Symposium Quantenkontrolle funktionaler Moleküle (SYFM)

gemeinsam veranstaltet von
Fachverband MO (Molekülphysik)
Fachverband Q (Quantenoptik und Photonik)

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Moleküle sind die kleinsten funktionalen Einheiten in weiten Bereichen der Biologie, Chemie und Physik. Gleichzeitig sind sie hochkomplexe Quantensysteme mit herausragendem Anwendungspotential zum Beispiel in der Informationstechnologie, für nanoskalige Maschinen oder molekulare Elektronik. Mit Ihrer Vielzahl von Freiheitsgraden der Elektronen- und Kernbewegung bieten sie die Möglichkeit, durch optische Anregung ihre eigenen Eigenschaften gezielt zu aktivieren und als Teil komplexer Anordnungen das globale Verhalten von Supramolekülen und makroskopischen Materialien zu steuern. Die oft nicht ausreichende Effizienz der dynamischen Prozesse kann durch gezielt modifizierte Lichtfelder ultrakurzer Dauer optimiert werden. Damit wird eine Quantenkontrolle auf der Zeitskala der intramolekularen Bewegungen möglich. Im Symposium soll der Fortschritt in diesem hochaktuellen Gebiet dargestellt werden.

Übersicht der Hauptvorträge und Fachsitzungen

(Hörsaal 6C)

Hauptvorträge

SYFM 1.1	Mi	14:00–14:30	6C	**Control strategies for molecular switches in donor-bridge-acceptor systems** — •Regina de Vivie-Riedle, Dorothee Geppert
SYFM 1.2	Mi	14:30–15:00	6C	**Ultrafast ring-opening and ring-closure reaction of photochromic molecular switches - indolylfulgides** — •Markus Braun, Simone Draxler, Thomas Brust, Stephan Malkmus, Florian Koller, Björn Heinz
SYFM 1.3	Mi	15:00–15:30	6C	**Mode-selective control in complex molecules with shaped femtosecond pulses** — •Marcus Motzkus, Tiago Buckup, Jürgen Hauer, Carles Serrat
SYFM 1.4	Mi	15:30–16:00	6C	**Quantum control of liquid-phase molecular dynamics and of catalytic surface reactions** — •Patrick Nürnberger, Daniel Wolpert, Horst Weiss, Gustav Gerber
SYFM 2.1	Mi	16:30–17:00	6C	**Photochromic systems and *moving* molecules and nano-objects** — •Luisa De Cola
SYFM 2.2	Mi	17:00–17:30	6C	**Mechanisms of electron induced switching at interfaces** — •Martin Wolf
SYFM 2.3	Mi	17:30–18:00	6C	**Physical mechanism of quantum control: ultrafast, robust and efficient by tailored intense resonant femtosecond laser pulses** — •Matthias Wollenhaupt, Tim Bayer, Cristian Sarpe-Tudoran, Thomas Baumert
SYFM 2.4	Mi	18:00–18:30	6C	**Quantum control of electric ring currents by circularly polarized laser pulses** — •Jörn Manz, Ingo Barth

Fachsitzungen

SYFM 1.1–1.4	Mi	14:00–16:00	6C	**Quantum control of functional molecules I**
SYFM 2.1–2.4	Mi	16:30–18:30	6C	**Quantum control of functional molecules II**

SYFM 1: Quantum control of functional molecules I

Zeit: Mittwoch 14:00–16:00 Raum: 6C

Hauptvortrag SYFM 1.1 Mi 14:00 6C

Control strategies for molecular switches in donor-bridge-acceptor systems — •Regina de Vivie-Riedle and Dorothee Geppert — Department Chemie, Universität München, Butenandtstr. 11, 81377 München, Germany

Bistable fulgides are promising candidates for optical molecular switches and are already tested as switchable bridge molecules in donor-bridge-acceptor (DBA) systems. The transition between the two stable isomers can be induced by laser light and is then mediated via conical intersections. Only one isomeric form allows energy transfer from D to A. Our aim is to formulate realistic strategies based on ab initio data to optimize the optical On/Off switching process. We perform quantum chemical calculations for the DBA moiety and detailed quantum dynamical studies for the switching process. Different control strategies for the switching process including intermediate targets on the excited state as well as ground state targets are developed. The optimized laser fields are found applying optimal control theory and are subsequently analyzed to enlighten the underlying control mechanisms. A decomposition extracts the leading field components and enables a simplified reconstruction by Gaussian laser pulses [1]. Our studies show that laser control of chemical relevant reactions is within reach for experimental realization with high quantum yield and opens the way to design functional molecular devices.

