MO 5: Quantenkontrolle 1

Zeit: Montag 14:00–16:00 Raum: VMP 6 HS-G

MO 5.1 Mo 14:00 VMP 6 HS-G

Optimization and application of heterodyne single-beam CARS microscopy — •Christina Müller[1], Tiago Buckup[1], Bernhard von Vacano[2], and Marcus Motzkus[1] — [1]Physikalische Chemie, Philipps-Universität Marburg, D-35032 Marburg, Germany — [2]Polymer Physics, BASF SE, D-67056 Ludwigshafen, Germany

Single-beam coherent anti-Stokes spectroscopy presents a unique and powerful approach to nonlinear microscopy since a complete CARS scheme is accomplished using just a tailored pulse from a fs-oscillator and coherent control of the non-linear signal generation by phase modulation of the excitation with a pulse shaper.[1] In a previous work, we have demonstrated that even a heterodyne detection scheme with an additional local oscillator can be implemented in the single-beam approach by pulse shaping.[2, 3] In this contribution, we present and compare two different approaches of modulating heterodyne signal for sensitive-lock-in detection. We show the careful optimisation of the experimental scheme analysing the different relations between local oscillator field and CARS signal intensity in order to improve the sensitivity of the method. After successful optimization the scheme is introduced to microscopic applications, combining chemical selectivity and a high spatial resolution of non-linear CARS processes with the sensitivity to low sample concentrations.

[1] N. Dudovich, D. Oron, Y. Silberberg, Nature 418, 512 (2002).
[2] B. von Vacano, T. Buckup, M. Motzkus, Opt. Lett. 31, 2495 (2006).
[3] B. von Vacano, J. Rehbinder, T. Buckup, M. Motzkus, Appl. Phys. B 91, 213 (2008).

MO 5.2 Mo 14:15 VMP 6 HS-G

New insights on the coherent control of selective bond breaking in model peptides — •Claus Peter Schulz[1], Ihar Shchatsinin[1], Tim Laarmann[1,2], Nickolai Zhavoronkov[1], and Ingolf V. Hertel[1] — [1]Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin — [2]Present address: HASYLAB at DESY, Notkestrasse 85, 22603 Hamburg

One of the goals in coherent control experiments on biologically relevant molecules is to establish an analytic tool. Recently, we have shown that selective bond breaking in model peptides, like Ac-Phe-NHMe, with shaped femtosecond laser pulses is achievable [1]. In this contribution we want to report on new insights into the photo induced bond breaking mechanism. A detailed analysis of the spectral content of the optimal pulse sequence indicates that the S_1 state acts as an isomer selective "doorway" in the highly non-linear excitation and fragmentation process.

[1] T. Laarmann, I. Shchatsinin, P. Singh, N. Zhavoronkov, C.P. Schulz, I.V. Hertel, J. Phys. B **41**, 074005 (2008).

MO 5.3 Mo 14:30 VMP 6 HS-G

Investigation of electronic and vibrational coherence in molecules by phase-locked double pulses — •Nils Krebs[1], Peter Baum[1], Jürgen Hauer[2], and Eberhard Riedle[1] — [1]Lehrstuhl für BioMolekulare Optik, LMU München — [2]Institut für Physikalische Chemie, Universität Wien

We investigated the femtosecond coherence dynamics of organic molecules in solution in a coherent control experiment based on transient absorption with double pulse excitation. The first pump pulse induces polarization oscillating with the transition frequency connecting ground and excited state. The second pulse with a well determined relative phase interacts constructively or destructively with the previously induced polarization. Hence, the obtained interference signal is a measure for electronic dephasing. A third pulse probes the excited state population at a ps delay time. The pulses were generated as identical copies of Fourier limited 11 fs visible pulses via a stabilized Michelson interferometer. In agreement with the expectation for a simple two-level system (here Perylene Orange in solution) the results are interpreted with the aid of homogeneous and inhomogeneous broadening mechanisms affecting the absorption line width. Extension of the pumping wavelength to the UV (350 nm, 22 fs pulse duration) allowed for the investigation of transient electronic and the vibrational phase effects during the excited state intramolecular proton transfer (ESIPT) of HBT. The double pulse temporal spacing was found to be the decisive control parameter for vibrational wavepacket control.

MO 5.4 Mo 14:45 VMP 6 HS-G

Generation and characterization of polarization-shaped broadband visible femtosecond laser pulses — •Tatjana Löhrig, Christoph Benjamin Schwarz, Frank Dimler, Florian Langhojer, Ulrike Selig, and Tobias Brixner — Institut für Physikalische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

The laser pulses presented here are generated in a noncollinear optical parametric amplifier (NOPA). Polarization shaping is achieved by an LCD pulse shaper specifically designed for broadband visible laser pulses. The pulses are spectrally dispersed by a volume phase holographic grating which exhibits a high and flat diffraction efficiency over the entire visible spectral range. All other optical components are reflective in order to minimize material dispersion.

A novel method of pulse characterization is described which is based on dual-channel spectral interferometry. It is a combination of dual-quadrature spectral interferometry to extract the phases of the two linearly independent polarization components of the pulse and a tomographic pulse measurement to yield the relative phase between them.

