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ADRIATICO RESEARCH CONFERENCE on

LASERS IN SURFACE SCIENCE

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Coherent Control - a brief overview
Optimization of Femtosecond Pulse Shaping using Feedback

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Coherent Control of Quantum Dynamics by Feedback-Optimized Femtosecond Laser Pulses

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Coherent Control - a brief overview
Optimization of Femtosecond Pulse Shaping using Feedback

- Chemical Reaction Dynamics controlled by Phase-Shaped fs-Laser Pulses
  - $Fe(CO)_5$
  - $CpFe(CO)_2Cl$

coworkers: A. Assion, V. Seyfried, M. Bergt, B. Kiefer, T. Brixner, T. Baumert, M. Strehle

support: DFG: SFB 347
SPP "Femtosecond Spectroscopy"
Fonds der Chemischen Industrie

time scale

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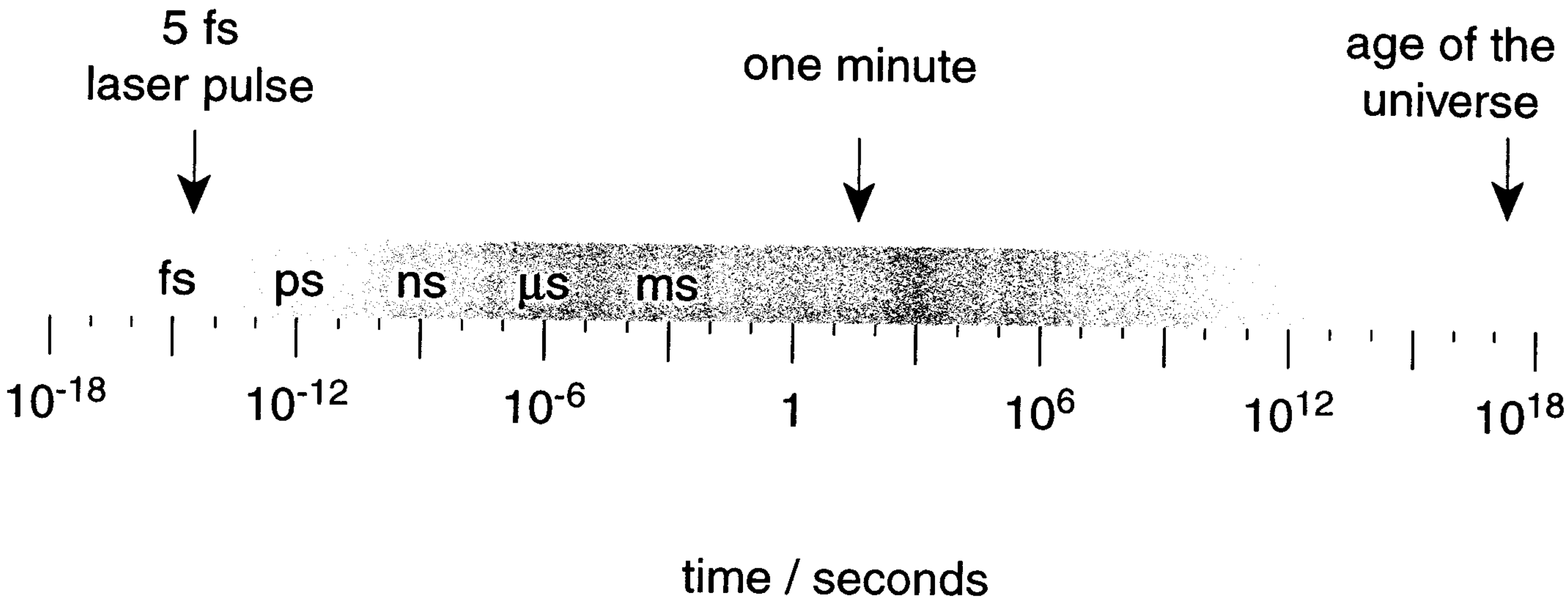


Femtosecond Time Scale

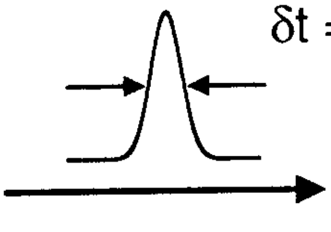
University of Würzburg, Department of Physics

1 fs = 10^{-15} sec   light travels: 0.3 μm

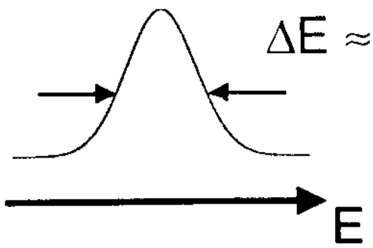
100 fs = 30 μm

---

pulse duration



energy distribution



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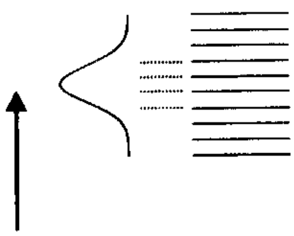
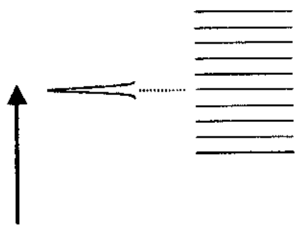
fs laser pulse



coherent superposition
$\Rightarrow$ wave packet

ns laser pulse



single stationary state

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atomic collisions (Ar^+-Ar; 10 - 10^3eV):	10 fs - 100 fs
molecular vibrational period:	100 fs - 1 ps
fastest molecular (dye) relaxation:	50 fs - 300 fs
interaction times in liquids:	10 fs - 1 ps
e^- - hole relaxation in semiconductors	50 fs - 200 fs

chemical reaction

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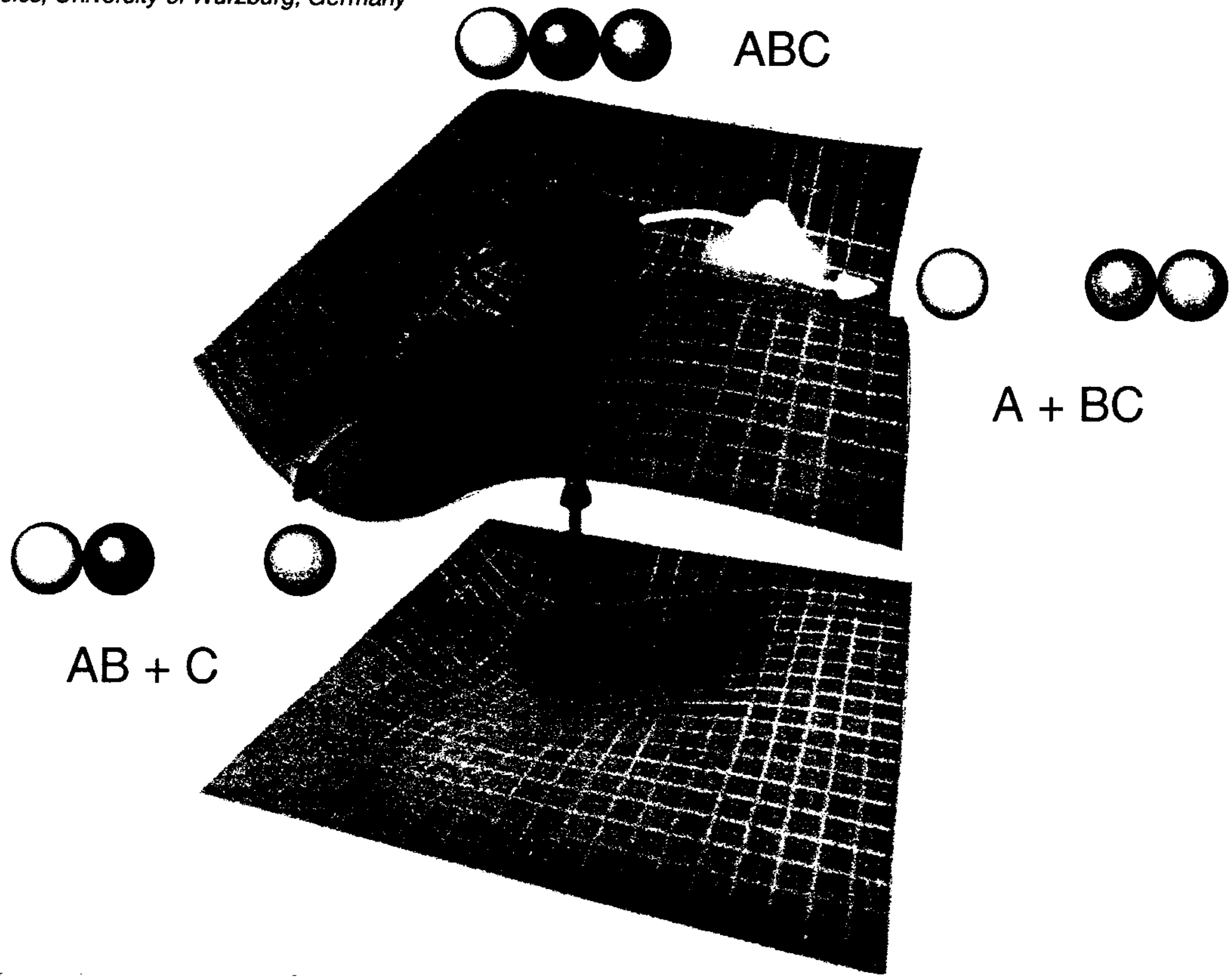

