Molecular Alignment and Orientation: From Laser-Induced Mechanisms to Optimal Control

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Abstract. Laser-induced alignment and orientation of molecules are challenging control issues with a wide range of applications, extending from chemical reactivity to nanoscale design [1].

One of the basic mechanisms for alignment (i.e. molecular axis parallel to field polarization) is related with the pendular states accommodated by the molecule-plus-field effective potential. The laser control of alignment can be reached by an adiabatic transport of an initial isotropic rotational state on some pendular state trapping the molecule in well aligned geometries. After the laser pulse is turner off, alignment can no more be observed, unless some sudden excitations are referred to, leading to high rotational populations [2]. Symmetry breaking mechanisms are to be looked for when orientation (i.e. molecular axis in the same direction than field polarization) is the goal of the laser control. Two mechanisms are considered. The first is based upon an asymmetric pulse combining a frequency w and its second harmonic 2ω resonant with a vibrational transition. Due to the polarizability, both even and odd rotational states are excited, resulting into some orientation effect [3]. A much more efficient mechanism is the so-called "kick" that a highly asymmetric sudden laser pulse can impart to the molecule. Half-cycle pulses, within the reach of today's experimental technology, are among good candidates for producing such "kick" [4]. Laser-induced alignment and orientation of some molecules like HCN, LiCl, LiF, are analysed emphasizing the above-mentioned basic mechanisms. More interestingly, an optimal control scheme for orientation, worked with genetic algorithms, also leads to a sudden pulsed field bearing the characteristic features of the kick mechanism [5]. Optimal pulse shaping for very efficient and long lasting orientation, on one hand, robustness with respect to temperature effects, on the other hand, are among our future prospects.

References

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