[1] D. Geppert and R. de Vivie-Riedle, J. Photochem. Photobiol. A 180 (2006) 282-288.

Hauptvortrag SYFM 1.2 Mi 14:30 6C

Ultrafast ring-opening and ring-closure reaction of photochromic molecular switches - indolylfulgides — •Markus Braun, Simone Draxler, Thomas Brust, Stephan Malkmus, Florian Koller, and Björn Heinz — Lehrstuhl für BioMolekulare Optik, Ludwig-Maximilians-Universität München, Oettingenstr. 67, 80538 München

Photochromic molecules are characterized by a reversible rearrangement of their structure (isomerization) due to light illumination, connected with changes in their color (probed by e.g. absorption in the UV/VIS and mid-IR or fluorescence). Indolyl substituted fulgides and fulgimides are known for their thermally stable and photochemically robust isomers (C, E and Z), which can be addressed optically by well-separated absorption bands. The control of the structural dynamics and reaction yield of the pericyclic isomerization is investigated by steady-state and fs-time resolved pump-probe spectroscopy. These experiments show that isomerization occurs within a few picoseconds or even faster. Photochemical parameters as temperature, polarity or optical excitation conditions allow for an optimization of the reaction yields. Application of optical prepulses influences the excited state dynamics and the isomerization yield.

Hauptvortrag SYFM 1.3 Mi 15:00 6C

Mode-selective control in complex molecules with shaped femtosecond pulses — •Marcus Motzkus[1], Tiago Buckup[1], Jürgen Hauer[1], and Carles Serrat[2] — [1]Physikalische Chemie, Philipps-Universität Marburg, D-35032 Marburg, Germany — [2]Departament de Fisica, Universitat Politecnica de Catalunya, 09822 Terrassa, Spain

Coherent control of vibrational modes is essential for manipulating photochemistry on ground and excites states since such oscillations may represent motion along the desired reaction coordinate. One well established mechanism for optimizing vibrations in time domain exploits pulse sequences with temporal spacing between the sub pulses equal to an integer multiple of the vibrational period. To explore the underlying mechanism it is important to understand if an optimally shaped laser pulse can overcome the limitations defined by Fourier-limited laser pulse of equal laser energy, i.e. the shaped pulse does not act merely as a filter but actual enhancement of population and coherence is obtained. We show that excitation with electronically resonant or far off-resonant laser pulse spectra cause no enhancement, while with near resonant excitation (blue or red shifted) the vibrational modes can be amplified by almost a factor of 2. A four-level density matrix simulation supports the experimental findings and underlines the crucial role of electronic resonances for effective mode-control.

Hauptvortrag SYFM 1.4 Mi 15:30 6C

Quantum control of liquid-phase molecular dynamics and of catalytic surface reactions — •Patrick Nürnberger[1], Daniel Wolpert[1], Horst Weiss[2], and Gustav Gerber[1] — [1]Physikalisches Institut, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]BASF AG, Polymer Research Division, 67056 Ludwigshafen, Germany

We present two concepts for the selective transfer of one molecular configuration into another one with femtosecond laser pulses. By selective excitation with optimized laser pulses, the photoisomerization efficiency of a molecular system can be controlled. The product yield can also be manipulated by selective de-excitation of an excited molecule with shaped laser pulses.

Furthermore, we demonstrate the formation of molecular bonds with femtosecond laser pulses. Hydrogen and carbon monoxide are streamed onto a palladium surface irradiated with the laser pulses. A time-of-flight mass spectrometer allows the detection of created product molecules. By applying a closed-loop quantum control scheme, we manipulate this formation of molecular bonds and selectively optimize the ratio of different reaction channels with specially adapted laser pulses.

SYFM 2: Quantum control of functional molecules II

Zeit: Mittwoch 16:30–18:30 Raum: 6C

Hauptvortrag SYFM 2.1 Mi 16:30 6C

Photochromic systems and *moving* molecules and nano-objects — •Luisa De Cola — Westfälische Wilhelms-Universität, Physikalisches Institut and CeNTech, Mendelstr. 7, 48149 Münster, Germany

In this contribution I will discuss the use of the photochromic behavior of metal complexes. In particular the possibility to switch on/off photoinduced processes (e.g., energy transfer) controlling the state of the photochromic unit will be illustrated [1]. The talk will then focus on the use of mesoporous materials for the entrapping of dyes and their release upon photoisomerization of azobenzene derivatives linked to the pores [2]. Finally it will be shown how zeolite L, biocompatible nano- microcontainers, upon functionalization can be non-covalently linked to living organisms [3]. The filled zeolite, with fluorescent dyes, will be used to monitor the motion of bacteria and their uptake by cells will be discussed.