Intramolecular Energy Redistribution

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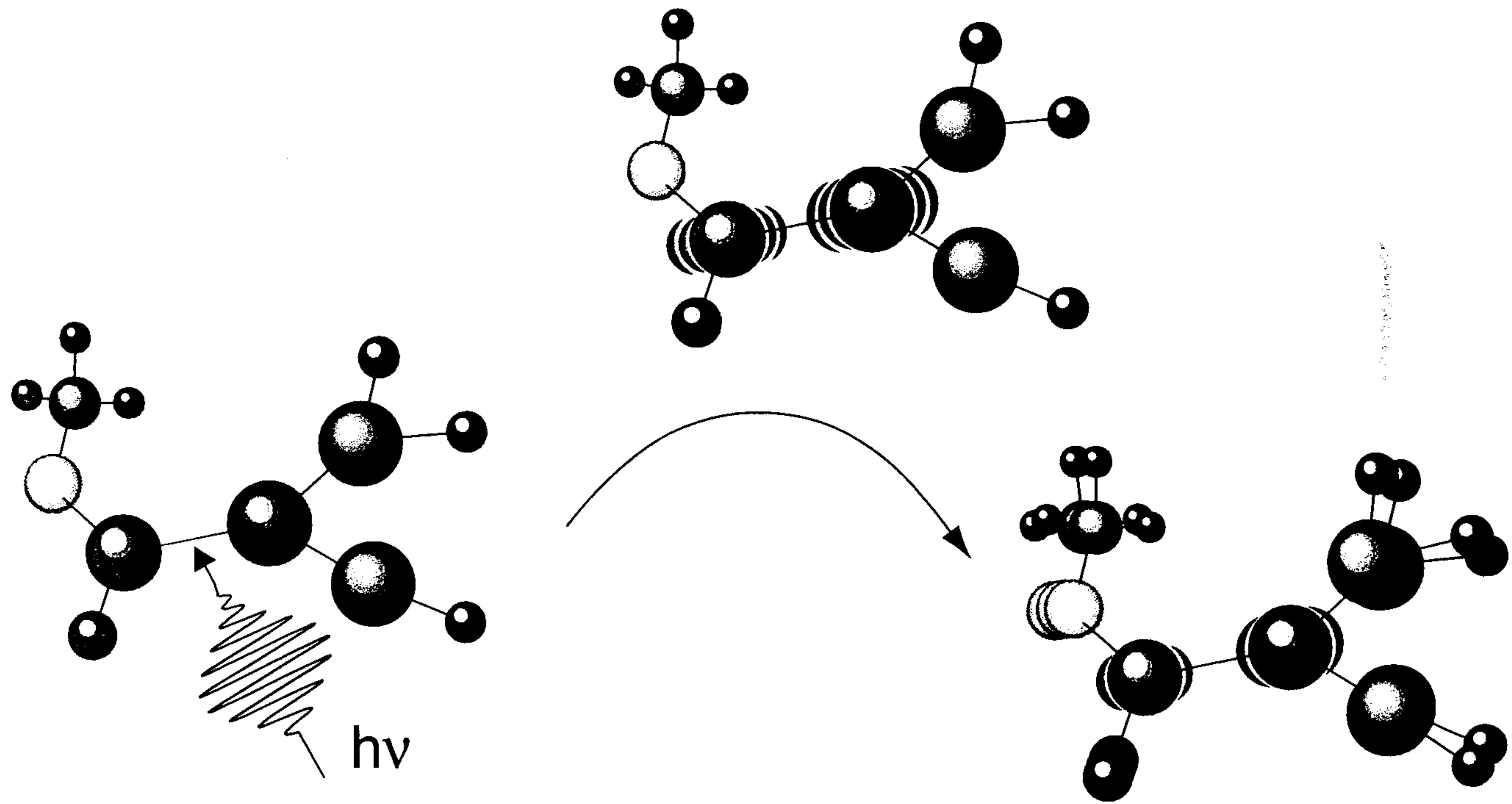

control of chemical reactions

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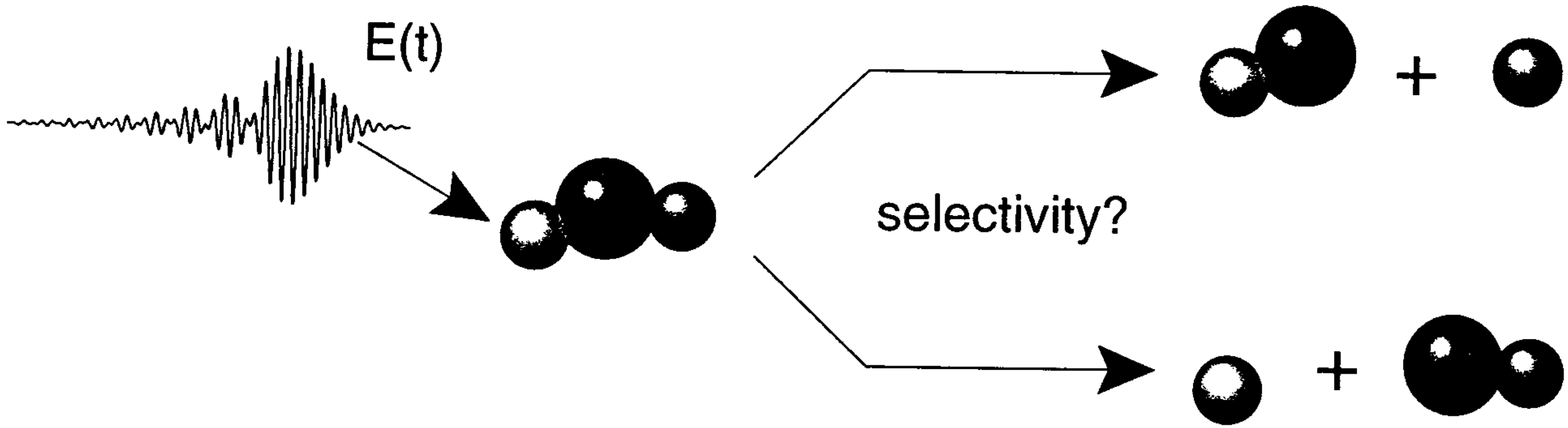


laser electric field E(t):

- calculation for real molecules extremely complicated (if not impossible)
- exact laboratory realization of predicted fields difficult

Quantum Control of Dynamics

Department of Physics, University of Würzburg, Germany

Brumer - Shapiro

CPL 126 , 54 (1986)

phase difference:

$\phi = \phi_{\omega} - \phi_{3\omega}$

Tannor - Kosloff - Rice

JCP 85 , 5805 (1986)

time delay Δt
between pump and probe

Rabitz

PRL 68 , 1500 (1992)

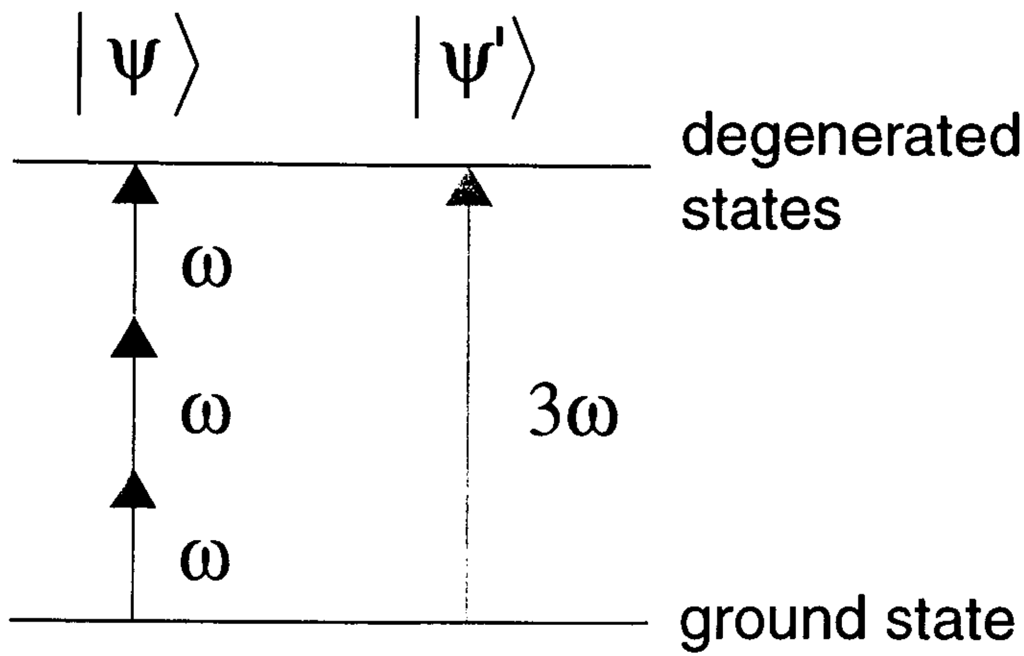
adaptive feedback control
of molecular motion

Brumer - Shapiro

Department of Physics, University of Würzburg, Germany

P. Brumer and M. Shapiro, Ann. Rev. Phys. Chem. 43, (1982)

- selectivity of product formation by tuning the phase difference between two laser sources at ω and 3ω
- exploiting the coherence properties of laser radiation in the frequency domain.



