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Quantum Control of Femtochemistry in the Gas Phase, Liquid Phase and on Surfaces

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By using coherent control techniques we control the behavior of quantum systems on their natural fs-time scale 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 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]. Structural changes of a molecule in the liquid phase have been controlled by laser-optimized photoisomerization of a cyanine dye molecule [4] and of retinal in bacteriorhodopsin [5]. So far, optimal control techniques have been restricted to gas phase and condensed phase optimization experiments. Recently we have demonstrated femtosecond laser-assisted catalytic reactions on a Pd(100) single crystal surface. By applying a closed-loop optimal control scheme, we manipulate these reactions and selectively optimize the ratio of different bond-forming reaction channels, in contrast to previous quantum control experiments aiming at bond-cleavage. The results represent a first step towards selective photocatalysis of molecules.

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