Selective multiphoton excitation of molecules on helium droplets by tailored laser pulses

Albrecht Lindinger

Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

Synopsis Pulse shaping is reported for selective multiphoton processes of molecules in solution and on superfluid helium droplets. Particularly phase and polarization tailored laser pulses are employed for multi-photon excited fluorescence of different dyes in a liquid and a good agreement between experiment and simulation is obtained. An improved fluorescence contrast between different dyes is achieved which is relevant for imaging applications. Moreover, the dynamics of alkali dimer ionization on helium droplets is controlled with shaped pulses by activating or inhibiting different ionization paths due to molecular wave function interference. This is a demonstration of coherent control for molecules on helium droplet surfaces.

Control of photo-induced molecular processes has recently gained large interest and several successful optimal control experiments have already been conducted in different research fields. In order to influence the molecular dynamics, laser pulse shapers were utilized which allow for simultaneous shaping of phase, amplitude, and polarization - hence all parameters of the light field [1]. Tailored laser pulses particularly enable multiphoton excitation processes for selective control of molecular systems.

In this contribution, new pulse shaping methods for coherent control of multiphoton processes are demonstrated. Laser pulse shaping on multiphoton excitations enables to exploit intrapulse interference effects. Here, tailored laser pulses are utilized for selective photo-induced fluorescence of dye mixtures. Special antisymmetric phase functions are applied for scans of the excitations [2], and improved fluorescence contrasts are reported for two and three photon transitions, which is relevant for microscopic imaging. Moreover, polarization shaping is employed to increase the contrast between different dyes, whereby one dye is selectively excited in one polarization direction and the other dye is simultaneously excited in the perpendicular polarization direction. The presented pulse shaping methods will be favorable regarding novel biophotonic applications in endoscopy and microscopy.

As a demonstration of coherent control on doped helium droplets pump-probe scans of the ionization of potassium dimers on helium droplets were conducted by using a pulse shaper [3], which enables to precisely tune the phase difference between the sub pulses. A significantly modified ion signal was observed by solely varying the phase difference between the two sub pulses from zero to π (see figure 1). This feature is interpreted by different ionization pathways via the first and second excited state [4]. When pump and probe have the same phase, the wave functions interfere constructively and two pathways are observed, whereas the pathway via the first excited $A^1\Sigma_u^+$ state is annihilated when there is a π shift between both pulses, which generates destructive interference. Hence, a phase shift between different sub pulses results in a remarkably different ionization process. The explanation based on wave function interference assumes that the molecular wave packet inherits the phase of the laser pulse and that coherence is maintained. The results are regarded as evidence for interference control of molecules on helium droplets. Finally, prospects for controlling the photoassociation of prebiotic molecules in helium droplets are given concerning possible routes to the origin of life.

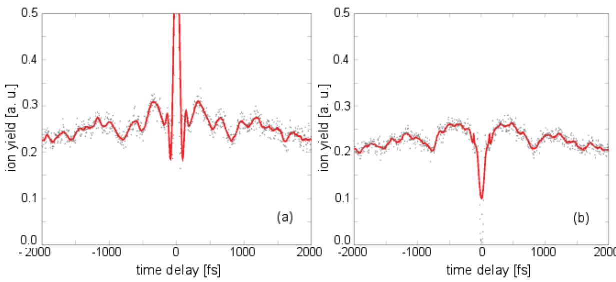

Figure 1. Pump–probe scans of the ionization of potassium dimers on helium droplets. The phase difference between the two pulses amounts to zero (a) and π (b). A significant change is observed by solely modifying the phase difference [3].

References

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E-mail: lindin@physik.fu-berlin.de