

Controlled molecules for ultrafast chemical dynamics studies

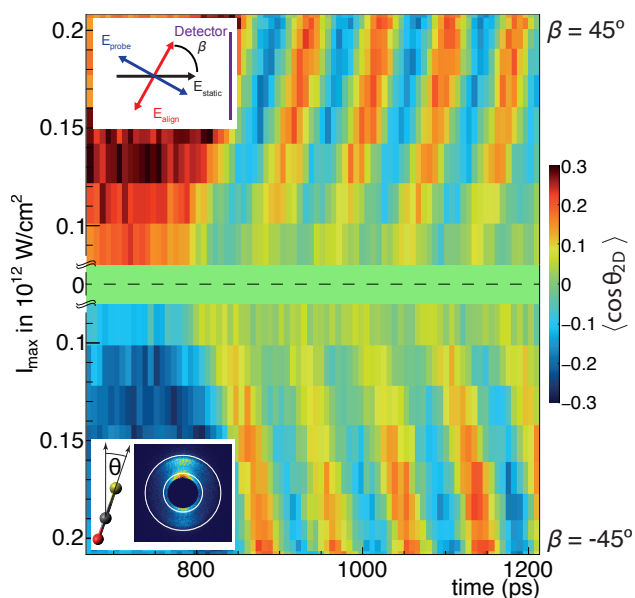
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A molecule's chemical behavior is governed by its electronic and nuclear properties. Imaging the temporal evolution of the valence electrons and the positions of the atoms during a chemical reaction provides direct insight into fundamental chemical processes. State-selected, strongly aligned, and oriented molecular ensembles allow for studies of ultrafast chemical dynamics directly in the molecular frame. Such dynamics can be extracted, for instance, *via* molecular-frame photoelectron angular distributions (MFPADs) and x-ray or electron diffraction.

We have developed techniques to manipulate the motion of molecules in cold supersonic beams using strong inhomogeneous electric and laser fields. The state-selected molecules are aligned or oriented by the combined effect of a dc electric field and moderately strong laser pulses with tunable duration between 50 fs and 500 ps. This allows for non-adiabatic as well as adiabatic alignment.

I will present our work on the alignment and orientation of carbonyl sulphide (OCS) and iodobenzene molecules: This includes, for example, the creation of coherent superpositions of pendular states in the strong field of the alignment laser, which resulted in pendular motion. Alternatively, strong laser-field-free orientation of ensembles of absolute-ground-state-selected OCS was achieved. Furthermore, MFPADs of indole and indole-water₁ clusters were used to topographically build up a 3D MFPAD. The results will be discussed using an approach based on the strong-field approximation.



Orientation dynamics of OCS