MAX-PLANCK-INSTITUT FÜR QUANTENOPTIK

International Workshop on Optimal Control of Quantum Dynamics: Theory and Experiment

July 25 - 27, 1999
Schloss Ringberg

Organizers:
K.L. Kompa, M. Motzkus, and R. deVivie-Riedle

MPQ 251

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Max-Planck-Institut für Quantenoptik
85740 Garching, Bundesrepublik Deutschland

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International Workshop
on
Optimal Control of Quantum Dynamics: Theory and Experiment

25.-27. July 1999
Ringberg Castle, Tegernsee, Germany

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Scope:

The objective of the conference is to discuss the recent experimental and theoretical results and progress of real time control of chemical dynamics and show perspectives of the future of computerized control experiments.
Special emphasis will be on the following topics:

- Coherent control in femtosecond chemistry
- Femtosecond pulse shaping techniques
- Optimal control theory
- Evolutionary algorithms
- Quantum control of molecular dynamics

Address during the Meeting:

Tagungsstätte der Max-Planck-Gesellschaft
Schloß Ringberg
D-83700 Rottach-Egern
GERMANY

Tel.: +49-8022-279-0
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Program

Sunday, 25 July 1999

shuttle service from airport and railway station to the castle

18:00 - 21:00 Registration and welcome party

Monday, 26 July 1999

Optimal control on quantum dynamics: introductionary lectures

chair: K.L. Kompa

8:30-9:30 H. Rabitz, Princeton University
"Making molecules dance and watching them in the act "

9:30-10:30 W.S. Warren, Princeton University
"Developing a unified view of coherent spectroscopy: what pulse shape control brings to optics"

10:30-11:00 coffee break

Femtosecond pulse shaping

chair: W. Warren

11:00-11:40 D. H. Reitze, A. Efimov, M. D. Moores, N.M. Beach, and J.L. Krause, University of Florida
"Enhancement of strong field molecule ionization using adaptive feedback"

11:40-12:20 K.A. Nelson, MIT
"Excitation, control, and monitoring of propagating coherent excitations: Spatiotemporal femtosecond pulse shaping and spatiotemporal imaging"

12:30-13:50 lunch

chair: C. Bardeen

13:50-14:30 T. Brixner, M. Strehle and G. Gerber, Universität Würzburg
"Feedback optimized compression and shaping of femtosecond laser pulses"

14:30-15:00 T. Hornung, R. Meier, D. Zeidler, and M. Motzkus,
MPI für Quantenoptik
"Adaptive phase modulation of sub 20 fs VIS pulses and two-photon-transition control in Sodium with evolutionary strategies"

Applications on prototype systems (I)

chair: T. Baumert

15:00-15:40 S.R. Leone, R. Uberna, and Z. Amitay, JILA, University of Colorado and National Institute of Standards and Technology
"Multistate phase control of wave packet dynamics"

15:40-16:10 coffee break

16:10-16:50 L. Wöste, FU-Berlin
"Metal Clusters: Good Model Systems for Coherent Control? "

16:50-17:30 M. Dantus, I. Pastrik, V. V. Lozovoy, E.J. Brown, B.I. Grimberg, Michigan State University
"Control and characterization of intramolecular dynamics with femtosecond four-wave mixing techniques"

17:30-18:10 T. Feurer, A. Glass, T. Rozgonyi, and R. Sauerbrey, Friedrich-Schiller-Universität Jena
"Coherent control of the photodissociation process of a diatomic molecule using a feedback controlled self-learning fs-laser system"

18:30-20:00 dinner "Bayerischer Abend"

20:00- Guided tour of Schloß Ringberg

Tuesday, 27 July 1999

Applications on prototype systems (II)

chair: D. Proch

8:30-9:10 L.D. Noordam, FOM-Institute AMOLF Amsterdam
"Quantum ladder climbing with chirped pulses"

Aspects of optimal control theory

chair: J. Krause

9:10-9:50 J. Manz, FU-Berlin
"Theory of selective preparation of enantiomers by laser pulses"

9:50-10:30 Y. Fujimura, Tohoku University
"Quantum control of reaction dynamics by a locally optimized control method"

10:30-11:00 coffee break

chair: H. Rabitz

11:00-11:40 K. Sundermann and R. de Vivie-Riedle, MPI für Quantenoptik
"Optimal control theory - a tool to discover new reaction mechanisms"

11:40-12:20 J. Cao, MIT
"Phase-coherence in ultrafast non-linear optical excitation"

12:20-13:00 Y. Yan, Hong Kong University
"Semiclassical methods in optimal control and detection of molecular dynamics"

13:00 -14:00 lunch

Applications on complex systems

chair: R. Sauerbrey

14:00-14:40 C. Bardeen, University of Illinois
"Applications of shaped femtosecond pulses in condensed phase molecular spectroscopy"

14:40-15:20 M. Bergt, T. Brixner, B. Kiefer, M. Strehle, and G. Gerber, Universität Würzburg
"Control of chemical reactions with optimally tailored femtosecond laser pulses"

15:20-16:00 E. Riedle, A.J. Wurzer, and S. Lochbrunner, Ludwig-Maximilians-Universität, München
"Highly localized vibronic wavepackets in large reactive molecules"

16:00-16:30 coffee-break

chair: K. Nelson

16:30-17:10 M. Gruebele, University of Illinois
"Ultrafast laser control of molecular vibrational dephasing"

17:10-17:50 J.L. Krause, University of Florida
"Quantum control in semiconductor heterostructures"

17:50-18:30 A. Sizmann and G. Leuchs, Universität Erlangen
"The multimode quantum structure of coherent solitons"

19:00- Barbecue on the terrace

Wednesday, 28 July 1999

departure, shuttle service to airport and railway station
and for those who are interested a lab tour of the MPQ is provided.

MAKING MOLECULES DANCE AND WATCHING THEM IN THE ACT

Herschel Rabitz
Princeton University

The ability to control events at atomic dimensions and ultrafast time scales opens up a number of applications with unprecedented capabilities. Manipulation of events in this domain demands new algorithms to work within the laws of quantum mechanics operative in the microworld. In addition, observation of the ultrafast controlled dynamical processes is leading to the technology for producing molecular-scale "movies" of the ensuing events. This research is concerned with algorithm development to (a) guide the laboratory control efforts, and (b) invert the "movies" for their atomic-scale information content.

"Developing a unified view of coherent spectroscopy: what pulse shape control brings to optics".

Warren Warren
Princeton University

Recent advances in laser technology have made it possible to create complex multiple-pulse sequences with pulse shaping and full interpulse phase control. Particularly in the infrared, where coupled vibrational excitations may reflect energy transfer, this has led to renewed interest in optical analogs of two-dimensional NMR spectroscopy. I will discuss the fundamental advantages and disadvantages of optical coherent spectroscopy, as compared to NMR, and point out the likely directions where coherent laser pulses can contribute new molecular information.

Enhancement of Strong Field Molecule Ionization Using Adaptive Feedback

David H. Reitze[1], Anatoly Efimov[1], Mark D. Moores[1], Nicole M. Beach[1], and Jeffrey L. Krause[2]
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University of Florida
Gainesville, FL

The interaction of intense femtosecond laser pulses with atoms and molecules and resulting ionization is, in general, a highly complex phenomenon which depends sensitively upon the pulse energy, spatial profile, and temporal pulse shape as well as on the specific properties of the atomic or molecular species. The ability to control these interactions is hampered by the strong field nature of the problem and its highly nonlinear nature. Nonetheless, adaptive learning algorithms [1] combined with ultrafast pulse shaping tehniques [2] are emerging as potentially powerful ways of controlling laser-matter interactions. *Experimental* adaptive feedback methods have the advantage of optimizing a particular set of pulse parameters or signal in the presence of noise and experimental uncertainty [3,4].

In this talk, we present results of recent experiments in which we "train" an amplified laser pulse to enhance the ionization rate during molecular ionization by adaptively controlling its phase [5]. Our experiments have a dual purpose. First, we seek to explore how the laser can learn to optimize a particular *complex* experimental signal, in this case a spectral blueshift of the pulse that occurs during strong field ionization [6]. The spectral shift has been shown to be a sensitive probe of the pulse intensity. Using the magnitude of blueshifted light in a molecular ionization experiment as an indicator of pulse intensity and shape, we show that the amplifier can actively determine the optimal amount of residual phase for enhancing the rate of ionization. Second, we seek to minimize phase dispersion in the amplifier using adaptive feedback. Controlling the amplitude and phase of femtosecond optical pulses and their interactions with matter is a topic of active research within the ultrafast laser community. In particular, minimization of the higher order frequency-dependent phase dispersion in ultrashort chirped pulse amplifiers has been found to be essential for obtaining high fidelity amplified pulses.

In our experiments, a genetic algorithm (GA) is used in conjunction with a chirped pulse amplifier to update the phase of a laser pulse through pulse shaping and then monitor the ionization process by measuring the pulse spectrum after the ionization has occurred. We examine how varying the complexity and size of the parameterization of the laser pulse affects the convergence of the GA and find that the optimization process is robust, finding the best pulse for enhancing ionization independent of initial conditions. We also compare our results to strong field ionization theory.

