Adaptive Femtosecond Quantum Control

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Obtaining active control over the dynamics of quantum-mechanical systems is a fascinating perspective in modern physics. The intrinsically broad spectral distribution and phase function of femtosecond laser pulses can be specifically manipulated by pulse shapers to drive molecular systems coherently into desired reaction pathways [1]. Following a suggestion of Judson and Rabitz [2], a computer-controlled pulse shaper is used in combination with a learning evolutionary algorithm and direct feedback from the experiment to realize a closed-loop setup for adaptive femtosecond laser pulse shaping [3]. Coherent control over quantum-mechanical processes is achieved in an automated fashion, without requiring any model for the system's response. This technique can be applied to actively control gas-phase photodissociation processes [4].

Recently, adaptive pulse-shaping techniques were transferred to the control of photoprocesses in the liquid phase [5]. Chemically selective molecular excitation is achieved by many-parameter adaptive quantum control, despite the failure of typical single-parameter approaches such as wavelength control, intensity control, or linear chirp control. Photoprocesses in two different molecular species can be controlled at the same time, even in the presence of complex solvent/solute interaction. Applications are envisioned in chemical analysis or in bimolecular reaction control where specific reactant molecules could selectively be "activated" for purposes of chemical synthesis.

A novel technological development further increases the possibilities and prospects of quantum control. Femtosecond polarization pulse shaping [6] allows to manipulate electromagnetic waves in a fundamentally new way. The polarization state of light—i.e., the ellipticity and the elliptical orientation—as well as the intensity and oscillation frequency can be varied as functions of time within a single femtosecond laser pulse. Thereby it is possible to gain access to three-dimensional properties and dynamics of quantum wavefunctions. First experiments illustrate the feasibility of feedback-controlled polarization shaping.

Due to the universality of closed-loop learning optimization of ultrashort laser pulses, a broad field of applications opens up in all areas where femtosecond laser technology is used.

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