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Field-free Alignment of Asymmetric-Top Molecules

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Fixing molecules in space enables a variety of investigations into molecular structure and dynamics, such as high harmonic spectroscopy or ultrafast diffractive imaging of molecular dynamics. Molecules can be aligned and oriented to the laboratory frame using suitable ac and dc electric fields [1,2]. State-selection can lead to a significant enhancement of the degrees of alignment and orientation [2,3]. We follow both, experimental and theoretical, paths to investigate techniques to improve 1D and 3D alignment and orientation. Experiments have resulted in both, in-field as well as laser-field-free, alignment and orientation of the linear carbonyl sulfide (OCS) molecule [4,5,6]. Many of these techniques are also applicable to more complex molecules and clusters [2,3,7].
Here, we discuss how laser-pulse shaping was utilized to show field-free 3D alignment of indole experimentally. More complex shaping, combined with learning-loop approaches, is planned to further enhance the degree of alignment and orientation with the goal of near-perfect 3D alignment and orientation of complex asymmetric top molecules. Theoretical simulations of the rotational behavior of such systems interacting with electric fields will be used to guide experiments.

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