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EXCITATION, CONTROL, AND MONITORING OF PROPAGATING COHERENT EXCITATIONS: SPATIOTEMPORAL FEMTOSECOND PULSE SHAPING AND SPATIOTEMPORAL IMAGING

Keith A. Nelson, MIT Department of Chemistry

Femtosecond pulse shaping methods are now rather well developed and their use for coherent control over molecular and material responses is well established. For most cases, the coherent response does not propagate in space. Therefore the excitation field needs to be specified as a function of time only, and the sample response needs to be monitored as a function of time only. For some excitations, however, coupling to electromagnetic radiation results in mixed (polariton) modes which move through the host medium at light-like speeds. Control over propagating responses requires specification of the excitation field and monitoring of the responses as functions of both time and position. Spatiotemporal femtosecond pulse shaping, through which a single input beam with a single femtosecond pulse is converted into a spatially and temporally shaped output, e.g. many separated beams with independently specified time-dependent waveforms, has recently been developed and implemented in an automated manner. Spatiotemporal imaging, through which the location and spatial characteristics of propagating responses can be monitored at specified times, also has been demonstrated. These techniques together provide the basis for control over coherent propagating responses. Applications include characterization and shaping of linear and nonlinear polariton responses and generation of specified, spatiotemporally shaped THz-bandwidth signals.

Feedback-Optimized Compression and Shaping of Femtosecond Laser Pulses

T. Brixner, M. Strehle, and G. Gerber
Physikalisches Institut, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Adaptive femtosecond pulse shaping in combination with Ti:Sapphire oscillator pulses has already been demonstrated [1,2]. Many experimental applications in the fields of optics and femtochemistry, however, require amplified laser pulses either transform-limited or specifically tailored.
We recently reported on automated compression of amplified femtosecond laser pulses [3] employing a programmable liquid crystal display (LCD) based pulse shaper following the design introduced by Weiner et al. [4]. This pulse shaper is feedback-controlled by an evolutionary algorithm. We are able to generate transform-limited laser pulses ($\tau \approx 80$ fs @ 800 nm) (see Fig. 1) with pulse energies of up to 0.6 mJ corresponding to a relative energy throughput of 65 %. In contrast to other groups [5,6], our pulse shaper is placed behind the CPA system. It hereby acts as a stand-alone device that can be operated with any kind of input laser pulses, no matter whether they originate directly from the laser system, from a frequency conversion unit or from any nonlinear process (e.g. hollow fiber spectral broadening). It is furthermore possible to shape both unamplified Ti:Sapphire oscillator pulses and amplified femtosecond laser pulses.

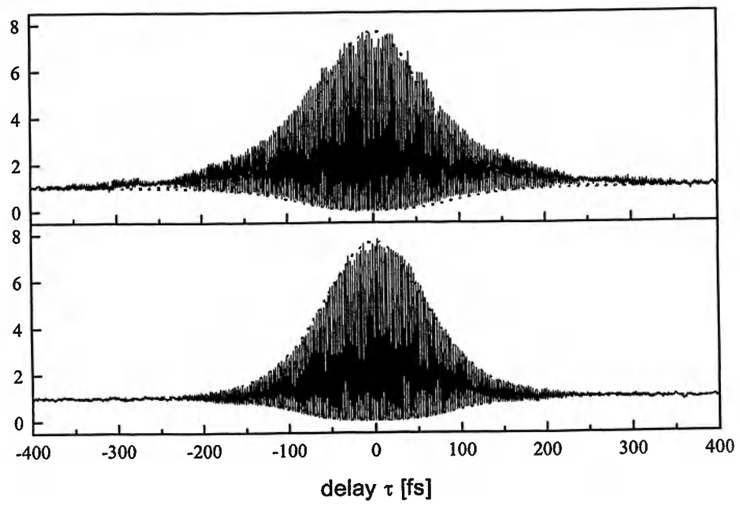

Fig. 1: Interferometric autocorrelations, recorded before and after the optimization procedure.

In a second experiment we employed a method to generate almost-arbitrarily shaped femtosecond laser pulses [7,8]. We therefore combined the TADPOLE method (**T**emporal **A**nalysis by **D**ispersing a **P**air **O**f **L**ight **E**-fields) [9], which is suitable for complete characterization of the electric field, and the frequency-domain pulse shaper (see Fig. 2). The measured pulse shape (which acts as feedback) is compared with the desired target pulse shape and a simple local convergence algorithm adjusts the voltages applied to the LCD in the pulse shaper such that the phase difference is compensated for each LCD pixel. An advantage of this non-iterative and thus „real-time" method (less than one second) is that fundamental limitations (e.g. pixelated spatial filters, inter-pixel gaps) and optical deficiencies (e.g. lens abberations) are corrected automatically. Consequently, specifically designed pulse shapes in either time or frequency domain can be provided with high fidelity.

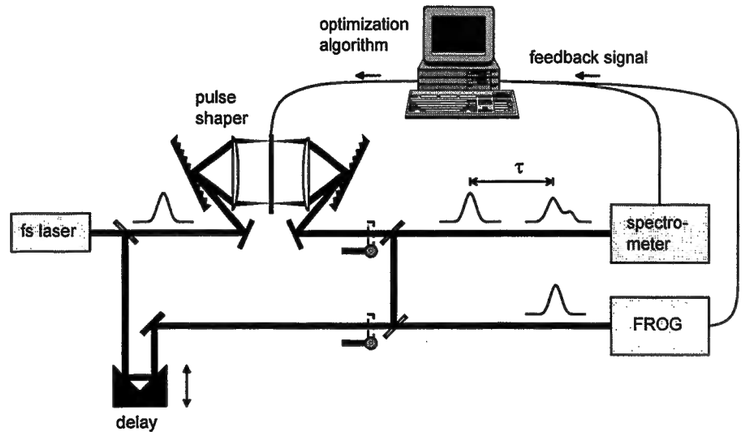

Fig. 2: Experimental setup for feedback-optimized shaping of femtosecond laser pulses.

References:

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[9] D. N. Fittinghoff et al.: Opt. Lett. **21**, 884 (1996)

Adaptive phase modulation of VIS pulses to below 20fs and two-photon-transition control in sodium with evolutionary strategies

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The adaptive phase modulation of tunable pulses in the visible generated by a noncollinear OPA is demonstrated. Pulse compression is achieved by a combination of pulse shaper and feedback-controlled evolutionary algorithm. By this, pulse durations below 20fs are produced through dispersive material in the beam path. This provides a useful tool for optimal control experiments.
Another application of evolutionary algorithms to a physical problem is the generation of both "bright" and "dark" pulses in the two-photon-transition of sodium leading to enhancement and extinction of the fluorescence from the excited state population is demonstrated. In both applications of evolutionary strategies, convergence is achieved securely and fast.

Adaptive pulse compression

The compression of broadband spectra produced by Optical Parametric Amplifiers (OPA) with non-collinear type phase-matching [1] to pulse durations below 20fs has commonly been achieved by prisms or gratings. Instead, a pulse shaper [2,3] is now used for this task. The desired shaping of the pulses can be contrived by an evolutionary algorithm in a feedback loop [4,5]. By this technique, phase functions of arbitrary appearance can be compensated.

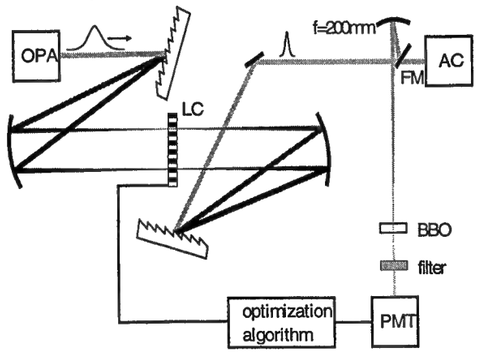

Fig. 1: Setup of the computer-controlled compressing unit

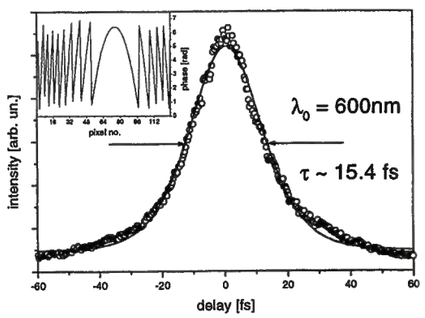

Fig. 2: Autocorrelation trace after phase optimization. Inset: Phase values retrieved by the algorithm.

The broadband pulse shaper is built up by reflective optics throughout (Fig. 1) [6]. A pixelated liquid crystal (LC) mask is placed in the Fourier plane which is capable of influencing phase (and amplitude) of every spectral component. The adjustment of the phase for maximum second harmonic (SH) signal is solved by an evolutionary algorithm. We chose a polynomial representation of the phase function with quadratic terms as lowest order and simulated realistic experimental conditions (dispersive material in the beam path between compressor and experiment) by installing an ethanol-filled cuvette in front of the SH crystal. The shaping apparatus is capable of adaptive pulse compression down to below 16fs (Fig. 2) by optimizing the polynomial coefficients. The optimization process typically takes three minutes.

