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Towards Control of 3D Alignment of Polyatomic Molecules

MAXIM ARTAMONOV, Northwestern University

3D alignment of polyatomic molecules is the subject of a growing number of studies fueled by a host of potential applications. In general, to fully control the rotations of an asymmetric molecule, it is necessary to define two nonequivalent directions in space by means of two non-collinear fields. To this end, several strategies have been proposed; however, the question of the best approach in achieving 3D alignment remains open. One method to control the rotations of an asymmetric top molecule in 3D space is by using a combination of long and short (relative to the molecular rotational period) linearly polarized pulses. The molecular axis with the largest polarizability is sharply aligned along the polarization vector of the long pulse while the short pulse, orthogonally polarized and applied at the peak of the long pulse, spins the molecule about the “held” axis. By rapidly truncating the long pulse following the short pulse turn-off this method can be adopted to furnish field-free 3D alignment, a particularly attractive regime in terms of possible applications. Another approach to induce field-free 3D alignment is by using two short laser pulses linearly polarized in orthogonal directions. The temporal parameters of the pulses are tailored to a given molecule through optimization in order to produce the best overall 3D alignment or enhance the alignment in a particular angle.