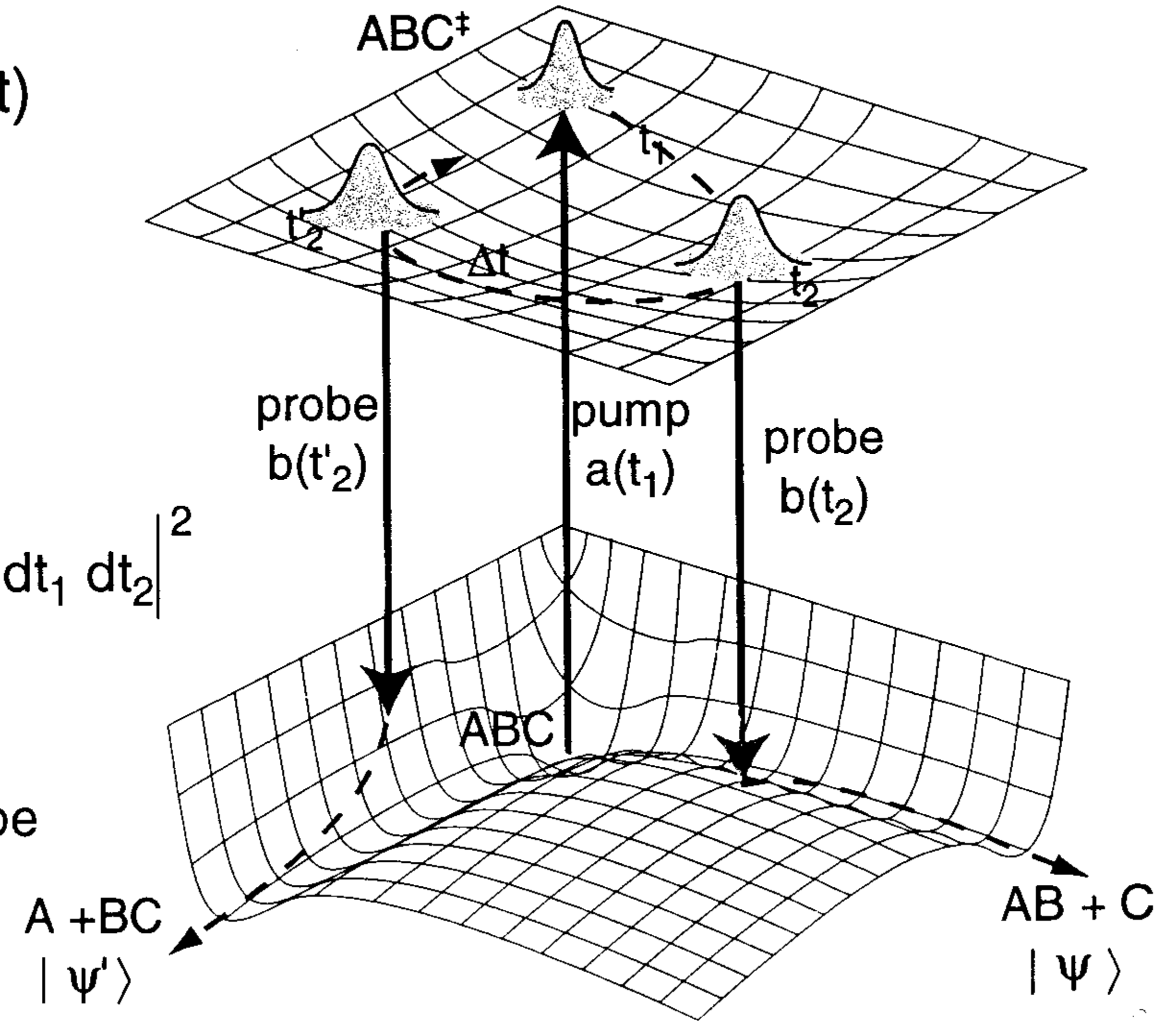
- two or more coherently locked laser fields
- scheme is analogous to the double slit experiment

Tannor - Kosloff - Rice

Department of Physics, University of Würzburg, Germany

D. J. Tannor, R. Kosloff, S. A. Rice, JCP 85, 5805 (1986)

Optimize pump a(t) and probe b(t) for exit in desired channel.



probability for 2-photon process with wavepacket propagation:

$$P_{if} = \left| \iint \langle \psi_f | \; b(t_2)\, e^{-iH(t_2-t_1)/\hbar}\, a(t_1) \,|\, \psi_i \rangle \; dt_1 \, dt_2 \right|^2$$

control parameters: delay $\Delta t = t_2 - t_1$, amplitude and chirp of pump and probe

molecular wavepacket dynamics and femtosecond time-resolved spectroscopy

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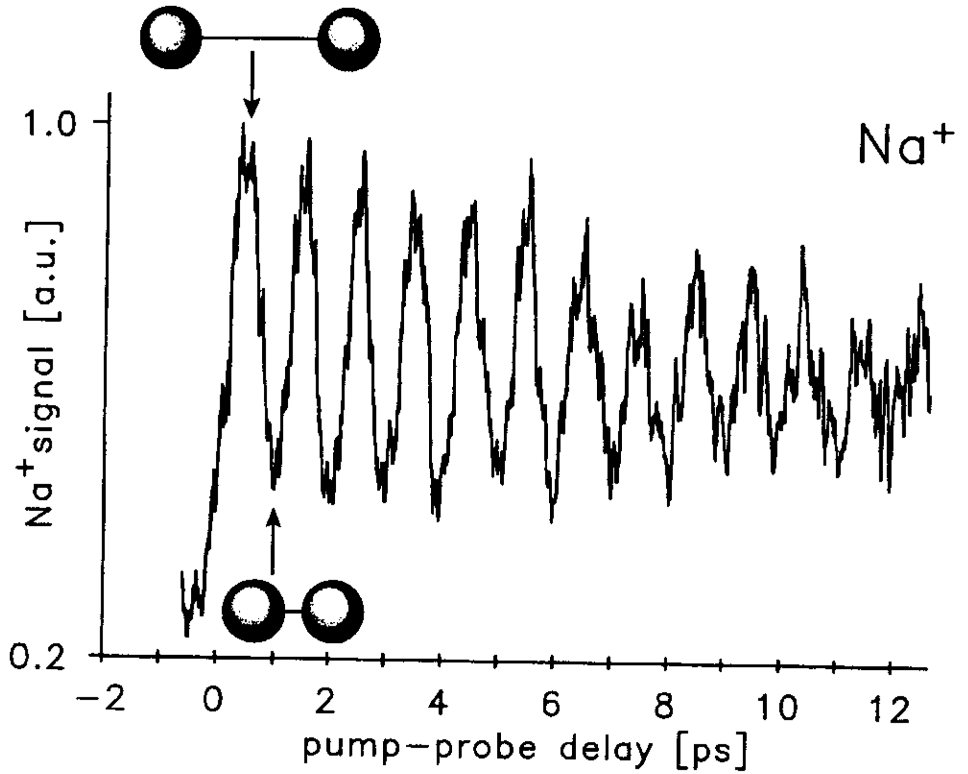
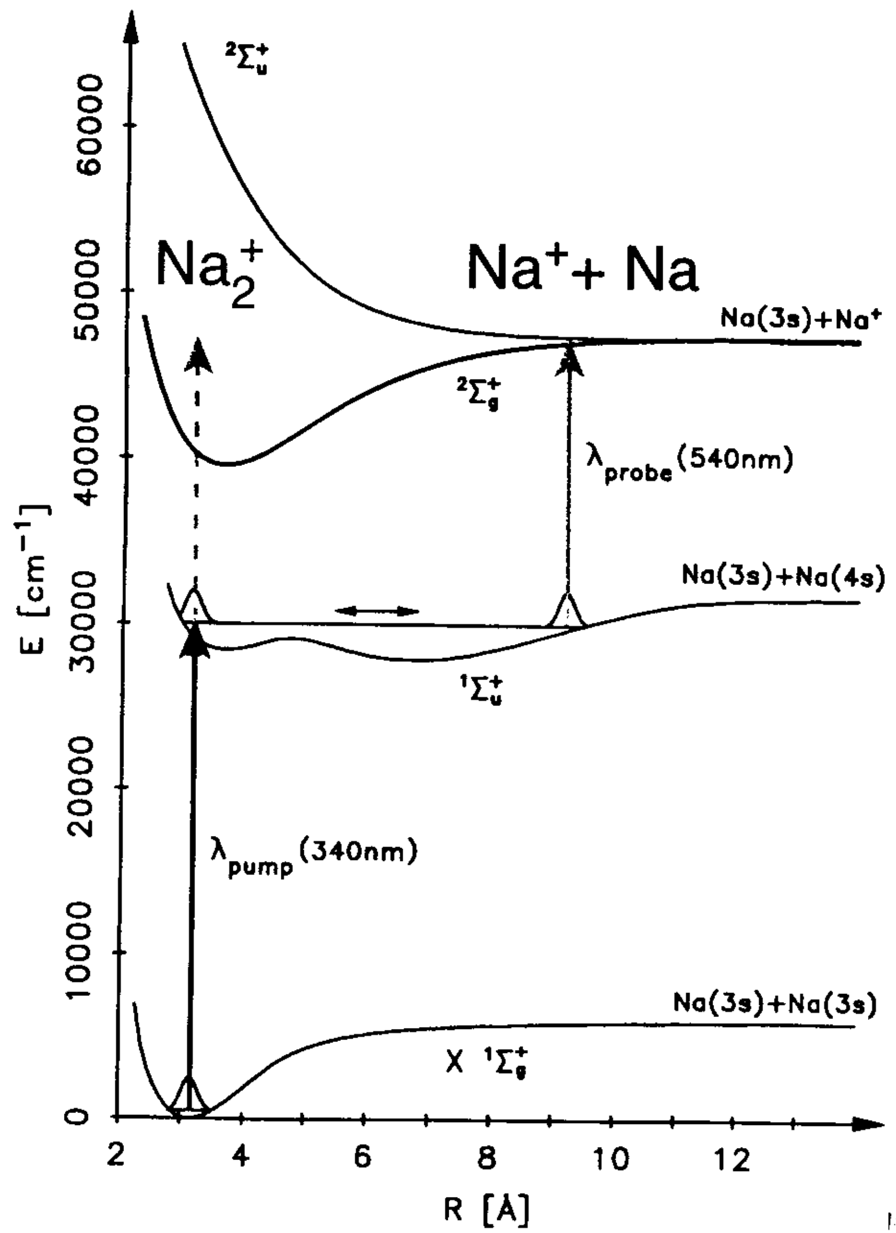




