OPTIMAL CONTROL OF MOLECULAR AND ELECTRON DYNAMICS

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By using coherent control techniques, it is possible to trace and control the behavior of quantum systems on their natural time scale. For molecular dynamics on the order of femtoseconds, this is achieved by applying ultrashort coherent light fields in the wavelength range from the IR to the UV. These laser pulses can be variably shaped in space and time using a laser pulse shaper consisting of some adaptive device such as a deformable mirror or a liquid-crystal display. [1]

Laser-optimized femtochemistry in the gas phase and liquid phase is one field in which this new technique is successfully employed. Automated optimization of branching ratios and total product yields of gas phase photodissociation reactions as well as chemically selective molecular excitation in the liquid phase is performed [2] [3].

A central question in chemistry and biophysics is how the structure and the dynamics of complex molecules evolve and are related to the primary processes. Structural changes of a molecule in the liquid phase have been induced by laser-optimized switching between the trans- and the cis-configuration in the photoisomerization of a cyanine dye molecule [4].

A new technological development further increases the possibilities and prospects of quantum control. With the technique of femtosecond polarization pulse shaping [5], it is now possible to vary intensity, momentary frequency, and light polarization (i.e., the degree of ellipticity as well as the orientation of the principal axes) as functions of time within a single femtosecond laser pulse. The time dependent variation of the polarization state of an ultrashort laser pulse has been used to optimize multiphoton ionization of dimer molecules K_2 from a cold molecular beam [6]. This experiment demonstrates a qualitative extension of quantum control mechanisms.

Adaptive high-harmonic XUV spectral shaping is performed by phase-only shaping of the fundamental (800 nm) driver laser pulse. A high degree of control is observed. Enhancement of single harmonics or group of harmonics as well as creation of spectral holes in the plateau region, i.e. suppression of particular harmonics, is achieved. Automated spectral shaping opens the door to XUV sub-femtosecond temporal pulse shaping [7].

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