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Manipulating the motion of large molecules: Information from the molecular frame

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Large molecules have complex potential-energy surfaces with many local minima. They exhibit multiple stereoisomers, even at the low temperatures (~1 K) in a molecular beam, with rich intra- and intermolecular dynamics. Over the last years, we have developed methods to manipulate the motion of large, complex molecules and to select their quantum states. We have exploited this state-selectivity, for example, to spatially separate individual structural isomers of complex molecules [1] and to demonstrate unprecedented degrees of laser alignment and mixed-field orientation of these molecules [2]. Such clean, well-defined samples strongly benefit, or simply allow, novel experiments on the dynamics of complex molecules, for instance, femtosecond pump-probe measurements, X-ray or electron diffraction of molecular ensembles (including 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 recently exploited these state-selected and oriented samples to measure photoelectron angular distributions in the molecular frame (MFPADs) from non-resonant femtosecond-laser photoionization [3] and using the X-ray Free-Electron-Laser LCLS. We have also investigated X-ray diffraction imaging and, using ion momentum imaging, the induced radiation damage of these samples using the LCLS. This work was carried out within a collaboration for which J. Küpper, H. Chapman, and D. Rolles are spokespersons. The collaboration consists of CFEL (DESY, MPG, University Hamburg), Fritz-Haber-Institute Berlin, MPI Nuclear Physics Heidelberg, MPG Semi-conductor Lab, Aarhus University, FOM AMOLF Amsterdam, Lund University, MPI Medical Research Heidelberg, TU Berlin, Max Born Institute Berlin, and SLAC Menlo Park, CA, USA. The experiments were carried out using CAMP (designed and built by the MPG-ASG at CFEL) [4] at the LCLS (operated by Stanford University on behalf of the US DOE) [5].

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