Z. Phys. D 36, 265-271 (1996)

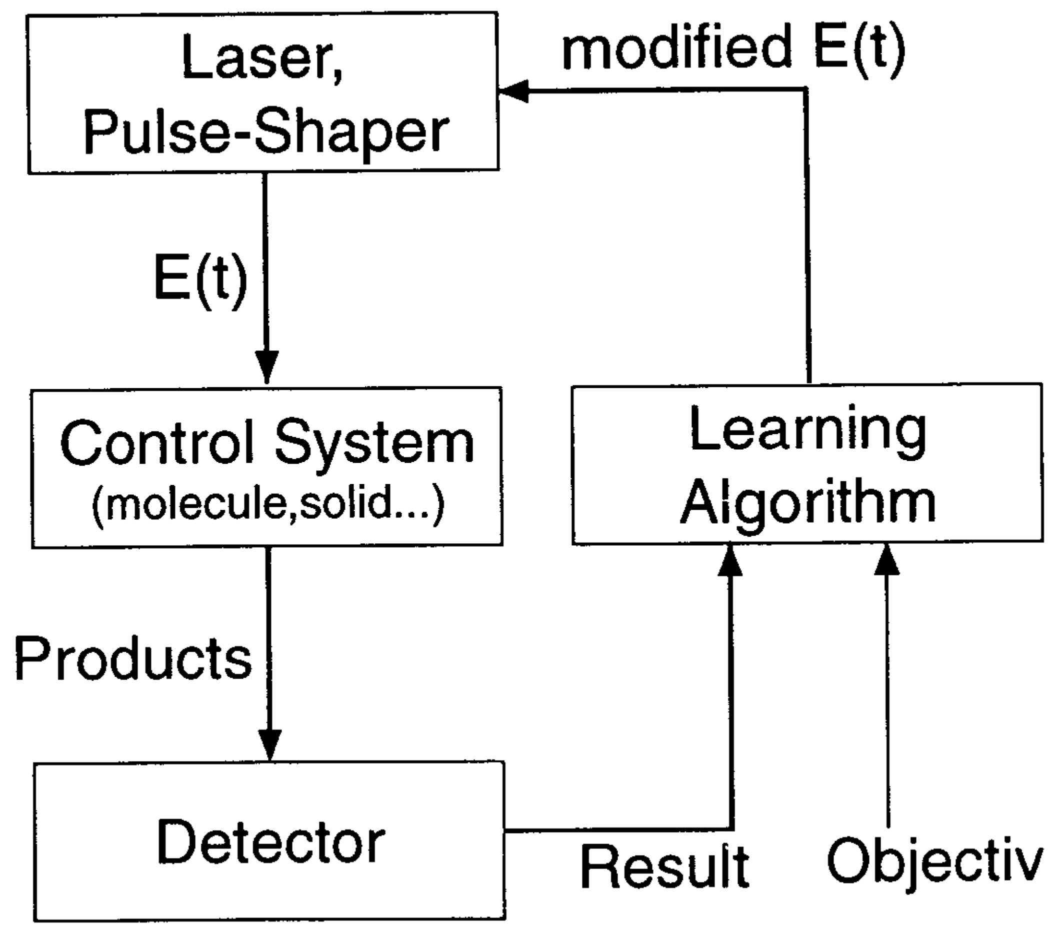
Rabitz

Department of Physics, University of Würzburg, Germany

R.S. Judson, H. Rabitz, PRL 68, 1500 (1992)

"optimal control therory" used to design electric fields such that the yield of a specified reaction product is maximized by directly including the experimental output

"teaching lasers
to control molecules"



adaptive optimization loop

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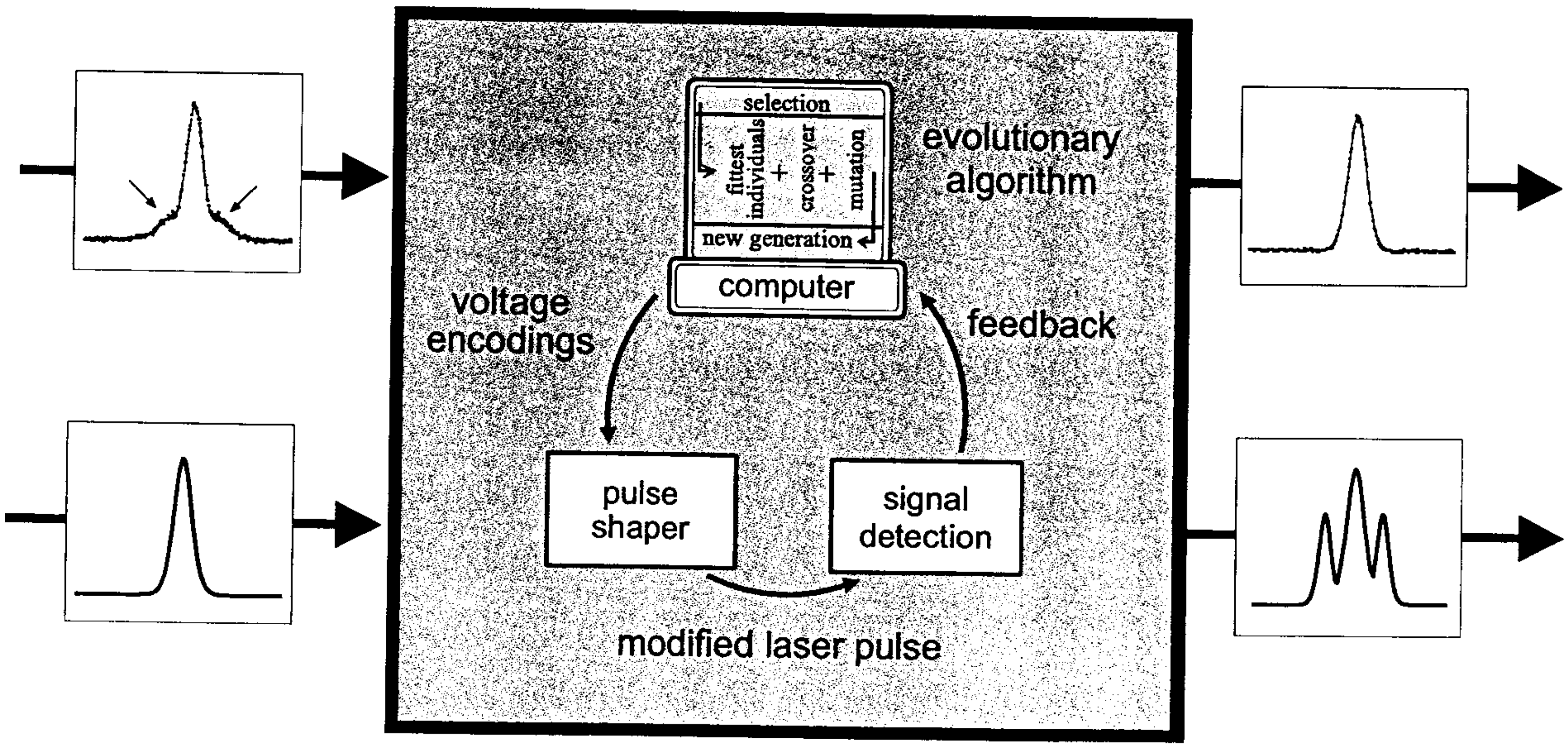


