Analysis and Control of Ultrafast Chemical Processes in Molecules and Aggregates

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With the advent of ultrafast laser sources, the utilization of pump & probe spectroscopy in the fs-regime has allowed for detailed insights to elementary dynamical molecular processes such as vibrations, fragmentation, isomerization, internal vibrational relaxation (IVR), neutralization, ionization, charge reversal and even photo-association. These phenomena were first investigated in great detail on simple molecular systems in the controlled environment of the gas phase. However, these fundamental studies soon lead to the realization that the observed photo-induced processes could not only be monitored, but also guided along a desired path in real time by the applied light fields. As a consequence, the employed laser pulses rapidly evolved from simple pump&probe sequences to highly structured light fields, optimally shaped in phase, amplitude and polarization. Exciting features could be extracted from the optimized pulse shapes, promoting a fruitful convergence with theory. These achievements demand for new challenges and applications: examples are the control of binary reactions and photo-catalytic processes together with a significantly increased spectral range and sub fs-time resolution for addressing the respective electron dynamics, exploiting the nonlinear response regime and reaching out for the atmosphere.