MO 24: Poster: Quantum Control

Time: Thursday 16:00–18:30 Location: Lichthof

MO 24.1 Th 16:00 Lichthof

Infrared Laser Driven Double Proton Transfer. An Optimal Control Theory Study — •Oliver Kühn and Mahmoud Abdel-Latif — Institut für Physik, Universität Rostock, D-18051 Rostock

Laser control of ultrafast double proton transfer is investigated for a two-dimensional model system describing stepwise and concerted transfer pathways [1]. The pulse design has been done by employing optimal control theory in combination with the multiconfiguration time-dependent Hartree wave packet propagation. The obtained laser fields correspond to multiple pump-dump pulse sequences. Special emphasis is paid to the relative importance of stepwise and concerted transfer pathways for the driven wave packet and its dependence on the parameters of the model Hamiltonian as well as on the propagation time. While stepwise transfer is dominating in all cases considered, for high barrier systems concerted transfer proceeding via tunneling can make a contribution.

[1] M. Abdel-Latif, O. Kühn, arXiv:0912.1715v1 [physics.chem-ph]

MO 24.2 Th 16:00 Lichthof

Open-loop and closed-loop quantum control in von Neumann time-frequency phase space — •Stefan Ruetzel[1], Christoph Stolzenberger[1], Frank Dimler[1], Susanne Fechner[1], David J. Tannor[2], and Tobias Brixner[1] — [1]Institut für Physikalische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Department of Chemical Physics, Weizmann Institute of Science, 76190 Rehovot, Israel

The parameterization of shaped laser pulses plays a decisive role in the outcome of quantum control experiments. Depending on the system under investigation and the control target, the experimentalist has to choose an appropriate parameterization to reveal possible control mechanisms.

We have introduced the von Neumann representation, a joint time-frequency description of shaped femtosecond laser pulses and have proposed a large variety of applications. Here we present time-frequency phase-space scanning procedures of shaped femtosecond laser pulses in the von Neumann formalism that can be applied to measure multidimensional molecular control landscapes. This technique has been utilized to analyze the intrapulse pump-dump mechanism of a laser dye in solution.

On the other hand, the von Neumann basis can be implemented into an evolutionary algorithm for closed-loop quantum control experiments. We show simulated as well as experimental results that prove the advantage of this laser pulse basis over conventional parameterizations.

MO 24.3 Th 16:00 Lichthof

Generation of shaped UV pulses for quantum control spectroscopy — •Jens Möhring[1], Tiago Buckup[1,2], and Marcus Motzkus[1,2] — [1]Physikalische Chemie, Philipps-Universität, D-35043 Marburg, Germany — [2]Physikalisch-Chemisches Institut, Ruprecht-Karls-Universität Heidelberg, D-69120 Heidelberg, Germany

The availability of phase and amplitude modulated UV pulses enables the extension of coherent control experiments to new molecular systems absorbing in the UV spectral region. Especially small prototype systems often show only UV absorptions bands and can therefore be studied by quantum control spectroscopy only if shaped UV excitation pulses are available. Here we show an improved setup for generation of phase and amplitude controlled femtosecond UV pulses and their characterization [1]. Central component is a micromechanical (MEMS) mirror 2D phase modulator for ultrashort optical pulse control. A duty cycle upgrade (333 Hz) of the spatial light modulator improves its application in spectroscopic experiments. The application of diffractive 2D shaping removes parasitic amplitude effects of the MEMS shaper and enables on the other hand the introduction of amplitude control on this phase only device. The setup is driven by a two stage non-collinear optical parametric amplifier with sum frequency generation and temporal compression in the UV, yielding a tunable source of 30 fs pulses. Altogether a versatile setup for quantum control spectroscopy in the UV is presented.

[1] Möhring, J. et al., JOSA B 26 (2009) 1538.