Appl. Phys. B **65**, *779 (1997)*

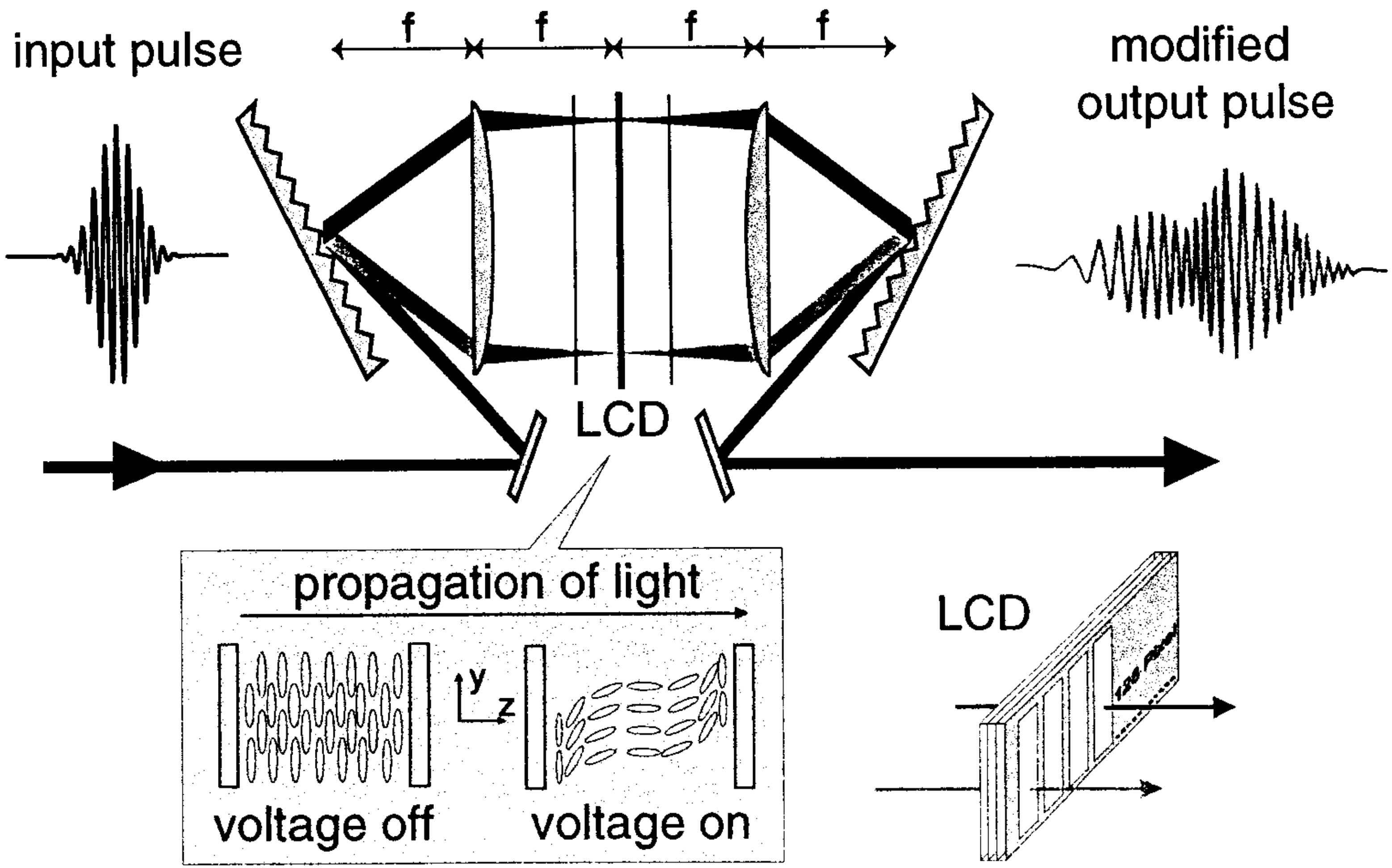
feedback: R.S. Judson, H. Rabitz: *Phys. Rev. Lett.* **68**, *1500 (1992)*

D. Yelin et al Opt. Lett. 22, 1793 (97)

C. J. Bardeen et al. CPL 280, 151 (97)

pulse shaper

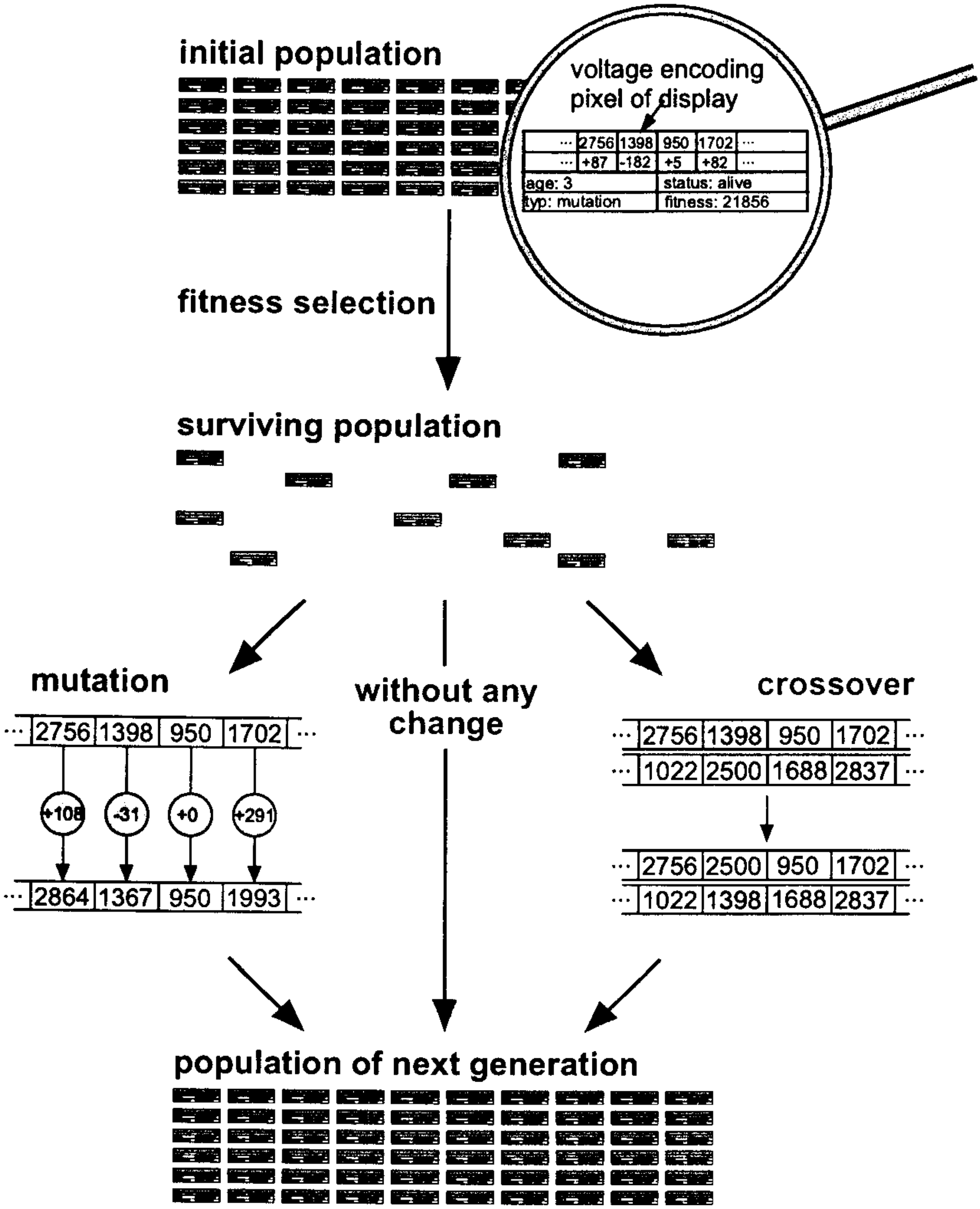
Department of Physics, University of Würzburg, Germany



A. M. Weiner et al.: Opt. Lett. ***15****, 326 (1990)*
IEEE -QE ***28*** *,908 (1992)*

Evolutionary Algorithm

Department of Physics, University of Würzburg, Germany



Appl. Phys. B 65, 779 782 (1997)

Applied Physics B Lasers and Optics

C Springer Verlag 1997

Rapid communication

Femtosecond pulse shaping by an evolutionary algorithm with feedback

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Received: 20 September 1997

Abstract. We report on computer controlled compression of femtosecond laser pulses using a programmable liquid crystal spatial light modulator which is feedback controlled by an evolutionary algorithm. This algorithm generates the optimal laser field on the basis of feedback from the experiment by optimizing the laser pulse iteratively. Without knowledge of the (chirped) input pulses, the experimental signal (second harmonic light=SHG) is maximized by the algorithm, thus resulting in fully compressed pulses. This method only makes use of the experiment's response (SHG signal) on the formed pulses. No other parameters need to be considered. This approach leads to many experimental applications in all fields of optics and ultrafast spectroscopy where particularly shaped pulses are advantageous.

PACS: 42.65.Re; 02.10.Jf; 07.05.Dz

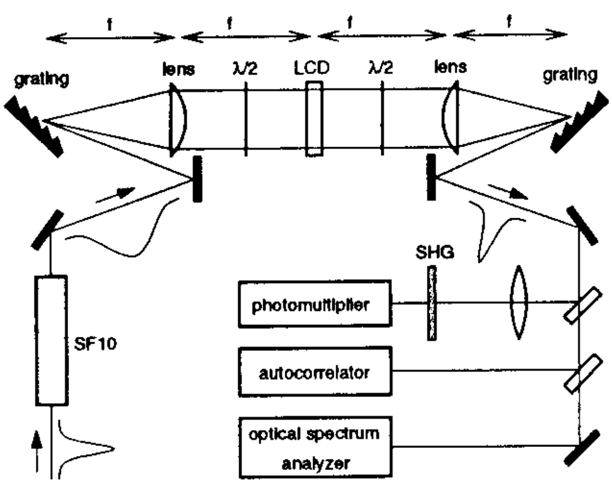

Fig. 1. Schematic experimental setup. A femtosecond laser pulse is tempo rally broadened by a SF10 rod, and recompressed by a pulse shaper

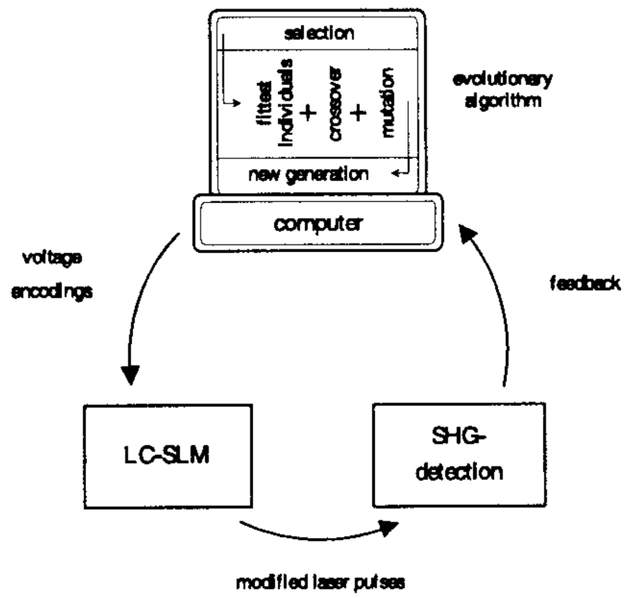

Fig. 2. Feedback loop. Given voltage encodings generate speci c second harmonic signals. The best patterns are selected and allowed to produce their offspring by mutation and crossover. The new generation is then test ed, and so forth