Adaptive two-photon-transition control in sodium

The feedback scheme was applied to control the efficiency of pumping the two-photon-transition 3s-5s in sodium [7].

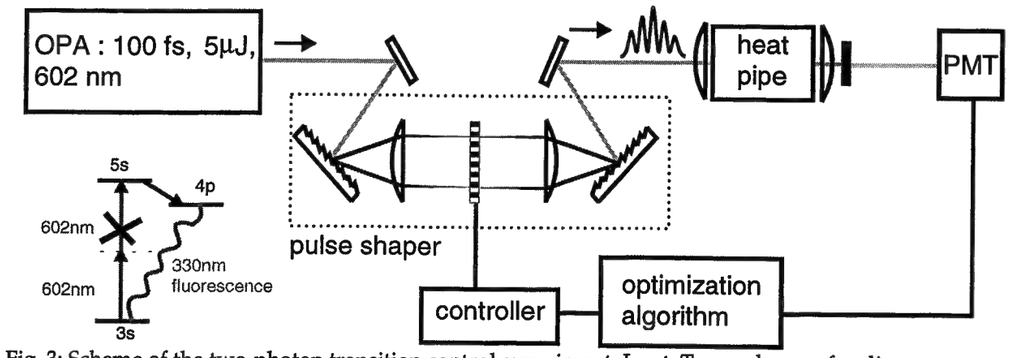

Fig. 3: Scheme of the two-photon transition control experiment. Inset: Term scheme of sodium.

The experimental setup is shown in Fig. 3. The population of 5s relaxes by collisions to 4p, a fluorescent level slowly decaying to the ground state (Fig. 3, inset). The resulting 330nm emission is a measure for the 5s population and served as feedback signal for the algorithm.
For the two-photon excitation probability S_2 all pairs of sequential one-photon transitions (ω_1, ω_2) are taken into account whose frequencies are present in the pulse spectrum and which may cooperate to reach the excited state $\omega_1 + \omega_2 = \omega_0$. The final population is calculated by considering all these paths. A control experiment may be designed to achieve two opposing optimization goals:
Maximize S_2: A Fourier limited pulse with a constant spectral phase leads to maximal transition. However, a phase shaped pulse with an antisymmetric phase function $\phi(\omega_0/2 - \Omega) = -\phi(\omega_0/2 + \Omega)$ yields the same result.
Minimize S_2: Theory predicts [7] that pulses with a symmetric spectral phase function $\phi(\Omega)=\cos(\beta\cdot\Omega)$ cancel the transition probability ("dark pulses").

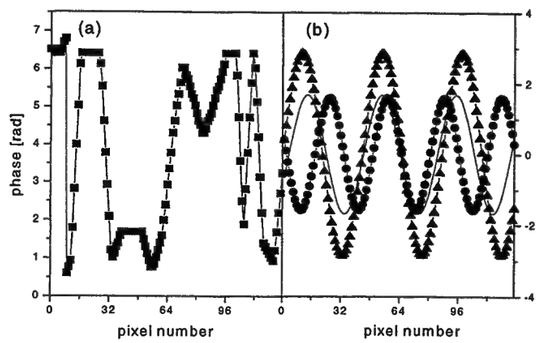

Fig. 4: Phase functions retrieved for the enhancement of the two-photon transition. a) Free optimization. b) Phases restricted to periodic functions.

In a first experiment, a spectral phase $\phi(n) = a \cos(b \cdot n + c)$ was put onto the mask, n being the pixel number. The task of the algorithm was to optimize the parameters a, b and c to achieve fluorescence enhancement and extinction, respectively. All phase filters retrieved show symmetry for extinguishing and antisymmetry (Fig. 4, right) for enhancing fluorescence.
In a further experiment, new solutions producing the same fluorescence rate as in the pervious 3-parameter approach were found by a linear interpolation of the optimized phase. As postulated by theory, there is still antisymmetry remaining in the phase functions found for the case of enhancing the fluorescence (Fig. 4, left).
The shaped pulses show a complicated phase and amplitude time structure. It is important to note that the duration of both "bright" and "dark" pulses is about 2 ps. Hence pulses with equal power density can either suppress or maximize population transfer only by virtue of their special pulse shape. As the problem is not specific to the particular system studied here, these solutions are of general interest for controlling two-photon processes in any coherently excitable media.

References

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Multistate Phase Control of Wave Packet Dynamics

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Phase and amplitude control of vibrational and rotational wave packets is investigated with precise detail in the lithium dimer E($^1\Sigma_g^+$) shelf state. This state is chemically interesting because it switches from covalent on the inner turning point to ionic on the outer turning point. The preparation and probing is carried out with an ≈100 fs Ti:sapphire/OPA ultrafast laser system. An important aspect of these experiments is the use of a single-mode cw laser for intermediate state selection prior to the preparation of the wave packet. Such intermediate state selection provides precise knowledge of the wave packet components and thus permits a very high degree of control over the exact coherent superposition. A second aspect is phase and amplitude control, using a liquid crystal spatial modulator to shape the electric field of the femtosecond pulses. This method is used to actively set independently the phases and amplitudes of as many as 10 rovibronic states in the prepared wave packet.

The experiments employ a three photon excitation sequence. A single mode cw laser is used to select an individual vibrational and rotational state in the A($^1\Sigma_u^+$) electronic state of Li_2. This is the intermediate state from which the wave packet is launched. A 100-200 fs pulse from a Ti:sapphire regenerative amplifier prepares the coherent superposition of states from among 10 rovibronic levels in the E electronic state. A second, time-delayed, ultrafast pulse ionizes the molecules at a specific localization of the wave packet (internuclear separation and rotational angle). The dimer ions are detected as an electrical current by a pair of biased electrodes in the lithium heat pipe. Phase and amplitude pulse shaping is performed with a 128 pixel liquid crystal spatial light modulator to alter the electric field of the laser pulse in either the wave packet preparation (pump) step or the detection (probe) step.

This technique is applied in experiments to control the time evolution of the assembled wave packet and to achieve focusing of the wave packet at a specific localization. Such focusing is reflected as maxima and minima of the probe ionization signal at specified times after the pump step. Amplitude control experiments are performed to alter the coefficients of the amplitudes of the individual rovibronic states that make up the wave packet. Different wave packets can be formed out of the same set of levels, but with altered amplitudes of the individual components, resulting in dramatically different time evolutions. Using the phase control capabilities of the liquid crystal modulator, rovibronic wave packets composed of eight strongly excited levels, each with a pre-set phase, are prepared. Specific limits on the degree of such phase control in wave packet focusing experiments have been derived, based on the signs of the various quantum beats. In one particular experiment, the signs of the individual quantum beats in the wave packet recurrences are all found to be positive. Thus with an initial uniform phase of all the wave packet states, a maximum in the ionization signal is produced at time t=0. At any later time, this maximum can be produced by phase control. However, an amplitude minimum of this same magnitude cannot be produced, since the phases of only eight quantum mechanical states can be controlled, whereas 18 quantum

mechanical beats contribute significantly to the wave packet time evolution. A phase function that produces a minimum can be found numerically, but in the present case it is not necessarily a true global minimum of the specific set of quantum beats. In the talk, the limitations on the maximum and minimum spatial localization of the wave packet and the origins of the signs of the beats are considered in further detail.

Also, two-level rotational coherence spectroscopy, which permits measurements of the frequency splittings between pairs of high rotational states, has been developed. In these experiments, the relative phase of the rotational quantum beats is controlled with an accuracy of 0.02 radians. By monitoring the modulation amplitude of the two-level rotational wave packet signals, while amplitude shaping the ionization (probe) pulse, Rydberg states embedded in the ionization continuum have been mapped. The presence of a single Rydberg state in the ionization continuum permits excitation of a single final state in the probe step. With this advantage, phase control of the detection (probe) step has been demonstrated for the first time. Thus it is possible to use tailored femtosecond probe pulses to select specific information in a wave packet prepared in a complex superposition of states. In addition, using a tunable ultrafast probe laser (OPA) and the two-level rotational wave packet signal amplitude as a tool for background-free measurements of the ionization cross section, the ionization cross section of the E electronic state versus final vibrational state in the ion has been mapped. This cross section is highly correlated with the ionization probability as a function of internuclear distances, which changes markedly in the E state of the lithium dimer.

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Metal Clusters: Good Model Systems for CoherentControl?

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The complexity of photochemical systems is a most importantparameter with regard to coherent control scenarios, in which active modesshould independently be addressable from the bath. This emphasizes to investigate model systems, which allow progressly toincrease the number of active and passive modes. We employed transient two-and multiphoton ionization spectroscopy on metal clusters of a selectedsize in order to probe their wavepacket dynamics, structural reorientations, charge transfers anddissociative events in their different vibrationally excited electronicstates including their ground state. The experiments were carried out onfree neutral particles in the vacuum, on cluster ions confined in a trap, on particles surrounded bynon-chromophormic ligands, and on clusters softly deposited on a surface.This allowed us to observe the influence of IVR-processes as a function ofthe internal degrees offreedom of the system. An important parameter in this regard is itstemperature, which defines the starting coordinate of the initiatedtrajectories at the moment of the pump-pulse and also the stabilizationtime for theresulting geometry. The most important parameter in this regard are theshape and amplitude of the employed excitation pulse. This rises hopes forcontrolling the intramolecular dynamics with different chirp settings. Their influence on fragmentation patterns will be discussed and comparedwith conventional photo- ionization and electron-impact experiments.