[1] for a recent review see: P. Belser, L. De Cola, et al., Adv Funct Mat 2006, 16, 195.

[2] P. Sierocki, H. P. A. Maas, P. Dragut, G. Richardt, F. Vögtle, L. De Cola, F. A. M. Brouwer, J. I. Zink, J.Phys.Chem. B 2006, 110, 24390 and paper submitted.

[3] Z. Popovic, M. Otter, G. Calzaferri, S. Huber, L. De Cola, submitted

Hauptvortrag SYFM 2.2 Mi 17:00 6C

Mechanisms of electron induced switching at interfaces — •Martin Wolf — Fachbereich Physik, Freie Universität Berlin, 14195 Berlin

The possibility to design molecules with dedicated functional properties opens the perspective to use molecules as building blocks for integrated functional devices. However, for applications like in molecular electronics it will be essential to control the switching between different molecular states and to connect the molecular system with the outside world by contacts with electrodes. This requires the synthesis and design of appropriate nanosystems and a basic understanding of structural and electronic properties including the interaction with solid interfaces. In addition, one would like to achieve active control of the switching between different molecular conformations by external stimuli like electromagnetic fields, forces and currents. In this talk I will present the approach of the collaborative research center Sfb 658 "Elementary processes in molecular switches at surfaces" to this problem and discuss our current understanding using several examples for (reversible) conformational switching processes at surfaces induced by charge transfer, electrical fields, currents and light probed with scanning tunnelling microscopy and photoemission spectroscopy.

Hauptvortrag SYFM 2.3 Mi 17:30 6C

Physical mechanism of quantum control: ultrafast, robust and efficient by tailored intense resonant femtosecond laser pulses — •Matthias Wollenhaupt, Tim Bayer, Cristian Sarpe-Tudoran, and Thomas Baumert — Universität Kassel, Institut für Physik und CINSaT, Heinrich-Plett-Str. 40, D-34132 Kassel, Germany

The success of optimal control strategies in controlling chemical reaction dynamics with tailored light pulses is contrasted by our limited understanding of the underlying physical processes. However, new perspectives for quantum control of functional molecules open up due to the uncommon molecular dynamics induced by shaped resonant intense pulses. In this contribution the physical mechanism of quantum control using resonant ultrashort optimal pulses is investigated on model systems (atoms and small molecules [1,2]). Switching among different final electronic states is realized by Selective Population of Dressed States (SPODS). In the experiment high selectivity of the dressed state population (more than 90%) is demonstrated. Because we achieve tunability of dressed state energies in the range of several hundred meV, our approach is attractive for control of functional molecules. SPODS proceeds within a few optical cycles. Therefore our strategy might be operative in the presence of decoherence processes as well. Our approach is general, because resonant control scenarios - as demonstrated here - will become increasingly important as shorter and shorter pulses with ultra broad spectra are available.

[1] M. Wollenhaupt et al., Chem Phys Lett 419, 184(2006).

[2] M. Wollenhaupt and T. Baumert, J Photochem Photobiol A 180, 248 (2006).

Hauptvortrag SYFM 2.4 Mi 18:00 6C

Quantum control of electric ring currents by circularly polarized laser pulses — •Jörn Manz and Ingo Barth — Freie Universität Berlin, Institut für Chemie und Biochemie, Takustr. 3, 14195 Berlin

Few cycle laser pulses with durations from several hundred as to few fs may induce electronic excitations of valence electrons in oriented molecules, atoms, or ions. In particular, circularly polarized laser pulses may induce electron circulation or electric ring currents and induced magnetic fields in ring-shaped molecules such as Mg-porhpyrin, around linear molecules such as AlCl, or atoms. These ring currents are more than hundred times stronger than traditional ones induced by permanent magnetic fields. Moreover, the laser pulses allow active control, i.e. they may serve as "traffic light"for electronic pathways along alternative molecular bonds in ring-shaped molecules. Furthermore, analytic expressions derived for the non-relativistic H-atom and for one-electron ions show that electrons in $2p_+$ or $2p_-$ atomic orbitals have the strongest electric ring currents and the strongest magnetic fields. The LCAO-MO method can be applied in order to estimate approximate values of the ring currents and induced magnetic fields in molecules, based on the analytical formulas for atomic orbitals.

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