Controlling Large Molecules at High Repetition Rates: Toward the "*Molecular Movie*"?

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Abstract: We have exploited electric fields to control the translations and rotations of complex molecules. This allows for the preparation of state-, size-, and structural-isomer selected samples of molecules fixed in space. These are ideal targets for the investigation of chemical reaction dynamics.

With our new experimental setup we aim for studying ultrafast "chemical" dynamics of large and complex molecules directly in the molecular frame. In first benchmark experiments we create supersonic, cold beams of prototypical iodobenzene (C_6H_5I) molecules at high repetition rates – up to 1 kHz. These molecular beams are quantum state selected by dc electric fields and, subsequently, laser aligned and mixed-field oriented by strong laser fields and weak dc electric fields. The resulting strongly aligned and oriented molecular samples are characterized by strong-field ionization using femtosecond laser pulses and velocity-map imaging of the produced ions to derive the angular distribution of the molecules. The degrees of alignment and orientation are characterized as a function of repetition rate, state selection, and laser parameters.

In the future, the high repetition rate production of these clean, well-defined samples will strongly benefit, or simply allow, novel time-resolved experiments on the dynamics of complex gas-phase molecules, for instance, femtosecond pump-probe measurements, X-ray or electron diffraction of molecular ensembles (including molecular-frame photoelectron angular distributions and diffraction-from-within experiments), or tomographic reconstructions of molecular orbitals. These samples could also be very advantageous for metrology applications, such as, for example, matter-wave interferometry or the search for electroweak interactions in chiral molecules. Moreover, they provide an extreme level of control for stereo-dynamically controlled reaction dynamics.

We have already exploited such state-selected and oriented samples to measure photoelectron angular distributions in the molecular frame (MFPADs) from non-resonant femtosecond-laser photoionization and using the Free-Electron-Lasers FLASH and LCLS. We have also investigated (coherent) X-ray-diffractive imaging and, also using ion-momentum imaging, the induced radiation damage of these samples due to the X-ray irradiation.