Control and characterization of intramolecular dynamics with femtosecond four-wave mixing techniques

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Femtosecond four-wave mixing (FWM) techniques are used to characterize and in some cases control molecular dynamics. Off-resonance transient-grating measurements reveal the dynamics of ground state molecules. On-resonance transient-grating measurements provide information about ground and excited state dynamics. Strategies for achieving control over population transfer and coherence will be discussed based on the density matrix formalism for FWM processes. Experimental demonstrations will be included.

Related Publications from our group:

1. E. J. Brown, Q. Zhang, and M. Dantus, *J. Chem. Phys.* **110,** 5772 (1999).
2. I. Pastirk, E. J. Brown, B. I. Grimberg, V. V. Lozovoy, and M. Dantus, *J. Chem. Soc. Farad. Discuss*. **113,** (discussion in July 1999).

Coherent control of the photodissociation process of CsCl using a feedback controlled self-learning fs-laser system

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Abstract

The time-resolved photo-induced dissociation process of a diatomic molecule (CsCl) was investigated both theoretically and experimentally. It is shown that by applying a sequence of two different laser pulses the dissociation process may be controlled, according to the Tannor-Rice scheme. Using feedback-controlled optimization algorithms a self-learning system has been realized, which is able to coherently control the product channel without a priori knowledge on the molecular potential surfaces.

The desire to control the reaction path of photo-induced chemical processes with high efficiency has been a major goal for some thirty years [1,2]. In the last ten years novel concepts emerged for the control of quantum dynamics [2,3,4]. Most of them rely substantially on a real time control of the wavepacket dynamics in a chemical system using ultrashort laser pulses. In addition to the progress in theory, new developments in laser technology, especially the ability to tailor ultrashort laser pulses in the time and frequency domain, had an enormous impact on the field. It was suggested by Judson and Rabitz [5] that by combining optimizing algorithms and the methods of femtosecond pulse shaping a self-learning system may be constructed which is able to control a femtosecond laser driven chemical reaction without a priori knowledge of the details of the reaction pathways. In the present work, following a pump-dump scheme proposed by Tannor and Rice [3] a sequence of two phase controlled laser pulses having different wavelengths is used to coherently control the photo-induced dissociation of CsCl. A theoretical analysis is also presented revealing the mechanisms that lead to the different product channels. In addition, a self learning system is realized which is able to optimize the efficiency of one product channel over the other without detailed knowledge of the system parameters. This experiment thus demonstrates that a self-learning phase controlled femtosecond laser system is able to control a quantum system according to the Tannor-Rice scheme.

The laser system delivered pulses at 497 nm with a minimum pulse width of 50 fs. Using a compressor consisting of SF10 prisms the pulse width could be varied. The pulses were frequency doubled in a 300 um BBO crystal. After separating the second harmonic radiation

from the fundamental a variable delay was introduced between the two pulses. Finally the beams were recombined collinearly and focused into the target chamber. Inside the evacuated target chamber the laser pulses interacted with a CsCl molecular beam, which emerged from an oven heated to 875 K. The Cs ions were detected by a quadrupole mass spectrometer (QMS). The QMS had no active ion source but rather the Cs ions emerging from the molecular beam after photodissociation were gathered by a simple ion optics. The electric field of the pulses emerging from the compressor were fully characterized by a second harmonic generation FROG system. The temporal duration of the second harmonic radiation at 248 nm was determined by a crosscorrelation measurement in the target chamber. The crosscorrelation measurement was not only used to determine the temporal duration of the laser pulse at 248 nm, but also to determine accurately the time delay zero between the two pulses, which will be important in the discussion of the experimental results.

The electronic ground state X is ionically bound and the two excited electronic levels A and B correspond to the ground and the excited electronic states of the Cs atom. These electronic levels and the continuum are the important levels because they are resonantly or almost resonantly coupled by the two wavelengths used in the experiment. In order to numerically simulate the wavepacket dynamics on the four potential levels the second order split operator method has been used. The wavepacket dynamics can be illustrated as follows. A short UV laser pulse (248 nm) excites the stationary ground state wavepacket to the repulsive first excited electronic level A. From there the wavepacket propagates towards larger internuclear separations. Since it is subjected to dispersion it spreads in space. At any given (positive) time delay the pulse at 497 nm interacts with the wave packet and gives rise to an approximately constant Cs ion signal via a two photon process. Since the two photon process is non-resonant and the interaction takes place with a fast moving wave packet the ionization process is inefficient and the ion signal that appears as a constant background is relatively weak. Choosing the time delay between the two laser pulses so that the second laser pulse (497 nm) is able to resonantly deexcite the wavepacket back to the ground state X changes the situation drastically. The numerical simulations show that the time the excited wavepacket needs to propagate to this point is about 160 fs. If the delay between the two pulses is 160 fs part of the wavepacket is transferred back to the ground state X via stimulated emission. Therefore, at a delay of 160 fs the yield of Cs ions will increase due to the additional channel that is opened up by deexcitation and subsequent ionization. Note, that this second channel is much more efficient than that producing the background. Since the intensity of both pulses high enough in order to transfer a substantial fraction of population (>10%), the dynamics corresponds to the strong field regime. Figure 1 shows the calculated Cs ion yield as a function of the delay between the two laser pulses (solid line). Positive delay times correspond to a situation where the pulse at 248 nm precedes the pulse at 497 nm. In addition the dots represent the experimental data. From the figure it is clear that the agreement between the calculated and measured curve is very good.

In order to realize a feedback-controlled self learning system, the experimental setup has been modified in the following way. Three variables, the separation of the two prisms in the compressor, the delay time between the two pulses, and the position of the focusing lens have been varied by computer controlled stepper motors. The Cs ion yield measured by the QMS was used as feedback signal. Two different algorithms optimizing the ion yield have been applied, namely a Simulated Annealing (SA) code and a Genetic Algorithm (GA). First, the optimization has been performed using propane instead of CsCl. The algorithm (Simulated Annealing) was able to maximize the propane ion yield as a function of the three parameters. The same SA algorithm was applied to the CsCl dissociation. The position of the three stepper motors as a function of the number of iterations is shown in figure 2a) to c). The corresponding Cs ion signal is shown in figure 2d). Clearly the algorithm is converging to settings of the three stepper motors where the Cs ion signal is maximized. For arbitrary settings of the control parameters before starting the optimization process the Cs ion signal is below the detection limit of 120 mV. After optimization the signal increased by a factor of 40 with respect to the detection limit. The most obvious difference between the two experiments (using propane and CsCl) is the optimal delay position found by the algorithm. It corresponds to a difference in delay of two times 0.023 mm or 155 fs in excellent agreement with the predictions by the numerical simulations.

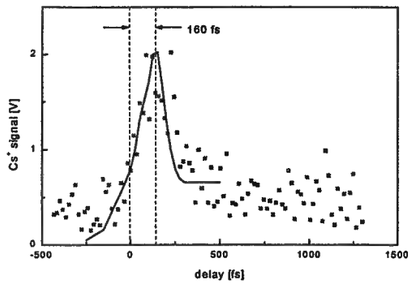

Fig.1: The Cs ion yield is shown as a function of the delay time between the two laser pulses. The dots represent experimental data and the solid line is the result of wavepacket simulation.
The parameters for both the simulation and the experiment were for 248 nm: 186 fs, 0.45 TW/cm^2 and for 497 nm: 86 fs, 4.7 TW/cm^2.

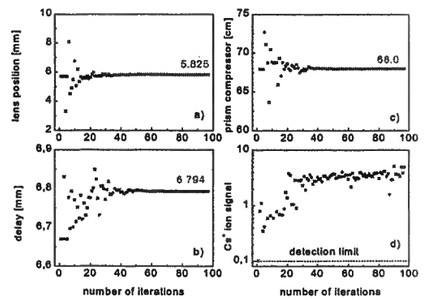

Fig.2: Optimal control of photodissociation channels in CsCl by using a Simulated Annealing algorithm. In the figures a) to c) the position of the three stepper motors is shown as a function of the number of iterations. Clearly all three motors converge to a final position. From the corresponding Cs ion yield shown in d) it is obvious that the algorithm successfully optimizes the ion yield.

In conclusion it has been demonstrated that by using a sequence of two femtosecond laser pulses the photodissociation of CsCl can be effectively controlled. Furthermore, a feedback-controlled self-learning system was demonstrated which is able to coherently control the product channel of the photo-induced dissociation process of CsCl. To the best of our knowledge this is the first realization of a self learning Tannor-Rice pump-dump scheme in the strong field regime.

