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Controlling the rotational motion of asymmetric top molecules by laser pulses HENRIK STAPELFELDT, Department of Chemistry, University of Aarhus

While the vast majority of previous studies on laser induced alignment of small molecules dealt with linear systems, interest is now shifting to asymmetric tops due to both the new physics involved and the broad range of applications. However, controlling the rotational motion of asymmetric tops represents, in general, a much harder task because they are characterized by three axes with different moments of inertia and different polarizability components. This talk will discuss recent studies on laser alignment of asymmetric tops. First, we show how the alignment dynamics induced by a single linearly polarized short laser pulse can be controlled by the fluence. When the fluence is increased the complex non-periodic revival structure of an asymmetric top approaches a simple periodic rotation around a single axis. Second, we introduce a new method for 3-dimensional (3D) alignment control by combining two linearly polarized pulses laser pulses, one short and one long compared to the molecular rotational periods. The long pulse strongly aligns the most polarizable molecular axis along its polarization axis while the orthogonally polarized short pulse sets the molecule in to controlled rotation about the axis aligned. As a result strong 3D alignment occurs immediately after the short pulse and is repeated periodically reflecting the revolution about the axis aligned. Our method opens new directions for field-free 3D alignment and for controlling internal rotations of molecules.