First examples are shown and compared to the results of conventional dual-channel spectral interferometry.

MO 5.5 Mo 15:00 VMP 6 HS-G

Photoelectron angular distributions from strong–field coherent electronic excitation — •Matthias Wollenhaupt, Marc Krug, Jens Köhler, Tim Bayer, Cristian Sarpe-Tudoran, and Thomas Baumert — UniversitätKassel, Institut für Physik und CINSaT, Heinrich-Plett-Str. 40,D-34132 Kassel, Germany

Photoelectron angular distributions (PADs) resulting from non–perturbative excitation of potassium atoms using shaped femtosecond laser pulses are presented. We study control exerted by (1) the polarization of an unshaped light pulse, (2) shaped linearly polarized light and (3) a combination of both degrees of freedom, i.e. polarization–shaped laser pulses. A theoretical approach to describe PADs from non–perturbative Resonance Enhanced Multi–Photon Ionization (REMPI) with ultrashort polarization–shaped laser pulses is presented and compared to experimental results. Applications of this technique to the generation and observation of atomic ring currents are discussed.

MO 5.6 Mo 15:15 VMP 6 HS-G

Robust Photon Locking — •Tim Bayer, Matthias Wollenhaupt, Cristian Sarpe-Tudoran, and Thomas Baumert — Universität Kassel, Institut für Physik und CINSaT, Heinrich-Plett-Str. 40, D-34132 Kassel, Germany

We experimentally demonstrate a strong–field coherent control mechanism that combines the advantages of Photon Locking (PL) and Rapid Adiabatic Passage (RAP). Unlike earlier implementations of PL and RAP by pulse sequences and chirped pulses respectively, we use shaped pulses generated by phase modulation of a femtosecond laser spectrum with a generalized phase discontinuity. The novel control scenario is characterized by a high degree of robustness achieved via adiabatic preparation of a state of maximum coherence. Subsequent phase control allows for efficient switching among different target states. We investigate both properties by photoelectron spectroscopy on potassium atoms interacting with the intense shaped light field. Since adiabatic Photon Locking combines efficient ultrafast switching with robustness, this scenario provides a novel tool for real–world applications in quantum control.

MO 5.7 Mo 15:30 VMP 6 HS-G

Simultaneous spatial and temporal control of optical near-fields at metal nanostructures — Martin Aeschlimann[1], Michael Bauer[2], Daniela Bayer[1], Tobias Brixner[3], Stefan Cunovic[4], •Frank Dimler[3], Alexander Fischer[1], F. Javier García de Abajo[5], Viktor Myroshnychenko[5], Walter Pfeiffer[4], Martin Rohmer[1], Christian Schneider[1], Felix Steeb[1], Christian Strüber[4], and Dmitri V. Voronine[3] — [1]TU Kaiserslautern, Kaiserslautern, Germany — [2]Universität Kiel, Kiel, Germany — [3]Universität Würzburg, Würzburg, Germany — [4]Universität Bielefeld, Bielefeld, Germany — [5]Consejo Superior de Investigaciones Cientificas, Madrid, Spain

Expanding the recently demonstrated adaptive control of optical near-fields [1] we investigate the temporal evolution of the local electronic excitation of nanostructures by spatially resolved cross correlation measurements using two-photon photoemission electron microscopy. Planar silver nanostructures manufactured by e-beam lithography are excited by polarization shaped pump pulses and the corresponding local excitation is probed by a time-delayed circularly polarized laser pulse. The experiments confirm that the polarization-shaped incident laser pulse does indeed switch between two distinct excitation patterns within a time scale that is only limited by the pulse duration of the laser system. First spatio-temporal control experiments show simultaneous subpicosecond and subwavelength resolution.

[1] M. Aeschlimann, et al.; Nature 446, 301 (2007).

MO 5.8 Mo 15:45 VMP 6 HS-G

Femtosecond quantum control calculations in the frequency domain for vibrational processes — •Caroline Gollub and Regina de Vivie-Riedle — LMU, Department Chemie und Biochemie, Butenandtstr. 11, 81377 München

In recent years, coherent control of molecular vibrational excitation with shaped mid-IR laser fields has been achieved. Inspired by the experiments, the question is investigated whether and how the results of optimal control experiments based on genetic algorithms (GAs) can be traced in the solution subspace of optimal control theory. Calculations on vibrational unitary transformations are performed with single- [1] and multi-objective [2] GAs in close analogy to the experiment. The minimum requirements on the pulse properties can be extracted from the resulting Pareto fronts. Additionally, a flexible, alternative optimization approach is proposed based on a modified ant colony optimization algorithm. The method allows to decrease the shaped pulse complexity and duration, without loss of efficiency. Short pulse durations are essential in the context of quantum information processing for fast quantum gates which are most robust in the presence of dissipation. The experimental implementation prospects of IR quantum gates in $W(CO)_6$ are discussed in condensed phase.

[1] C. Gollub and R. de Vivie Riedle, Phys. Rev. A, 78 (2008), 033424.

[2] C. Gollub and R. de Vivie Riedle, New J. Phys., (2008) accepted.