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Quantum ladder climbing with chirped pulses

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Abstract. Enhanced vibrational ladder climbing has been demonstrated in NO using picosecond, chirped, free-electron laser pulses. Three steps of the vibrational ladder are climbed within a single infrared laser pulse. Quantum interference in the total yield of rotational pathways is observed.

1. Quantum ladder climbing in atoms and molecules

Laser excitation of molecules into a specific highly-excited vibrational state is a controllable way to enhance their chemical activity. Direct population of such a highly-excited state by absorption of a single photon is very inefficient. Step-wise excitation of a sequence of vibrational states is a more promising route. However, this method is frustrated by the anharmonicity of the molecular potential: an infrared laser which has the appropriate frequency to drive a molecule from the vibrational ground state v=0 to v=1 is non-resonant with consecutive vibrational transitions (v=1 to v=2, v=2 to v=3 .....). To overcome this bottleneck, the use of broadband short laser pulses that contain more frequencies in their spectrum has been suggested [1]. Such broadband pulses can be ``chirped", that is, their frequency can be made to change in time to follow the consecutive vibrational transitions. This chirping should strongly enhance the transfer to the higher states. Moreover, ultrafast excitation of a specific bond in a poly-atomic molecule by chirped short pulses can outpace intramolecular relaxation processes and, hence, enables bond-selective excitation.

In a series of experiments [2-4] we demonstrated efficient quantum ladder climbing of *electronic* transitions in *atomic* rubidium (5s -> 5p -> 5d). Using chirped 780 nm pulses of a large range of energies we found the following two regimes: (I) At low fluence the chirp enhances the population transfer if the pulses are chirped such that the laser frequencies match the consecutive transitions. The total population in the upper state oscillates as a function of chirp due to interfering pathways: sequential photoabsorption (5s -> 5p -> 5d) and direct two-photon absorption (5s -> -> 5d). (II) At high fluence the chirped pulse transfers all population to the upper state following the sequential pathway (the frequencies in the pulse are ordered in the intuitive way), moreover even for counter-intuitive chirp all population is transferred in the direct two-photon process. Remarkbly only unchirped pulses, having the highest intensity, are unable to inverse the population!

Up till now, laser experiments have not been very successful in driving *vibrational* ladder climbing in *molecules* for two reasons. Firstly, the required radiation to drive vibrational transitions is in the infrared (IR). In that wavelength regime, lasers that are both intense and tunable are scarce. Secondly, the bandwidth of most high power IR lasers is too small to resonantly drive consecutive transitions in the anharmonic potential. Nowadays, free-electron lasers provide tunable ultrashort infrared pulses with a sufficiently broad frequency spectrum to drive several consecutive transitions. We report on population transfer to high vibrational states of NO using broad band, chirped, IR laser pulses from the free-electron laser FELIX.

We studied [5,6] the enhancement of population transfer through the vibrational ladder of NO as a function of the chirp of an ultrashort IR laser pulse. The experiment was organized as follows. Short (0.37 ps) bandwidth-limited IR pulses from a free-electron laser drove the vibrational ladder climbing process in the diatomic molecule NO. The laser was tuned to the frequency matching the vibrational transitions in NO with sufficient bandwidth to support the v=0 to v=1 transition up the v=2 to v=3 transition.

2. Interfering rotational pathways in the vibrational ladder

As an example we show the population in v=3 as a function of the chirp (α) of the infrared laser pulses. Negative values of α correspond to a decreasing laser frequency as a function of time. Hence for negative α, the laser frequency follows the decreasing vibrational transition frequencies. Figure 1 shows the three main observations reported here: (I) The broad bandwidth of the IR laser pulse enables population of the v=3 vibrational state by three consecutively resonant photons. (II) A strong enhancement of this population is observed when the chirp follows the consecutive steps up the vibrational ladder. (III) The population in excited vibrational states shows an oscillatory behaviour as a function of the chirp of the IR laser pulse.

The interferences originate from sequential transfer to the same final ro-vibrational states via different intermediate states. The interference is due to the accumulated phase difference of pathways involving different intermediate rotational states, as confirmed in a multistate time dependent quantum calculation (curve in figure 1).

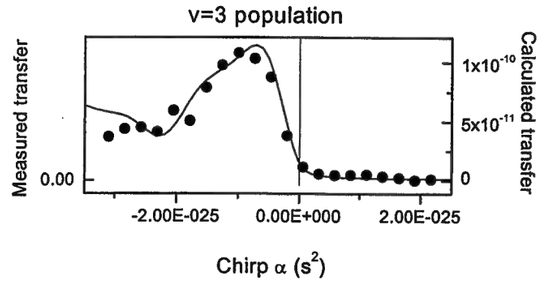

Fig. 1: Population of the v=3 vibrational mode of NO as a function of the chirp. For negative chirp the swept laser frequency follows the frequency of consecutive vibrational transition. The experiment (dots) shows indeed enhanced population transfer for negative α, as well as the destructive interference of the rotational pathways for $\alpha \sim -2\ 10^{-25}$.

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Theory of Selective Preparation of Enantiomers by Laser Pulses

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After a brief summary of the history of the field, including the fundamental papers of Shapiro and Brumer [1], Hänggi [2,3], and Quack [4], I shall present a new approach which is being developed in cooperation with the group of Y. Fujimura [5]. Specially, we consider polyatomic nonplanar molecules in the electronic ground state, with symmetric double well potentials supporting two enantiomers, which are related to each other by a reflection. Initially, we assume that the system is prepared in a symmetric state, e.g. the vibrational ground state, representing a racemate with equal probability for the two enantiomers. Then we show that one may use optimal elliptically polarized laser pulses in order to prepare localized non-symmetric states, representing specific enantiomers. Our approach will be demonstrated by a one-dimensional quantum mechanical model simulation for the system H_2OPSH, based on an ab initio potential energy surface, and dipole couplings. Specifically, we show that the laser pulse can be used to drive the dihedral angle of the non-planar fouratomic fragment OPSH towards specific configurations of the enantiomers, assuming preorientation of H_2OPSH. I shall also discuss some extensions which are based, in part, on some stimulating relation between this application, which basically involves the control of the rotation of the hydrogen atom of the terminal SH bond around the PS axis, and complementary developments for laser pulse control of hydrogen transfer reactions, which have been carried out recently by our group [6].

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QUANTUM CONTROL OF REACTION DYNAMICS BY A LOCALLY OPTIMIZED CONTROL METHOD

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Quantum control consists of manipulating nuclear wave packets to a desired reaction channel by the use of tailored pulses. The main purpose of this paper is to present an application of a quantum control method developed in our group to simple chemical reaction dynamics[1,2]. The method is based on a local optimal control theory. The important point in our optimization theory is that the Schrödinger equation can be linearized in the short time limit. The optimization procedure is carried out in each short time stage to reach the desired final state. The present optimization is not limited to the control of reaction dynamics in weak intensity fields, but can be applicable to reaction system dynamics in strong intensity fields. We apply the control method to several types of reaction dynamics such as ring puckering isomerization, nonadiabatic reaction of NaI[3] and HCN isomerization. HCN is treated as an example of a 2-dimensional reaction. Nuclear wave packet propagations under controlled pulse conditions are analyzed, and mechanisms of quantum control are discussed. Finally, we show briefly that an enantiomer selective reaction can be controlled by our locally optimized control method[4].

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Optimal control theory - a tool to discover new reaction mechanisms ?

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The rapid development of ultrafast pulse shaping technologies gives hope to reach the goal of laser pulse control of chemical reactions. The combination of pulse shaping techniques and sophisticated genetic algorithms has shown first experimental success in the control of branching reactions, Ref.[1]. The laser fields used in Ref.[1] have a similar complicated structure as fields that have been predicted by optimal control theory (OCT) (Ref.[2]) for other purposes (Ref.[3],[4]). Using OCT allows an "ab initio" computation of laser fields not only for state selective transitions but also for the control of molecular properties expressed in terms of the expectation value of semi positive operators (Ref.[5],[6]). In our contribution we address the relevance of OCT on its own and also its impact on experiments. Starting from the experimental point of view, one could claim in the extreme that OCT is redundant, since the experimentally employed genetic algorithms are supposed to find efficient laser fields without any knowledge of a theoretically optimized field.

In order to demonstrate the power of OCT we present three examples:

- the photoisomerization of Li_2Na (Ref.[3]),
- the control of the collision pair Na^*-H_2 (Ref.[6]),
- and hydrogen transfer systems (Ref.[7]).

These examples show how OCT helps not only to find optimized pump-dump schemes and optimized frequency ranges for the laser pulses but also new reaction mechanisms. Especially the ability of OCT to discover optimal reaction mechanisms calls for high dimensional system descriptions. This requires a replacement of the reaction path by reaction surfaces and the use of parallel high performance computers allowing the treatment of high dimensional systems (Ref.8).

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Abstract for Quantum Control Workshop, Garching, Germany, July 1999

Phase coherence in non-linear optical excitation and condensed phase quantum dynamics

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The first part of the talk is based on a serious of papers in collaboration with Bardeen, Che, Yakovlav, and Wilson. The topic is intra-pulse phase-coherence in non-linear optical processes, The second part of the talk is based on my recent effort to formulate dissipative dynamics as an eigen-value problem. This formulation allows us to treat coherent excitation and optical spectroscopy in the condensed phase and in the gas phase on the same footing.

