Molecular Spectroscopy with phase-locked ultrashort pulses at multiple wavelengths

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Many precise investigations of microscopic interest and high selectivity call for phase-locked pulses at freely chosen centre wavelengths throughout the UV, visible, and NIR and with high intensities. Through precise engineering of multicolor light fields, these pulse trains will be synthesized. Possible applications are in CARS spectroscopy, in coherent control of chemical reactions and in 2D optical spectroscopy.

Particularly for dynamic processes in molecules that proceed via conical intersections of the relevant potential energy surfaces, pulses with a duration of around 10 fs will be needed. Generation of such demanding pulses seems feasible with state-of-the-art optical parametric amplification techniques, but they are still some way short of routine use. For characterization and proper interaction with molecular samples of interest new approaches will have to be developed. It will only be through these most advanced experiments in molecular dynamics that a reliable comparison can be made with modern ab-initio and quantum dynamics calculations.

The PhD project will focus on the development of a heterodyne detected two-color photon-echo experiment with UV excitation and visible probe. Both pulses should have spectral width on the order of 1000 cm^{-1} and their relative phase will have to be fixed. Part of the effort will be to test the different possibilities to realize this goal. With this pseudo-2D spectrometer it will be possible to directly observe excitonic and adiabatic couplings in prototype molecules. These couplings are typically observed through reaction rates corresponding to decay times of well less than 100 fs. The conventional pump-probe spectroscopy can, however, not reveal the underlying details. This should be possible with the envisioned method.

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