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

Osman Atabek (osman.atabek@ppm.u-psud.fr)
Laboratoire de Photophysique Moléculaire du CNRS
Bâtiment 213
91405 Orsay Cedex
France

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
depolarization) is related with the pendular states accommodated by the molecule-
for 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 turned 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
depolarization) is the goal of the laser control. Two mechanisms are considered. The
first is based upon an asymmetric pulse combining a frequency $\omega$ and its second
harmonic $2\omega$ 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 algo-
rithms, 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
3. C. Dion, A. D. Bandrauk, O. Atabek, A. Keller, H. Umeda and Y. Fujimura,