I. PHASE COHERENCE IN NON-LINEAR OPTICAL EXCITATION

The primary result reported here is the study of intra-pulse coherence in non-linear optical processes, which has become a novel approach in quantum coherence control. The goal of such control is to design laser pulses to drive a quantum system toward a specific target by means of phase coherence. This approach achieves selectivity in a chemical reaction by quantum mechanical interference and therefore differs fundamentally from conventional photo-excitation based on intensity or frequency modulations and from traditional chemical and mechanical manipulations. Much effort has been devoted to using pulse pairs and pulse sequences in creating and annihilating wave-packets. To explore the phase coherence within a single pulse, we have studied chirp effects in non-linear strong-field excitation processes. The following results demonstrate that chirping the pulse is a unique method in the phase control of population transfer, adiabatic inversion, wave-packet squeezing, and other quantum effects, which cannot be achieved otherwise.

1. Theoretical analysis[1] leads us to the intriguing conclusion that nearly complete electronic population inversion of molecules can be achieved with intense positively chirped broadband laser pulses (molecular π pulses), as a combined result of vibrational coherence and adiabatic inversion. Strong field quantum calculations demonstrate inversion probabilities of up to 99%. The results are robust with respect to changes in light field parameters as well as to thermal and condensed phase conditions, are supported by experimental evidence, and have many potential applications.

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2. Stationary and non-stationary three-level models[2] are formulated to analyze ultrafast two-photon processes with emphasis on electronic and nuclear coherence of a near-resonant intermediate state. By identifying and characterizing three intra-pulse dynamical mechanisms (sequential-resonance, chirp-following, and time-delay), a clear physical picture can be developed for understanding the subtle chirp dependence in intra-pulse pump-probe processes. This analysis provides a possible interpretation for a recent three-photon experiment.

II. KINETIC SPECTRAL ANALYSIS METHOD

The above theoretical models are developed for isolated gas phase molecules. For condensed phase systems, the predominant approach is to propagate the equation of motion from a given set of initial conditions. Numerous theoretical studies have contributed to the development and application of molecular dynamics methods, including Langevin dynamics, wave-packet propagation, and surface hopping. Simulation results are then fitted to kinetic models to demonstrate the underlying mechanism. Therefore, the interpretation of the kinetics is indirect and is sensitive to the choice of initial conditions and the length of trajectories. In addition, the accuracy of the propagation depends crucially on the time-step and deteriorates over long times. These problems are inherent in most molecular dynamic methods.

The goal of kinetic spectral analysis[3] is to develop a complementary approach, which is built on eigen-structures of dissipative systems instead of dynamic trajectories. For a large class of processes, the system of interest can be isolated and the condensed phase effect can be treated as dissipation. In this reduced description, the dissipative equation of motion defines an underlying eigen-structure, namely the kinetic spectrum, which dictates the dissipative dynamics and exhibits various kinetic behaviors on different time-scales. Within this unified framework, observed rate behavior, bi-exponential and multi-exponential decay, recurrences and oscillations are different components of the same kinetic spectrum; thus, existing kinetic and rate theories can be systematically analyzed, tested, and extended. Preliminary result along this line has been obtained for electron transfer in mixed-valence systems.

Just as the spectrum of the Shrödinger equation completely determines the dynamics of a quantum system, the eigen-structure of the reduced description completely determines the kinetic of a dissipative system. The approach provides not only a numerical alternative to dynamic simulations but also a different perspective on dissipative systems. For example, with the introduction of dissipative eigen-states, quantum control in gas phases and in condensed phases can be formulated on the same footing. In particular, relations can be established between photo-excitation and kinetic modes, which can be used as a guide for the design of laser pulses for selective photo-chemistry.

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Semiclassical Methods in Optimal Control and Detection of molecular dynamics

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We present some theoretical and computational considerations on the molecular dynamics in condensed phases in both the optimal control and the related transient absorption spectroscopic processes. We first demonstrate the similarity between the theoretical formulation of optimal control and that of the spectroscopic process. Consider a two-surface molecular system. In the control theory, the optimal field $E(t)$ that drives the molecular system to the pre-defined target $\hat{A}$ at the target time t_f is determined by [1]

$$K_+^*(t;t_f) + K_-(t;t_f) = \lambda E(t). \tag{1}$$

In this equation, K_+ and K_- are the so-called the pump and the dump control kernels, respectively. In the density matrix formulation, they are given by

$$K_\pm(t,t_f) = i\mathrm{Tr}\{\hat{A}\mathcal{G}(t_f,t)\rho_\pm(t)\}. \tag{2}$$

Here, $\rho_\pm(t) = D_\pm\rho(t)$, with $D_- = \mu|g\rangle\langle e|$ being the dipole-exciton annihilation operator, and $D_+ = D_-^\dagger = \mu^\dagger|e\rangle\langle g|$ the dipole-exciton creation operator. In eq. 1, $\mathcal{G}(\tau,t)$ is the Liouville-space (or density matrix) propagator in the presence the control field.

In comparison, the dispersed transient absorption coefficient signal in the presence of arbitrary excitation field can be expressed as [2]

$$\alpha(\omega,t_d) = -2\,\mathrm{Im}\int_0^\infty dt\, e^{-i\omega t}[R_+^*(t,t_d) + R_-(t,t_d)]. \tag{3}$$

Here, R_+ and R_- are respectively the spectroscopic pump (absorptive) and dump (stimulate-emitting) kernels given by

$$R_\pm(t,t_d) = i\mathrm{Tr}[D_\mp\mathcal{G}(t+t_d,t_d)\rho_\pm(t_d)]. \tag{4}$$

Note the similarity between the control and spectroscopic kernels (cf. eqs. 2 and 4).

The difficulty in the evaluation of these kernels stems from the cooperative influence from both external driving and dissipation. A generalised Fokker-Planck approach [3] being under development to this fundamental problem of quantum statistical dynamics will be briefly advertised.

The main focus of this presentation is on the weak response regime in which the dynamic kernels of both control and spectroscopy can be represented in terms of response functions that do not involve excitation field. For example, in the one-photon response regime, $K_- = 0$ and [1]

$$K_+^*(t;t_f) = \int_0^{t_f} d\tau\, M_H(t,\tau)E(\tau). \tag{5}$$

Here M_H is a Hermitian symmetrised version of following one-photon control response function [1].

$$M_e(t,\tau) = \mathrm{Tr}\{\hat{A}_e\mathcal{G}_{ee}(t_f-\tau)\mathcal{G}_{eg}\rho_g(T)]\}. \tag{6}$$

Similarly, the spectroscopic kernels R_+ and R_- can be expanded at the second order in the pump field, leading to the expressions that are characterised by the third-order nonlinear spectroscopic response functions [4]. The quasi-/semi-classical dynamics evaluation of both the control and spectroscopic response functions will be detailed together with the resulting optimal fields [5] and pump-probe transient absorption signals [6].

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Applications of Shaped Femtosecond Pulses in Condensed Phase Molecular Spectroscopy

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We describe the use of chirped laser pulses to control both vibrational coherences and population distributions of molecules in condensed phases. By tailoring the phase structure of low intensity pulses, the time-dependent shape of vibrational wavepackets can be controlled on both the electronic ground and excited states of a molecule. At higher pulse intensities, it becomes possible to control the amount of population transferred between the ground and excited states, leading to the possibility of a molecular π pulse. Applications of chirped pulses in the study of the dynamics of complex molecules like bacteriorhodopsin, and in biological fluorescence imaging, will be discussed.

Control of Chemical Reactions with Optimally Tailored Femtosecond Laser Pulses

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Microscopic control of chemical reactions, i.e., selective cleavage or formation of chemical bonds on a molecular level, is an old dream. The most promising way for this purpose is to specifically modify the electric field of a femtosecond laser pulse in order to control the reaction pathways. To solve the given problem it was suggested to directly include the reaction product yield in an optimization procedure [1] which allows a totally automated search for the best adapted laser pulse shape. An evolutionary computer algorithm which mimics the biological evolution („survival of the fittest") is applied to find an optimum in this high dimensional search space. The computer algorithm addresses a programmable femtosecond pulse shaper [2], uses the desired reaction output as feedback, and iteratively improves the applied laser field (Fig. 1). Thus the system itself solves its own Schrödinger equation in real-time without any prior knowledge on the system under the constraints set by the laboratory conditions. Some implementations have been demonstrated for automated femtosecond pulse compression [3,4] and optimized electronic population transfer in a dye molecule [5]. Our goal is to go beyond the manipulation and optimization of the electronic population transfer in some parent molecule.
First we demonstrate the applicability of the described method on the example of the photodissociation reaction of the $Fe(CO)_5$ molecule [6]. The product ratio $Fe(CO)_5^+/Fe^+$ was both maximized and minimized. The best adapted laser pulses have been transform limited and of a duration of several picoseconds, respectively.