Recompression of fs-Laser Pulses Broadened by Material Dispersion

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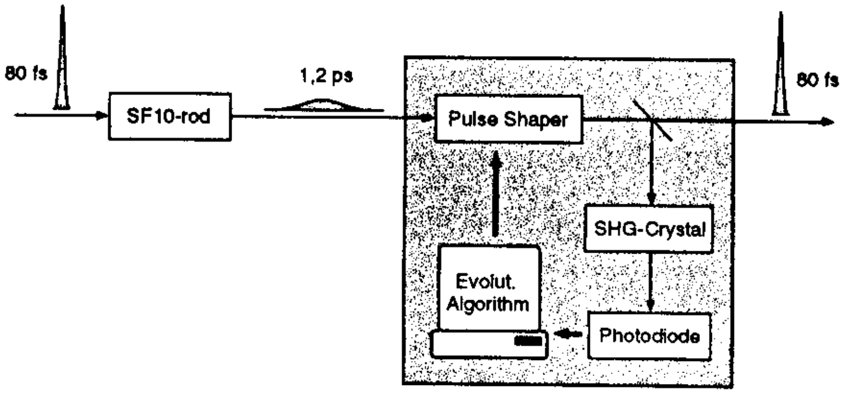
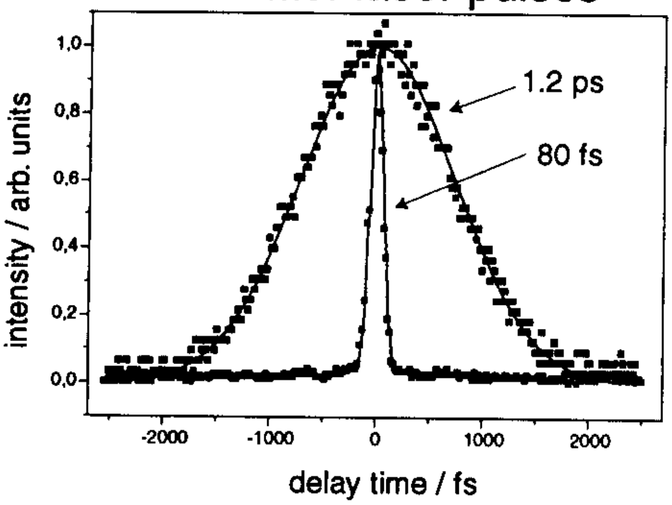
Experimental setup

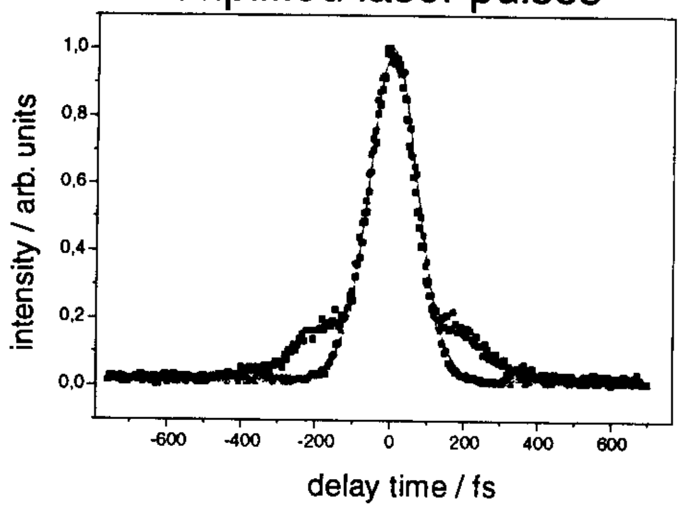


Recompression of broadened fs-laser pulses

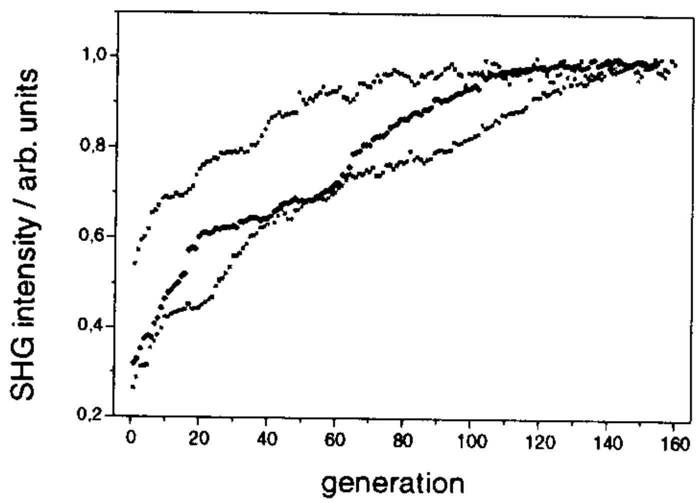
oscillator laser pulses



amplified laser pulses



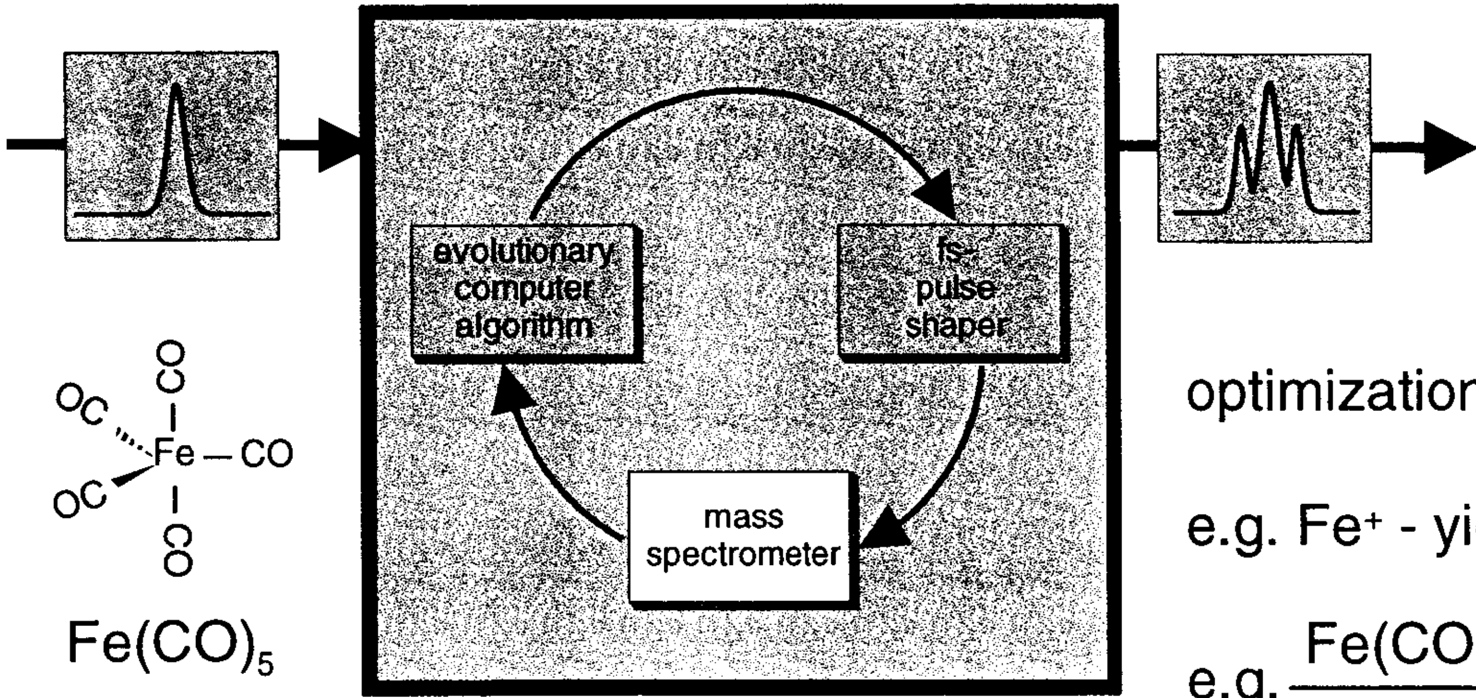
Evolution of the feedback signal



Appl. Phys. B **65**, 779 (1997) ; Appl. Phys. B **68**, 281 (1999)

