MO 17: Quantum Control

Time: Wednesday 16:30–18:15 Location: TOE 317

MO 17.1 Wed 16:30 TOE 317

Direct mid-infrared femtosecond pulse shaping with a calomel acousto-optic programmable dispersive filter — •Patrick Nuernberger[2,4], Raman Maksimenka[1], Kevin F. Lee[2], Adeline Bonvalet[2], Thibault Vieille[1,2], Cestmir Barta[3], Miloš Klima[3], Thomas Oksenhendler[1], Pierre Tournois[1], Daniel Kaplan[1], and Manuel Joffre[2] — [1]FASTLITE, Centre scientifique d'Orsay - Bât. 503, 91401 Orsay, France — [2]Laboratoire d'Optique et Biosciences, Ecole Polytechnique, CNRS UMR 7645, INSERM U696, 91128 Palaiseau, France — [3]BBT Materials Processing, Doubicka 11, 184 00 Prague, Czech Republic — [4]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

Direct amplitude and phase shaping of mid-infrared femtosecond pulses is realized with a calomel-based acousto-optic programmable dispersive filter transparent between 0.4 and 20 μm. The shaped pulse electric field is fully characterized with high accuracy, using chirped-pulse upconversion and time-encoded arrangement spectral phase interferometry for direct electric field reconstruction techniques. Complex mid-infrared pulse shapes at a center wavelength of 4.9 μm are generated with a spectral resolution exceeding by more than a factor of 5 the reported experimental resolutions of calomel-based acousto-optic filters.

MO 17.2 Wed 16:45 TOE 317

Femtosecond Pulse-Shaping and Characterization in the Mid-Infrared — •Rene Costard, Christian Greve, Erik T. J. Nibbering, and Thomas Elsaesser — Max Born Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max Born Strasse 2A, D-12489 Berlin, Germany

Femtosecond mid-infrared (IR) pulses are now commonly used for nonlinear time-resolved vibrational spectroscopy. Pump-probe or multidimensional photon echo experiments of transient vibrational excitations allow for elucidation of anharmonic couplings and vibrational energy flow pathways. So far, these experiments have typically been performed using the output of a parametric frequency converter, with the central frequency of the IR pulses as experimental parameter. Controlling the amplitude and phase of these pulses, however, allow for a full coherent control of vibrational excitations in molecular systems. We present experimental results of amplitude and phase shaping of ultrashort pulses around 3 μm, which are generated by taking the idler output of an optical parametric amplifier using KTP, pumped by a Ti:sapphire chirped-pulse amplification system. Directing these pulses through a 4f setup with a germanium acousto-optic modulator in the Fourier plane, enables independent shaping of amplitude and phase to generate e.g. double pulses with adjustable time separation or arbitrarily chirped pulses. We fully characterize the amplitude and phase of these shaped mid-IR pulses by cross-correlation frequency-resolved optical gating (XFROG) with well-characterized 800 nm pulses.

MO 17.3 Wed 17:00 TOE 317

The von Neumann representation as a parameterization for polarization-shaped laser pulses — •Stefan Ruetzel[1], Anja Krischke[1], Tobias Brixner[1], and David J. Tannor[2] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — [2]Department of Chemical Physics, Weizmann Institute of Science, 76190 Rehovot, Israel

Polarization-shaped laser pulses offer a wide range of applications in femtosecond spectroscopy and coherent control. The description of such laser pulses is generally provided in time or frequency domain. Time–frequency descriptions descriptions were shown to be useful in the past but have been limited to linearly polarized fields. Here we introduce the von Neumann description as a parameterization for polarization-shaped laser pulses. The electric field is expanded in terms of Gaussian-shaped transform-limited subpulses located on a discrete time–frequency lattice, each with a specific polarization state. On the one hand this formalism can serve as a new description of polarization-shaped laser pulses, simplifying the interpretation of the time- and frequency-dependent polarization state of the light field. On the other hand the polarization state of the laser pulse can directly be defined in the joint time–frequency domain, which allows for an intuitive parameterization of such laser pulses with a reduced number of variables compared to common laser pulse descriptions. Possible applications for future experiments will be presented.