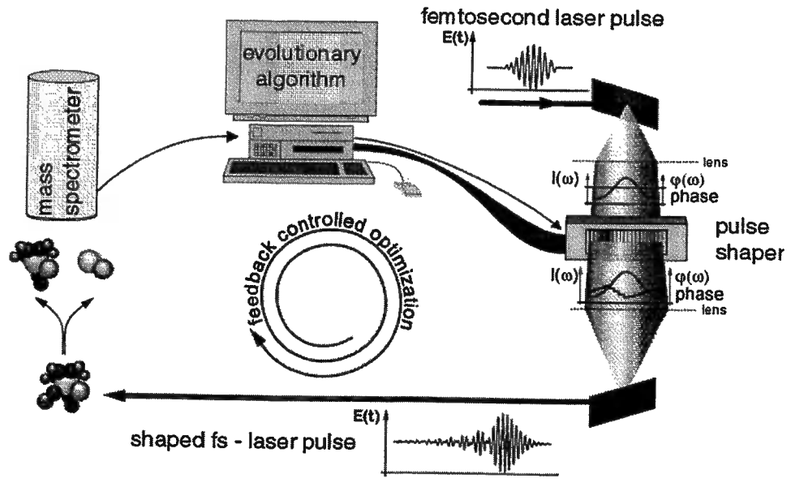

Fig. 1. Femtosecond laser pulses are modified by a computer-controlled pulse shaper. Ionic fragments from molecular photodissociation or ionization processes are recorded with a time-of-flight (TOF) mass spectrometer. This signal is used directly as feedback in the controlling evolutionary computer algorithm to optimize the desired photodissociation reaction.

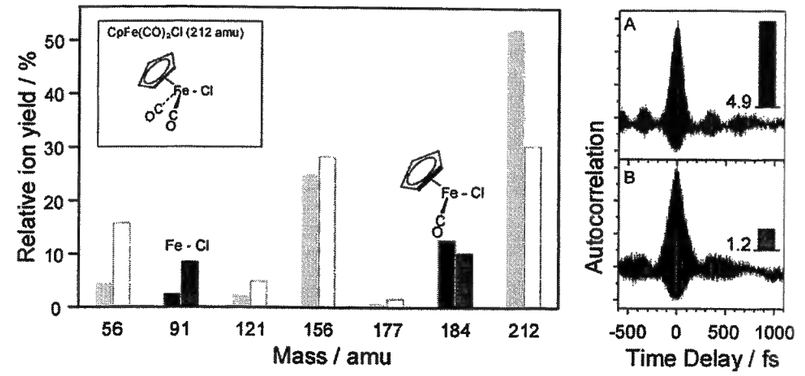

Fig. 2. Relative $CpFe(CO)_2Cl$ photodissociation product yields. Starting from the parent molecule, two different bond breaking reactions lead to chemically different final products. The branching ratio $CpFeCOCl^+/FeCl^+$ is maximized (solid blocks) as well as minimized (open blocks).

These results are in agreement with previous single femtosecond laser pulse experiments.
In addition to that we show that this coherent control scheme can also be applied to control chemical reactions where the photoproducts are chemically different from each other and from the parent molecule [7]. The branching ratio of the two photoproducts $CpFeCOCl^+$ and $FeCl^+$ of the dissociation reaction of $CpFe(CO)_2Cl$ was maximized and minimized and could thus be changed by a factor of 4 (Fig. 2). It should be noted that no information whatsoever about the sample substance is needed in the optimization procedure. The experiments reported here are an important step towards synthesizing chemical substances with higher efficiencies while at the same time reducing unwanted byproducts. The method can be applied to the liquid phase as well.
The optimally adapted laser fields generally show a very complex structure. An objective of our experiments is of course not only to optimize an experimental result, but to use the "optimized" phase information to derive a better understanding of the basic processes involved. In that respect an experiment with atomic calcium where we controlled the single and double ionization may serve as a model system. With the optimized fields the ionization pathways may be better understood.

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Highly localized vibronic wavepackets in large reactive molecules

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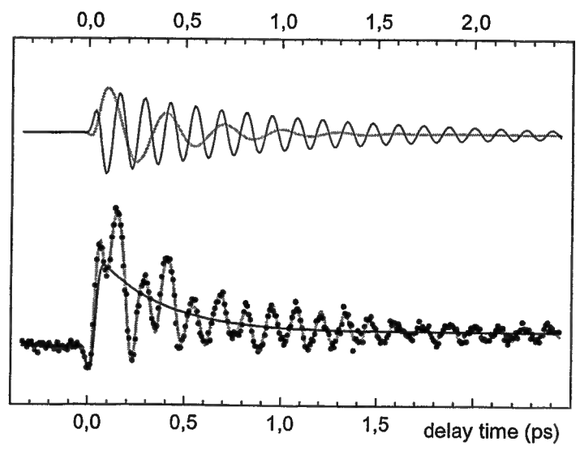
Coherent control of vibrational wavepacket motion towards a selected reaction goal requires persistent coherent motion of the molecule in a limited number of modes. Such coherent motion can readily be observed in small isolated molecules. Even large dyes molecules and a few systems of biological interest were recently found to display periodic oscillations of the transient absorption that are linked to normal modes.

For reactive systems the situation is much more complicated. The molecule changes its geometry on the way from educt to product significantly and as the molecular wavepacket moves away from the Franck-Condon point, the relevant topology of the potential energy surface varies. This leads to a change in normal mode frequencies and it is feared that the vibrational coherence is lost as a consequence.

We have monitored the evolution of vibronic wavepackets in two prototype molecules in solution with ultrahigh temporal resolution. Tunable 20 fs visible pump and probe pulses were produced in noncollinearly phase matched optical parametric amplifiers [1].

In azulene we found that the internal conversion from the S_1 state accelerates from 1.7 ps at the vibrationless S_1 state down to 400 fs at 1300 cm^{-1} excess energy [2]. Further increase up to 5500 cm^{-1} does not lead to any significant changes in the rate, however, strong oscillatory contributions to the signal are found. These correspond to vibrations at 160 cm^{-1} and 200 cm^{-1} that cannot be directly excited in the optically induced transition from the ground state.



The excited state proton transfer in 2-(2'-hydroxyphenyl)benzo-thiazole (HBT) is found to proceed in 60 fs and it is accompanied by extremely strong coherent wavepacket motion. This can be seen from the nearly 100 % signal modulation found in the typical experimental pump probe trace presented in the figure (lower part). A detailed analysis shows that the dominant contributions are vibrations at 110 cm^{-1} and 255 cm^{-1} (depicted in the upper part of the figure). Two other modes contribute with smaller amplitudes.

In azulene only 2 out of the 48 vibrational modes are found to oscillate coherently while in HBT there are only 4 out of 69 modes contributing to the coherent wavepacket motion. The vibrational dephasing time is typically between 0.5 and 1 ps. This high degree of localization of the vibronic wavepackets and the relatively slow dephasing is a strong indication that coherent control should be quite feasible in large molecules in solution. This is the class of systems that can undergo chemical changes of real interest and should therefore be a worthwhile objective in future experiments.

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"Ultrafast laser control of molecular vibrational dephasing"

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Experiments and calculations will be presented which show that the localized nature of vibrational dephasing allows successful laser control of energy flow in polyatomic molecules. Due to the properties of IVR, retardation of energy flow is a relatively robust and general process which can be applied in many degrees of freedom.

Quantum Control in Semiconductor Heterostructures

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Abstract

We investigate the possibility that phase and amplitude tailored, ultrafast laser pulses can be used to control photoexcited carriers in semiconductor heterostructures. Recent experiments have demonstrated the ability to create and observe electronic wave packets in multiple quantum well systems. These wave packets can be detected by a variety of means, including time- and position-resolved absorption. They also emit terahertz radiation, which provides a sensitive probe of the dynamics. In this work, we show that excitonic wave packets in superlattices can be controlled using simple, experimentally achievable femtosecond light pulses. In particular, we demonstrate that electronic wave packets can be localized in a desired well, at a desired time. Control of the terahertz radiation can also be attained, using a genetic algorithm to determine the optimal pulse parameters. Alternatively, assuming a fixed laser field, the parameters of the well can be optimized to produce a desired result. Finally, we investigate the effects of various dephasing mechanisms, and their implications for control.

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"The multimode quantum structure of coherent solitons"

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Ultrashort pulses propagate as solitons in optical fibers when dispersion and nonlinearity balance each other to form a wavepacket of remarkable stability. Optical solitons in fibers are an experimentally accessible model system for studying the nonlinear dynamics of a quantum field theory based on the quantum nonlinear Schrödinger equation [1]. In the context of fiber-optic communication, optical solitons are attractive for high-bitrate long-distance transmission systems [2] and for all-optical cascadable switching [3].

The quantum uncertainty of coherent solitons in fibers was observed to become "squeezed", i.e. to become reduced below the vacuum level in some field quadrature [4–7]. The quantum properties of optical solitons are particularly interesting for defining fundamental limits in high-precision measurements or in terabaud communication systems [8]. The quantum nature of light also gives rise to new opportunities in quantum information processing. Therefore, studying the quantum properties of optical solitons is an important field of research. The potential reward is the complete quantum control for the engineering of quantum states.