MO 24.4 Th 16:00 Lichthof

Combining fs pulse tailoring and self-phase modulation for nonlinear microscopy — •Tillmann Kalas, Jens Köhler, Cristian Sarpe-Tudoran, Matthias Wollenhaupt, and Thomas Baumert — University of Kassel, Institute of Physics and Center of Interdisciplinary Nanostructure Science and Technology (CINSaT), D-34132 Kassel, Germany

Nonlinear label-free microscopy is a powerful tool for the investigation of physical and biological samples with high spatial resolution. Often intrinsic Second- or Third-Harmonic Generation as well as Coherent Anti-Stokes Raman Scattering is used as contrast mechanism.
We make use of fs pulse shaping in combination with self-phase modulation (SPM) in order to generate the nonlinear signals [1, 2]. Extending our previous studies [1], fs laser pulses are amplitude and phase modulated in a narrow spectral interval and focused into transparent samples. SPM leads to a redistribution of the power spectral density (PSD) depending on the nonlinear index of refraction. In particular the intensity of previously removed spectral components is recovered. Hence, observation of these intensities holds the possibility to distinguish between different materials. We demonstrate high nonlinear contrast within index matched photonic fibers combining the fs pulse shaping technique with a commercial laser-scanning-microscope. Moreover, the influence of additional spectral phases on the self-phase modulated PSD is studied and results are given.

[1] A. Präkelt *et al.*: Appl. Phys. Lett. **87**(12), 121113(2005)
[2] M. C. Fischer *et al.*: Opt. Lett. **30**(12), 1551(2005)

MO 24.5 Th 16:00 Lichthof

Coherent Control of multiphoton-processes in colloidal semiconductor nanocrystals — •Martin Ruge, Matthias Wollenhaupt, Thomas Baumert, and Alexander Horn — Universität Kassel, Institut für Physik und Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), Heinrich-Plett-Str. 40, D-34132 Kassel, Germany

In the recent past semiconductor nanocrystals - further referred as quantum dots (QDs) - have gained interest due to their atomic-like energy level structure. Coherent control of quantum states in single QDs has been demonstrated by double pulses [1]. So far, experiments were performed on epitactically grown QDs either sandwiched between semiconductor materials or grown on the surface of the latter and therefore the resonance energies are restricted to the infrared spectral range [1,2]. Here we present coherent control of multiphoton induced photoluminescence (mpl) of colloidal semiconductor quantum dots with one photon resonance energies in the visible spectral range by phase-modulated femtosecond laser radiation. The beam is focused in a dispersion of QDs and the luminescence is investigtated in dependence on the applied phase modulation functions and parameters. Part of the signals can be explained in terms of higher order spectral interference other parts rely on further control mechanisms. The results obtained within the scope of this experiments could lead to new applications e.g. color generation in QD doped materials like glasses.

[1] N. H. Bonadeo *et al.*, Science 282 (1998) 20
[2] S. Stufler, Phys. Rev. B 73 (2006) 125304

MO 24.6 Th 16:00 Lichthof

Ultrafast Resonant Strong-Field Control on K_2 — •Tim Bayer, Matthias Wollenhaupt, Cristian Sarpe-Tudoran, and Thomas Baumert — University of Kassel, Institute of Physics and CINSaT, D-34132 Kassel, Germany

Selective Population of Dressed States (SPODS) is an elementary mechanism of resonant strong-field control. Recently, SPODS realizations based on chirped laser pulses, pulse sequences and combinations thereof were demonstrated experimentally on **K** atoms [1] and discussed in terms of control landscapes of coherent electronic excitation [2]. Moreover, the applicability of SPODS to the coherent control of chemical reactions was pointed out by wave packet calculations on $\mathbf{K_2}$ [3]. The proposed pulse sequence scenario allows to switch quantum mechanical population transfer efficiently from a state of maximum electronic coherence among different electronic target states, with attosecond precision. In the experiment, we use phase shaped 30 fs, 800 nm laser pulses, e.g. multi-pulse sequences from sinusoidal phase modulation and double pulses from phase-step modulation, to excite $\mathbf{K_2}$ in a supersonic beam. The final populations achieved in the vari-

ous excited molecular states are probed by a time-delayed 570 nm laser pulse from an optical parametric amplifier. Employing energy resolved photoelectron spectroscopy, we investigate the switching capabilities of the molecular SPODS scheme in terms of efficiency and robustness.