MO 17.4 Wed 17:15 TOE 317

Resonant Strong-Field Control of Electron Dynamics in K_2 — Tim Bayer, •Hendrike Braun, Cristian Sarpe, Matthias Wollenhaupt, and Thomas Baumert — University of Kassel, Institute of Physics and CINSaT, D-34132 Kassel, Germany

The strong-field control mechanism SPODS (Selective Population of Dressed States) has been demonstrated on atoms by using variously shaped femtosecond laserpulses [1,2,3]. The applicability of SPODS to the coherent control of molecules and chemical reactions was pointed out by wave packet calculations on K_2 [4]. Here, we present an experimental demonstration of the molecular strong-field control scheme proposed in [4]. Employing pulse sequences from sinusoidal and blurred step phase modulation respectively, we steer the molecular system, i.e. nuclear wave packet, from a well-defined ground state *via* a transient state of maximum electronic coherence towards a preselected target state. By exerting control on the dressed states of a resonant subsystem we are able to selectively adress different target channels within a manifold of final states with high efficiency. That is, the control of electronic coherences gives rise to effective manipulation of nucelar coherences. Since dressed state splittings in the order of several 100 meV are readily achieved experimentally, the devised SPODS control scheme offers prospect to various applications in femtochemistry.

[1] M. Wollenhaupt *et al.*, Phys. Rev. A **68**, 0154011 (2003)
[2] M. Wollenhaupt *et al.*, Phys. Rev. A **73**, 063409-1 (2006)
[3] T. Bayer *et al.*, Phys. Rev. Lett. **102**, 023004 (2009)
[4] M. Wollenhaupt *et al.*, JPPA **180**, 248 (2006)

MO 17.5 Wed 17:30 TOE 317

Product control of conical intersection driven photochemical reactions by steering electronic wavepackets — •Philipp von den Hoff and Regina de Vivie-Riedle — Department Chemie, Ludwig-Maximilians-Universität München, D-81377 München, Germany

Electrons and their dynamics are involved in bond breaking and formation, thus the idea to steer chemical reactions by localization of electronic wavepackets seems natural. The formation of a localized electronic wavepacket requires the superposition of two or more appropriate electronic states through e.g. an external electric field. The guiding of such an electronic wavepacket is only possible within the coherence time of the system. Here, we present a new UV-pump-IR-control scheme that allows us to control the product ratio of conical intersection driven photochemical reactions by steering an electronic wavepacket [1]. To test the proposed scheme, we constructed a two dimensional model system. Our calculations show, that we are able to steer the final product ratio very precisely by changing the carrier envelope phase of the control IR-pulse.

[1] P. von den Hoff, R. Siemering and R. de Vivie-Riedle, Ultrafast Phenomena XVII, Oxford University Press, (2010), in Press.

MO 17.6 Wed 17:45 TOE 317

Efficient and robust strong-field control of populationtransfer in sensitizer dyes with designed femtosecond laser pulses — Johannes Schneider, •Matthias Wollenhaupt, Andreas Winzenburg, Tim Bayer, Jens Köhler, Rüdiger Faust, and Thomas Baumert — University of Kassel, Institute of Physics and CINSaT, D-34132 Kassel, Germany

We demonstrate control of electronic population transfer in molecules with the help of shaped femtosecond laser pulses. To that end we investigate two photosensitizer dyes in solution being prepared in the triplet ground state. Excitation within the triplet system is followed by intersystem crossing and the corresponding singlet fluorescence is monitored as a measure of population transfer in the triplet system. We record control landscapes with respect to the fluorescence intensity on both dyes by a systematic variation of laser pulse shapes combining second order and third order dispersion. In the strong-field regime we find highly structured topologies with large areas of maximum or minimum population transfer being robust with respect to the applied laser intensities. We then compare our experimental results with simulations on generic molecular potentials by solving the TDSE for

excitation with shaped pulses. The analysis of the regions of maximum or minimum population transfer reveals that coherent processes control the outcome of the excitation process. The physical mechanisms of joint motion of ground and excited state wave packets or population of an vibrational eigenstate in the excited state permits us to discuss the molecular dynamics in an atom-like picture.

MO 17.7 Wed 18:00 TOE 317

Prospects of Incoherent Control by Continuous Measurements — •Felix Platzer and Klaus Hornberger — Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Straße 38, 01187 Dresden, Germany

In contrast to state-selective coherent control, which proves to be fragile when applied to large quantum systems such as poly-atomic molecules, incoherent dynamics induced by continuous measurements can provide a more robust means of steering quantum dynamics towards desired target states. Here, we demonstrate the potential use of non-selective measurements of position or, more generally, of configurational degrees of freedom of molecules, for the optimization of transition probabilities. Numerical simulations of wave packet dynamics in the presence of optimized position measurements are presented and their analytical description in terms of the corresponding master equation undertaken.