Towards Supercontinuum Spectroscopy and Control of Ultrafast Molecular Processes

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Elementary to the endeavor of manipulating the state of a molecular systems through the interaction with light is the capability to address the time scales as well as resonance conditions involved in driving a particular molecular process [1-5]. In view of this, the precision with which a molecular system can be controlled in its interference to a desired final state is largely determined by the bandwidth of the field driving the manipulation. The possibility of employing the octave-spanning radiation obtained from the filamentation of standard amplified femtosecond pulses in gaseous media is explored in this context [6]. The generation of white-light pulses and their compression to sub 7 femtosecond pulses in a single filamentation process in air is presented. The control over the full spectral bandwidth via a liquid crystal based modulation technique is demonstrated by the transcription of parametric phase functions in the construction of a desired pulse form. The spectroscopic application of this capability is further exemplified by an optimization of the spectral phase over the white-light envelope in a genetic algorithm controlled feedback loop. This was applied to driving the sequential electron photo-detachment and multi-photon ionization processes within the $Ag_3^{-/0/+}$ system under the conditions of space-charge limited densities in a radio frequency ion trap. For this case, pulses composed of ultrashort substructures with a variable time sequence of different spectral components and a total duration spanning into the picosecond domain are obtained from the optimization. Accompanying this, the versatility of analyzing the synthesized pulse forms via a transient grating based four-wave mixing FROG is shown, with the capability to indiscriminately characterize octave spanning pulses with substructures ranging the lower femto- to picosecond domain in intuitive traces. The technique described above presents a general instrumentation for precision control of ultrafast molecular processes.

[1] R.S. Judson, H. Rabitz, "Teaching lasers to control molecules" *Phys. Rev. Lett.* **68**, 1500 (1992).

[2] R. Kosloff, S. A. Rice, P. Gaspard, S. Tersigni, D. J. Tannor, "Wavepacket Dancing: Achieving Chemical Selectivity by Shaping Light Pulses" *Chem. Phys.* **139**, 201 (1989).

[3] P. Brumer, M. Shapiro, "Laser Control of Molecular Processes" *Annu. Rev. Phys. Chem.* **43** 257 (1992).

[4] T. Baumert, J. Helbing, G. Gerber, L. Wöste, A. H. Zewail, J. Troe, J. Manz, T. Kobayashi, V. S. Letokhov, U. "Chemical Reactions and Their Control on the Femtosecond Time Scale" *Solvay Conference on Chemistry*, **vol. 101**, John Wiley and Sons; NewYork (1997).

[5] *Analysis and Control of Ultrafast Photoinduced Reactions*, O. Kühn, L. Wöste (Eds.) Springer Series in Chemical Physics, **vol. 87**, Springer; Berlin (2007).

[6] B. E. Schmidt, W. Unrau, A. Mirabal, S. Li, M. Krenz, L. Wöste, T. Siebert, "Poor man's source for sub 7 fs: a simple route to ultrashort laser pulses and their full characterization" *Opt. Express* **16**, 18910 (2008).