[1] T. Bayer *et al.*, Phys. Rev. Lett. **102**, 023004 (2009)
[2] T. Bayer *et al.*, J. Phys. B **41**, 074007 (2008)
[3] M. Wollenhaupt *et al.*, J. Photochem. Photobiol. A **180**, 248 (2006)

MO 24.7 Th 16:00 Lichthof

Evolutionary sculptured 3-dimensional photoelectron wave packets — Marc Krug, •Manuel Gerlach, Qingqing Liang, Tim Bayer, Jens Köhler, Matthias Wollenhaupt, and Thomas Baumert — Universität Kassel, Institut für Physik und CINSaT, Heinrich-Plett-Str. 40, D-34132 Kassel, Germany

We present an approach to sculpt 3-dimensional photoelectron wave packets resulting from the excitation and ionization of atomic potassium using polarization-shaped fs laser pulses. Polarization shaped pulses are generated employing a Fourier-transform pulse shaper providing full control over the ellipticity of all spectral pulse components. The projections of the photoelectron wave packets are measured by photoelectron imaging spectrometry [1]. In a first experiment an evolutionary algorithm is used to optimize the photoelectron distributions in order to match a predefined shape. The optimal photoelectron wave packet is analyzed employing a tomographic technique based on sequential measurement of projections from polarization shaped pulses rotated about the propagation axis [2]. This results in photoelectron angular distributions observed from different views. A tomography algorithm reveals the real 3-dimensional photoelectron angular distribution. The control mechanism of the atomic excitation is discussed by comparison of simulated results based on temporarily varying polarization states of the used laser light.

[1] M. Wollenhaupt *et al.*, Appl. Phys. B 95: 245-259 (2009)
[2] M. Wollenhaupt *et al.*, Appl. Phys. B 95: 647-651 (2009)

MO 24.8 Th 16:00 Lichthof

Control of doped He nanodroplet dynamics with shaped femtosecond pulses — •Terry Mullins[1], Christian Giese[1], Barbara Grüner[1], Frank Stienkemeier[1], Marcel Mudrich[1], Roland Wester[1], and Matthias Weidemüller[2] — [1]Physikalisches Institut, Universität Freiburg, Hermann-Herder-Str. 3 79104 Freiburg, Germany — [2]Physikalisches Institut, Universität Heidelberg, Philosophenweg 12 69120 Heidelberg, Germany

We investigate the coherent excitation and ionization dynamics of doped He nanodroplets interacting with shaped ultrashort laser pulses. Atoms or molecules can be doped onto the nanodroplets and are thereby cooled. Excitation and ionization dynamics of the dopant atoms/molecules can be strongly influenced by their interaction with He on short time-scales and coherence effects can play an important role [1,2]. By applying shaped ultrashort pulses the quantum dynamics of the coupled alkali-droplet system can be coherently controlled. We are working towards addressing the vibrational wavepacket dynamics of Rb_2 and even the RbHe exciplex formation. The pulses are shaped using an SLM-based 4f setup [3]. Independent phase and amplitude shaping of the excitation pulse is possible and can also be applied within a feedback loop.

[1] M. Mudrich et al., Phys. Rev. A 80, 042512 (2009).
[2] M. Mudrich et al., Phys. Rev. Lett. 100, 023401 (2008).
[3] A. M. Weiner et al., J. Quant. Elec. 28, 908 (1992).

MO 24.9 Th 16:00 Lichthof

State selection in rotational wave packets by tuning from nonadiabatic to adiabatic interaction — •Nina Owschimikow[1], Burkhard Schmidt[2], and Nikolaus Schwentner[1] — [1]Institut für Experimentalphysik, Freie Universität Berlin — [2]Institut für Mathematik, Freie Universität Berlin

Non-adiabatically excited rotational wave packets display periodically recurring revivals of alignment after the termination of the excitation pulse. The amplitude of these oscillations decreases as the adiabatic limit is approached with increasing pulse duration. We show by numerical simulations for the nitrogen-molecule that this can be described as a convolution of pulse envelope and sinusoidal rotational response and it results in a unique dependence for all J quantum states on the ratio of pulse duration to rotational period.

In a thermal ensemble of molecules, a wide distribution of rotational levels is populated, with the crossover from the non-adiabatic to the adiabatic limit varying according to rotational frequency. Increasing the excitation pulse duration restricts the coherent post-pulse response to the cold part of the thermal distribution, thus creating a "cool" wave packet in a thermal bath. A pair of pump and dump pulses with the duration of the dump pulse tuned to interact non-adiabatically only with the rotationally "cool" molecules additionally serves to limit the coherence to the hot part of the distribution, thus allowing for state selectivity in spite of the non-resonant excitation mechanism.