Spectral filtering of coherent solitons in optical fibers was found unexpectedly to produce "photon-number squeezed states" [5, 9], i.e. states with a photon-number uncertainty below the Poisson-limit of coherent states. This method of quantum noise reduction was experimentally optimized. It yielded up to 3.8±0.2 dB of photocurrent noise reduction below shotnoise in direct detection, from which a reduction of the photon-number uncertainty by 6.4 dB below the Poisson limit before detection can be inferred [10].

The mechanism for quantum noise reduction by spectral filtering can be understood in terms of the multimode quantum structure of solitons [11–14]. Spectral correlations play a crucial role, which was recently observed for the first time [13]. Although being stationary objects in a classical field theory, solitons develop a wealth of internal multimode quantum correlations as they propagate down the fiber. Spectrally resolved quantum noise measurements revealed modes anticorrelated in photon-number within the soliton spectrum [13]. If the anticorrelated modes are detected simultaneously and coherently, their fluctuations cancel each other. Thus a particularly "quiet" photocurrent far below the shot-noise limit is observed, similar to what was predicted for twin-beam correlations from fibers [15]. A bandpass filter that removes the outlying spectral sidebands of coherent fundamental solitons and transmits 82 % of the pulse energy was predicted to eliminate 77% of the photon-number uncertainty (6.5 dB of photon-number noise reduction below the Poisson limit) [16]. Up to 8.1 dB of quantum noise reduction were predicted for pulses of more than the fundamental soliton energy [17].

There are several promising directions for further research in this field: firstly, higher-order solitons show strong correlations and enhanced nonclassical effects [18, 19]. Secondly, the internal quantum structure of interacting coherent solitons shows interesting correlations for quantum entanglement and quantum nondemolition measurements [18]. Thirdly, the squeezing mechanism of a nonlinear fiber interferometer, which produces highly nonclassical solitons, will be better understood with spectrally resolved quantum measurements. Quantum entanglement may also be generated by interference of two amplitude-squeezed states from such fiber interferometers [20]. Furthermore, the multimode quantum analysis of Raman-self-frequency-shifted solitons gives insight into an unexpected Raman-assisted squeezing mechanism [10]. Finally, photon-number noise reduction by filtering is a promising perspective for soliton communication systems because spectral filters will be employed for the reduction of the timing jitter and of dispersive waves [21].

The method of spectral filtering is in the process of unraveling a field of multimode quantum optics with the potential to synthesize and to control nonclassical states of light in a new way.

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History of the Ringberg Castle

Historical Background

Schloß Ringberg, overlooking the Tegernsee in the foothills of the Bavarian Alps, is a monument commemorating the idiosyncrasy, originality, and remarkable single-mindedness of two men: Duke Luitpold in Bavaria (Herzog Luitpold in Bayern), a member of the Wittelsbach family who ruled Bavaria over 800 years, and his friend, the alround artist, architect and interior decorator, Friedrich Attenhuber. The castle is entirely their creation, from the massive Renaissance-inspired exterior, right down to the fittings and furniture which, in every detail, were designed by Attenhuber himself and executed by native craftsmen. Attenhuber also painted every single picture hanging in the castle. He found his models in the farmhouses around the Tegernsee. The castle embodies all trends of art and styles which dominated the first half of this century, combined with local Alpine originality and the individual creative power of its constructors.

Schloß Ringberg dominated the lives of the two men to the point of becoming an obsession. They had met in Munich, while Duke Luitpold was studying philosophy and history of art at the Ludwig-Maximilian University (1910 - 1914). The idea of building a castle to realise an architectural dream and erect a new family seat probably came to them whilst travelling through Europe between 1910 and 1912. The location of the castle above the Tegernsee is said to have been decided upon one day in 1911, when Duke Luitpold was out hunting in the area with his cousin. A painting by Attenhuber in the entrance hall of the castle shows this scene. From 1913 onwards, work at the castle was personally supervised by Friedrich Attenhuber, who subsequently lived and finally died there in 1947. After his death, Duke Luitpold continued to devote all his energies to the construction of the castle, selling family estates to pay for the building and travelling the more than 50 kilometres from his home in the Hotel Vier Jahreszeiten in Munich to supervise the progress of the work. He never actually lived at the castle but preferred to lodge at the Hotel Bachmair when he had to stay overnight. When he died in 1973 at the age of 82, he left Schloß Ringberg unfinished.

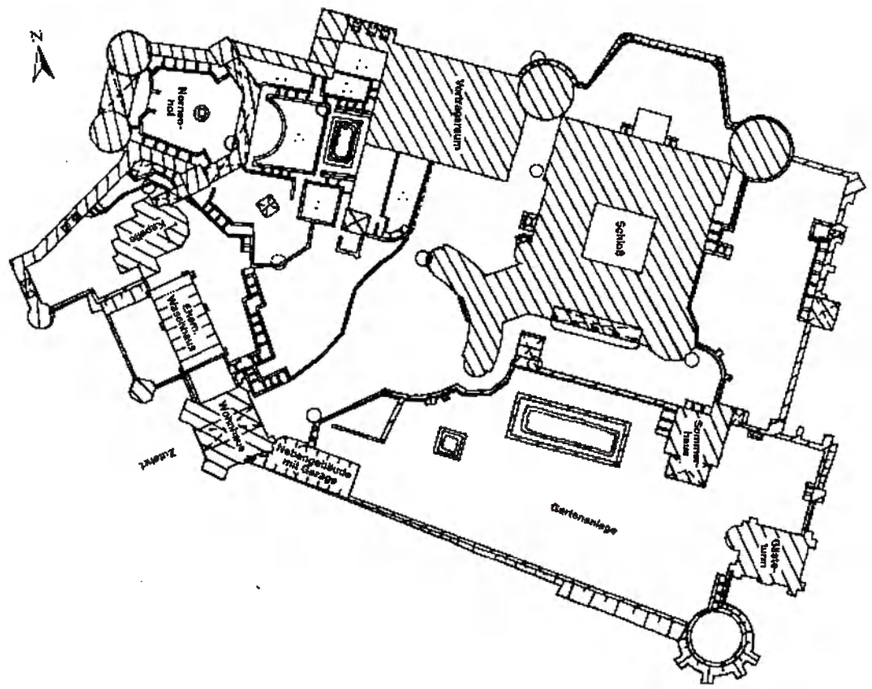
The House of Wittelsbach's Love for Architecture

Duke Luitpold was certainly not the only member of the Wittelsbach family to pursue architectural passions and ambitions. The city of Munich, capital of Bavaria, owes much of her distinctive atmosphere to the monumental buildings erected by King Ludwig I, great admirer of the classical period, in the last century. Ludwig II, the "fairy-tale king" and friend and patron of Richard Wagner, built the famous romantic castles of Neuschwanstein and Linderhof in the Bavarian Alps, and Schloß Herrenchiemsee situated on a small island in the Chiemsee, realising his vision of Versailles. Thus inspired by his ancestors, Duke Luitpold made his own dreams come true. The site he chose to erect his castle is one of great splendour. To the north stretches the Tegernsee, to the south the valley of Kreuth, while the south-east the Wallberg and Setzberg mountains tower the horizon.

Schloß Ringberg and the Max-Planck-Gesellschaft

During the mid-sixties, Duke Luitpold began to consider what should become of Schloß Ringberg after his death, since he had no direct heir. He requested the Bavarian authorities to grant his castle the status of a historical monument as a way of avoiding heavy tax duties, but his application was rejected by the Miesbach finance authorities. Thus he was confronted with the choice of either selling Schloß Ringberg to the German Federal Organisation of Trade Unions, or making a present of it to the Max-Planck-Gesellschaft.Fortunately, he chose the latter. The contract was signed in 1967 and the castle passed into the hands of the Max-Planck-Gesellschaft after the Duke's death in 1973. At first, the Max-Planck-Gesellschaft was not too enthusiastic in accepting the inheritance. It was clear that modifications of the castle for the purpose of serving the sciences, as was the will of the testator, would necessitate considerable financial expenditure. The problem was solved in that provision was made in the Duke's last will to leave also his other property to the Max-Planck-Gesellschaft for exclusive use in the preservation of the castle. It is thanks to the foresight and engagement of the former president of the Max-Planck-Gesellschaft, Prof. Dr. Reimar Lüst, that Schloß Ringberg has been transformed into a meeting place where scientists can exchange ideas and discuss problems with their colleagues from all over the worls in beautiful surroundings and in a relaxed mountain atmosphere high above the daily business activities. At the beginning of this era, the castle could accommodate 12 guests but nowadays, thanks to a generous donation, 33 guest beds are available. the centre of communication is nowadays well accepted by the scientific community and generally fully booked ahead for more than a year.

As in former times, people around the Tegernsee are highly interested in what is going on at Schloß Ringberg. Tourists are informed as follows: "In the castle there is now the Max-Planck-Gesellschaft to investigate the conditions of recreation in Bavaria".



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