Controlled molecules for chemical dynamics studies

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Manipulating the motion of large, complex gas-phase molecules poses severe challenges and chances. It would allow novel approaches to investigate these systems with unprecedented detail. However, it requires disentangling the high density of states present in these heavy molecules, i.e., the manipulation of molecules in so called high-field-seeking states. Moreover, large molecules exhibit multiple structural isomers, even under the cold conditions of molecular beams.

We are developing methods to manipulate the translational and rotational motion of large molecules for the investigation of their molecular dynamics and chemical reactivities. This includes the deflection [1,2], focusing [3], and deceleration [4] of these molecules with electric fields, as well as the control of their rotational motion using electric-field orientation, laser alignment, and mixed field orientation [1,5]. We have demonstrated the spatial separation of individual quantum states, conformers (structural isomers) [3], and sizes of neutral clusters [6] and exploited the produced state-selected samples to produce one-dimensionally and three-dimensionally mixed-field-oriented molecular samples. We can now routinely perform these experiments at a kHz repetition rate [7].

We have exploited such state-selected and oriented samples to measure photoelectron angular distributions in the molecular frame (MFPADs) from non-resonant femtosecond-laser photoionization [8] and using the free-electron lasers FLASH and LCLS. We have investigated their (coherent) X-ray-diffractive imaging [9] and, also using ion-momentum imaging, the induced radiation damage of these samples due to the X-ray irradiation.

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, for instance, in studies of the reactivities of conformer selected molecular beams with trapped ions.

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