

# Automated Coherent Control of Chemical Reactions and Pulse Compression by an Evolutionary Algorithm with Feedback

A. Assion<sup>1</sup>, T. Baumert<sup>2</sup>, M. Bergt<sup>1</sup>, T. Brixner<sup>1</sup>, B. Kiefer<sup>1</sup>, V. Seyfried<sup>1</sup>, M. Strehle<sup>1</sup>, and G. Gerber<sup>1</sup>

<sup>1</sup> Physikal. Institut, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

<sup>2</sup> Deutsches Zentrum für Luft- und Raumfahrt, D-82230 Weßling, Germany

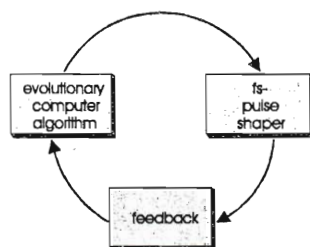
E-mail: gerber@physik.uni-wuerzburg.de

**Abstract.** We experimentally demonstrate automated coherent control of a photodissociation reaction as well as femtosecond pulse compression using a pulse shaper and an evolutionary algorithm with feedback.

## 1. Introduction

The electric field of femtosecond laser pulses can be shaped nearly arbitrarily by modifying the relative phase of the spectral components. We combine a programmable pulse shaper and an evolutionary algorithm with feedback in order to automatically form femtosecond laser pulses. The phase function is improved iteratively by directly including a specific experimental output in an optimization procedure which enables totally automated search for the best adapted laser pulse shape [1]. An evolutionary computer algorithm addresses the programmable femtosecond pulse shaper, uses the desired experimental output as feedback, and iteratively improves the applied laser field (see Fig. 1). This opens up technological as well as molecular physical applications. We use the method to compress complex phase-shaped laser pulses from a chirped pulse amplification (CPA) Ti:Sapphire femtosecond laser system, and to optimize different reaction channels of the photodissociation processes of  $\text{Fe}(\text{CO})_5$ .

The pulse shaper is based on the design of Weiner et al. [2]. It is set up as a zero dispersion compressor with a liquid crystal spatial light modulator (SLM) in its Fourier plane. With this device different optical path lengths can be introduced to the spatially separated spectral components of the laser pulse,



**Fig. 1.** Femtosecond laser pulses are modified by a computer-controlled pulse shaper. An evolutionary computer algorithm addresses the programmable femtosecond pulse shaper, uses the desired experimental output (signal of a 2-photon photodiode or the amount of ionic fragments recorded by a time-of-flight mass spectrometer) as feedback, and iteratively improves the applied laser field.

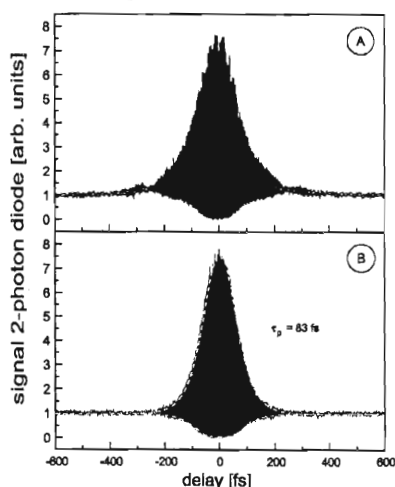
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resulting in a shift of the relative phases. A detailed description of the experimental setup and the algorithm is given in Ref. [3].