Experimental Setup

Physics Department, University of Würzburg, Germany



$Fe(CO)_5$

optimization of

e.g. Fe^+ - yield

e.g. $\frac{Fe(CO)_5^+}{Fe^+}$ - ratio

result of optimization

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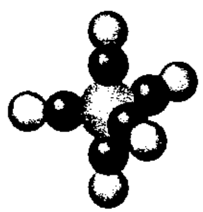
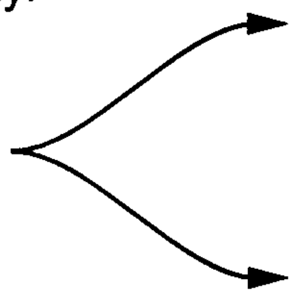
educt

ironpentacarbonyl



$Fe(CO)_5$



products

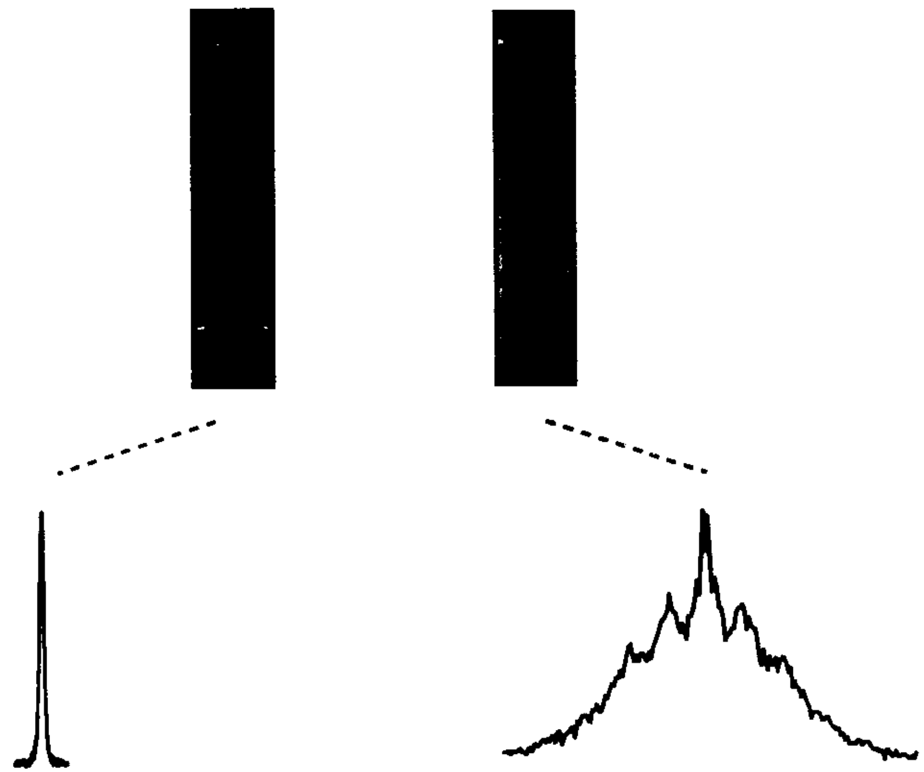
$Fe(CO)_5^+$

Fe^+

product yields

4,9 : 1

0,07 : 1



autocorrelation of optimized laser pulse (maximal ratio)

autocorrelation of optimized laser pulse (minimal ratio)

control of chemical reactions

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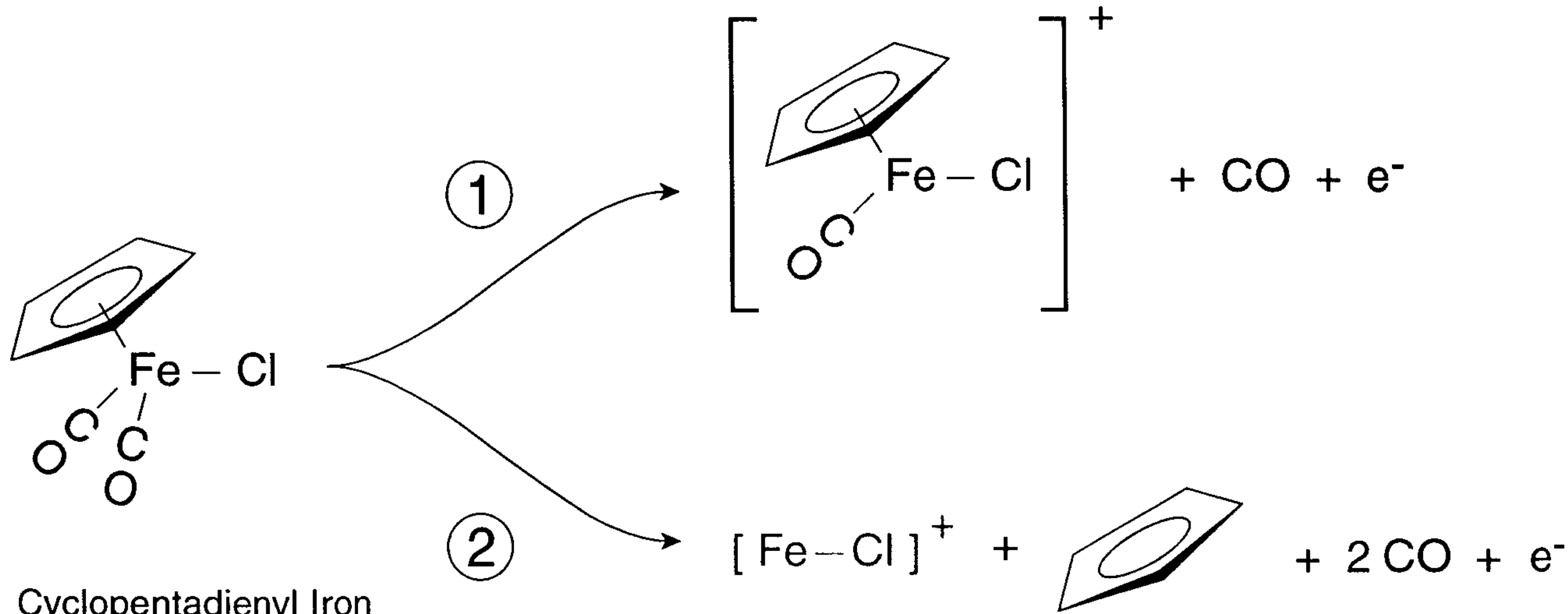

Cyclopentadienyl Iron
Dicarbonyl Chlorid

feedback controlled pulse shaping

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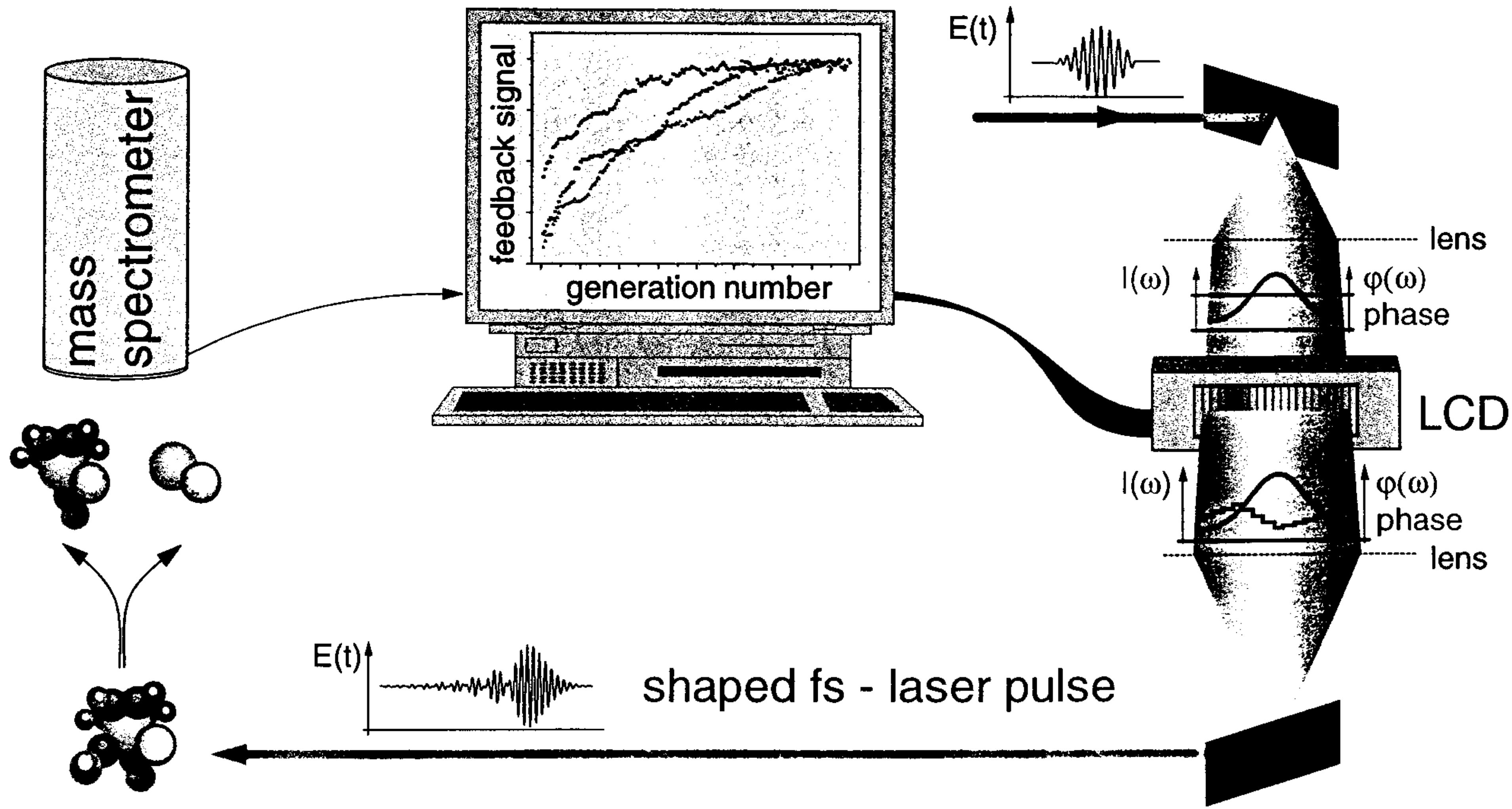

Science **282**, 919 (1998)

