Molecular dynamics of state-selected molecules fixed in space

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State-selected aligned and oriented molecular ensembles serve as ideal samples to study ultrafast molecular dynamics in the molecular frame. Possible probing mechanisms are the investigation of molecular-frame photoelectron angular distributions or the detection of structural changes via X-ray and electron diffraction.

We have developed techniques to manipulate the motion of molecules in cold supersonic beams using strong inhomogeneous electric and laser fields at 1 kHz repetition rate. State-, conformer-, and size-selection is achieved, for instance, using the electric deflector, the analogue of the Stern-Gerlach deflector and the electrostatic bender for charged particles. The state-selected molecules are aligned by strong laser fields or oriented in combination of laser fields and static electric fields. The laser pulses for alignment and orientation are provided by a kHz Ti:Sapphire laser system. The laser pulse length can be varied continuously between 50 fs and 500 ps. This allows for the manipulation of the rotational degrees of freedom of the molecules non-adiabatically (impulsively) as well as adiabatically, and the study of the intermediate regime.

Here, we will present our work on the alignment and orientation of iodobenzene and carbonyl sulphide (OCS). We will discuss our findings on the (non) adiabaticity of the alignment and orientation of OCS for various pulse lengths between impulsive and adiabatic alignment and orientation. Intruiging effects of state- and laser-intensity-specific alignment, orientation without alignment, and selective detection of molecular eigenstates with ultrashort laser pulses are observed.

These samples allow the recording of molecular-frame photoelectron angular distributions (MFPADs) to image the electronic and nuclear structure. In pump-probe type experiments one can thus follow the dynamics of these controlled systems.