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.