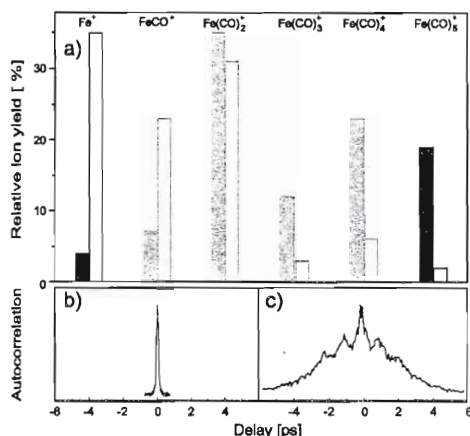
## 2. Results and Discussion

First we demonstrate the applicability of this method on the automated compression of modulated femtosecond laser pulses from a CPA Ti:Sapphire laser system. The CPA technique implies a lot of optical components which introduce phase distortions. Since the amplified laser pulses are usually required to be bandwidth-limited, careful alignment of the laser system is necessary. Higher order dispersion terms can be compensated for only with extensive experimental effort, if at all. To optimize the output of our CPA system we employ a 2-photon photodiode which is irradiated upon by the modified laser pulses. The shorter the laser pulses, the higher will be the signal of the nonlinear photodiode. The evolutionary algorithm improves iteratively the 2-photon signal and thus the phase function until an optimum is found. Interferometric autocorrelations of the unshaped input pulse and the optimized output pulse are shown in Fig. 2. The wings in the autocorrelation (Fig. 2A) could not be removed by a simple compressor alignment, but the algorithm yields a bandwidth-limited pulse (Fig. 2B) without any prior knowledge of the input phase function. Therefore pulses from a misaligned laser system can be improved automatically, without the user having to worry about complex phase functions.

Another application of the feedback-controlled femtosecond pulse shaper is found in femtochemistry. Microscopic control of chemical reactions on a molecular level is an old dream in chemistry. We experimentally demonstrate how automated coherent control of the photodissociation of  $\text{Fe}(\text{CO})_5$  can be achieved. Interaction of 800 nm photons with a  $\text{Fe}(\text{CO})_5$  molecular beam leads to different multi-photon processes. One possible process is direct ionization of the parent molecule, another possibility is a combination of ionization and fragmentation processes in which the  $\text{Fe}(\text{CO})_5$  molecules loses an electron and also from one up to all five of its carbonyl ligands. The reaction products are



**Fig. 2.** Interferometric auto-correlations. The input pulse (A) clearly exhibits wings which cannot be removed by simple compressor alignment. They indicate a complex phase function which is automatically compensated for by the evolutionary algorithm, resulting in a bandwidth-limited pulse (B).



**Fig. 3** a) The ratio of  $\text{Fe}(\text{CO})_5^+/\text{Fe}^+$  product yields is maximized (filled blocks), and minimized (empty blocks) by the optimization algorithm, yielding significantly different abundances of  $\text{Fe}^+$  and  $\text{Fe}(\text{CO})_5^+$ . The autocorrelations of the corresponding laser pulses are also included. A nearly bandwidth-limited pulse (b) leads to the production of mainly  $\text{Fe}(\text{CO})_5^+$  and a long laser pulse (c) to the production of mainly  $\text{Fe}^+$ .

detected in a time-of-flight (TOF) mass spectrometer and relative ion yields of the products are used as feedback. We maximized and minimized the branching ratio  $\text{Fe}(\text{CO})_5^+/\text{Fe}^+$ , yielding significantly different abundances of reaction products, as illustrated in Fig. 3. The maximization procedure yields the product distribution of Fig. 3a (solid blocks). The result of this optimization is a very short (bandwidth-limited) laser pulse (Fig. 3b). If, on the other hand, we choose to minimize the  $\text{Fe}(\text{CO})_5^+/\text{Fe}^+$  ratio, that is, to maximize the inverse, an optimum is found as well (Fig. 3a, open blocks). This optimization leads to a long laser pulse of ps duration (Fig. 3c). The optimized branching ratios could be changed by a factor of 70. The results are in complete agreement with previous experiments of pure pulse length variation [4].

### 3. Conclusion

The results demonstrate totally automated coherent control of photodissociation reactions as well as automated pulse compression. For this purpose we employ a femtosecond pulse shaper which is feedback-controlled by an evolutionary computer algorithm. It should be noted that the applied method needs no calibration and no information about the system for a successful optimization.

### References

- 1 R. S. Judson, H. Rabitz, *Phys. Rev. Lett.* **68**, 1500 (1992).
- 2 A. M. Weiner, D. E. Leaird, J. S. Patel, J. R. Wullert, *IEEE J. Quantum Electron.* **28**, 908 (1992).
- 3 T. Baumert, T. Brixner, V. Seyfried, M. Strehle, G. Gerber, *Appl. Phys. B* **65**, 779 (1997).
- 4 L. Bañares, T. Baumert, M. Bergt, B. Kiefer und G. Gerber, *J. Chem. Phys.* **108**, 5799 (1998).