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result of optimization

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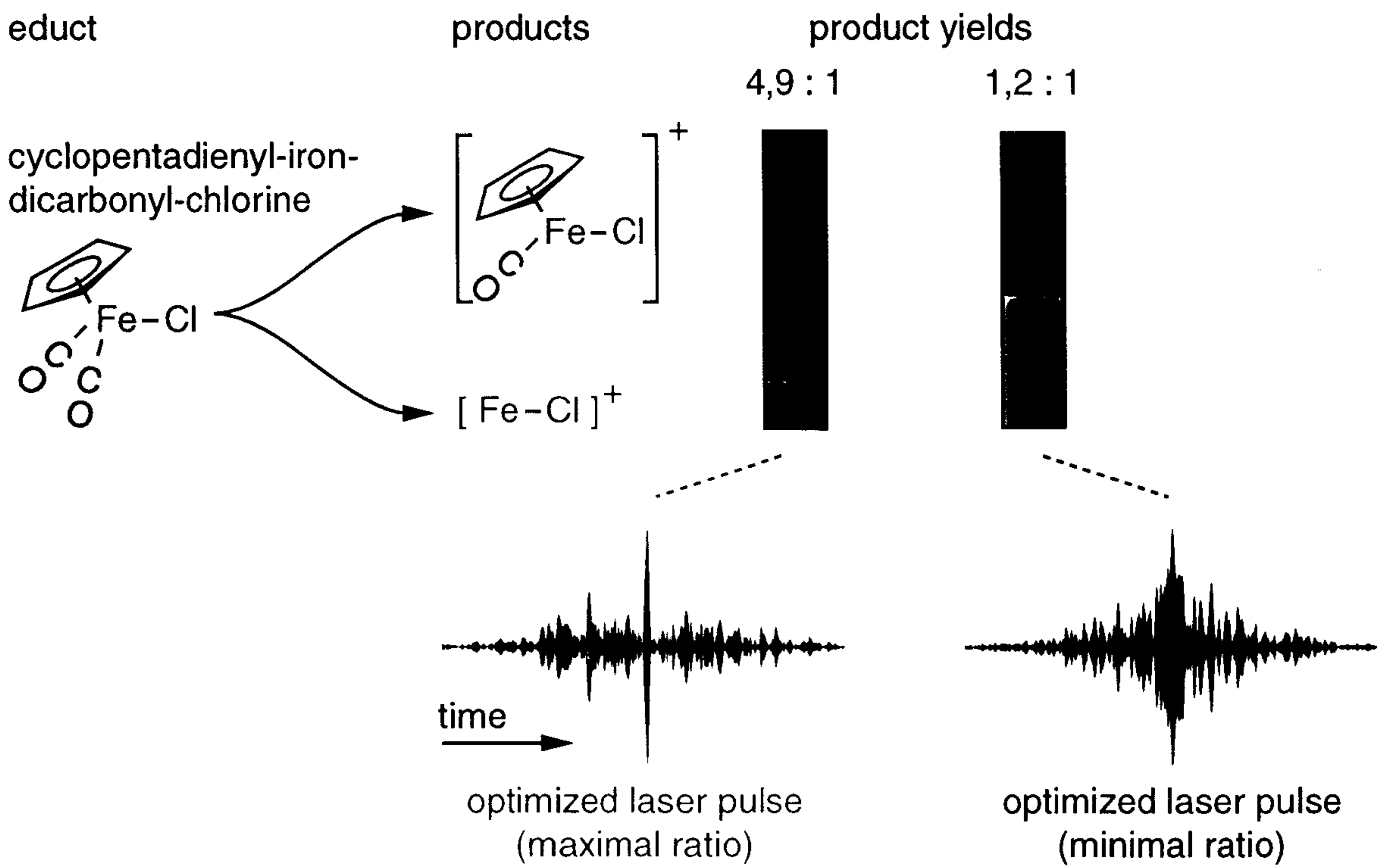

fitness curves for $CpFe(CO)_2Cl$ optimization

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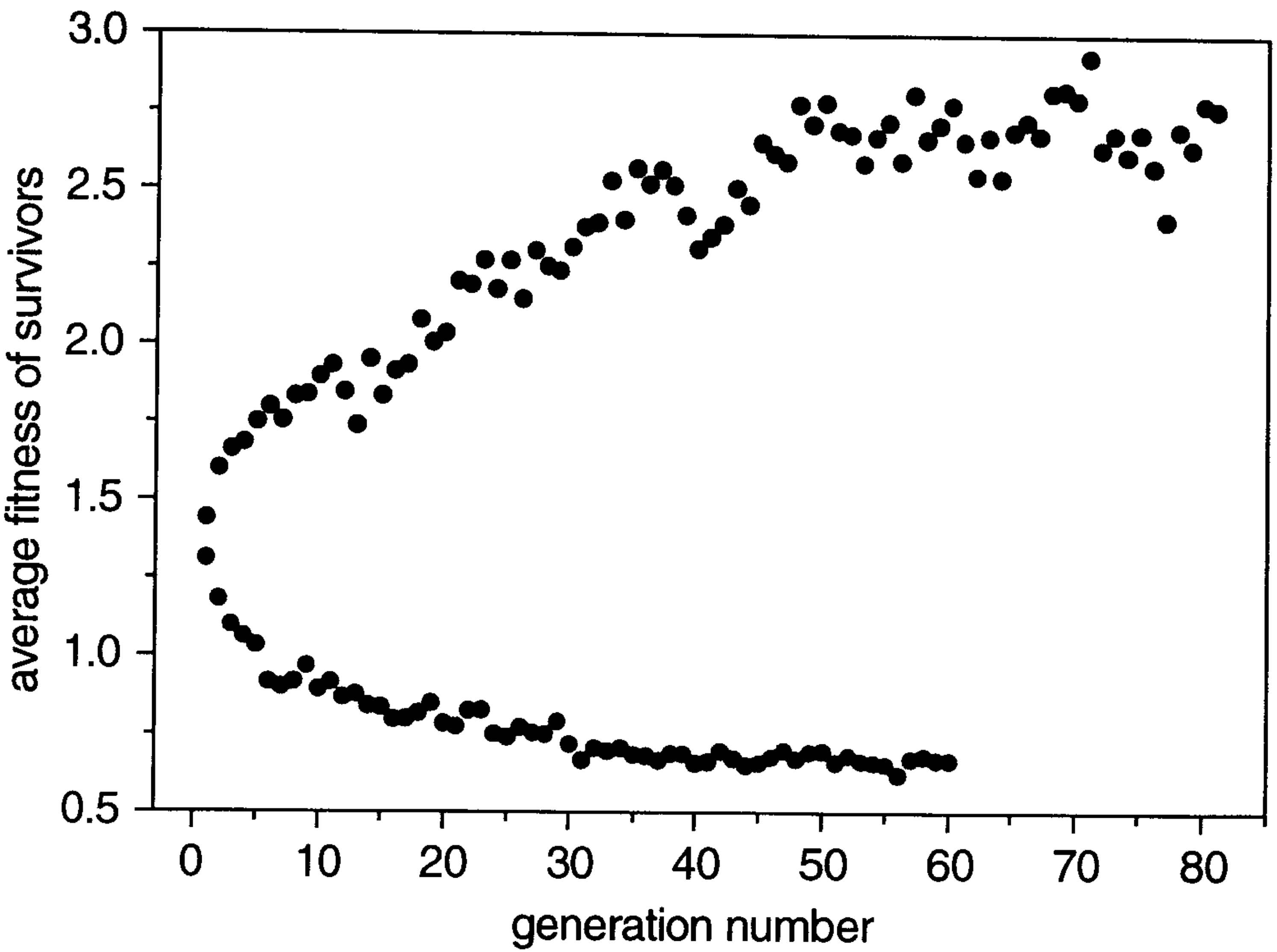

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Summary

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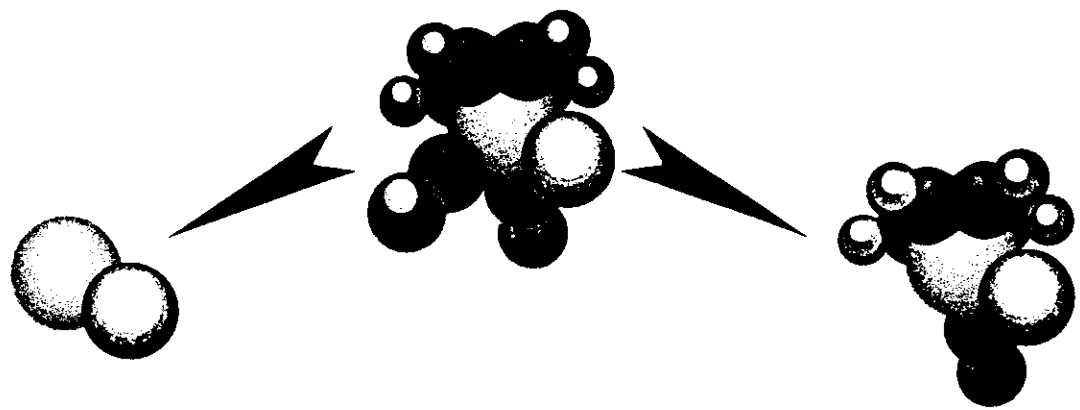
- automated coherent control of photodissociation reactions
- feedback-optimization of phase-shaped fs-laser pulses using an evolutionary algorithm
- $CpFe(CO)_2Cl$: significant changes of ratios of the chemically different products $CpFeCOCl^+$ and $FeCl^+$



Note: no information whatever is needed about the sample substance in the optimization procedure - which always starts with completely random gene configurations