MO 61: Quantum Control I

Zeit: Freitag 10:30–12:30 Raum: 6F

MO 61.1 Fr 10:30 6F

Excitation of C_{60} with Temporally Shaped Laser Pulses: Coherent Heating of Nuclear Motion — •Claus Peter Schulz, Ihar Shchatsinin, Tim Laarmann, Nickolai Zhavoronkov, and Ingolf Volker Hertel — Max Born Institute, Max-Born-Str. 2a, 12489 Berlin-Adlershof, Germany

Femtosecond laser pulses tailored with closed-loop, optimal control feedback were used to optimise the C_2 evaporation from C_{60} [1]. A characteristic pulse sequence results in significant enhancement of the C_{50}^+ yield, a typical fragment of vibrationally hot C_{60}, in comparison with the response to a single pulse of the same energy and overall width. The separation between subsequent pulses is close to the vibrational period of the radial symmetric $a_g(1)$ breathing mode. In a two-colour pump-probe experiment similar modulations were observed in the ion signal of multiply charges fragments. The observed period (80-127 fs) of this oscillation depends on the degree of ionisation and the deposited energy. Comparison with TDDFT calculations indicates that the shaped laser pulse excite giants oscillations in C_{60}, which prevails for several cycles.

[1] T. Laarmann, I. Shchatsinin, A. Stalmashonak, M. Boyle, N. Zhavoronkov, J. Handt, R. Schmidt, C. P. Schulz, and I.V. Hertel, Physical Review Letters, in press (2006)

MO 61.2 Fr 10:45 6F

Laserinduced breathing mode in highly excited C_{60} — •Jan Handt[1] and Rüdiger Schmidt[2] — [1]Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Strasse 38, D-01187 Dresden — [2]Institut für Theoretische Physik, Technische Universität Dresden, D-01062 Dresden

We present, a theoretical study of the excitation of C_{60} by strong laser pulses. We use the so-called nonadiabatic quantum molecular dynamics (NA-QMD) which combines self-consistently the electron dynamics treated with time-dependent density functional theory (td-DFT) and classical molecular dynamics. Exciting the molecule via the t_{1g} resonance our calculations show a strong multielectron excitation of C_{60}. Surprisingly most of the vibrational energy is stored in the $a_g(1)$ breathing mode. The period of the breathing mode depends on the deposited energy. We show, that oscillations observed in pump-probe and optimal-control experiments are due to this effect [1]. [1] T. Laarmann, I. Shchatsinin, A. Stalmashonak, M. Boyle, N. Zhavoronkov, J. Handt, R. Schmidt, C. P. Schulz, and I.V. Hertel, Physical Review Letters, in press (2006)

MO 61.3 Fr 11:00 6F

Optimale Kontrolle von laserinduzierten Reaktionen von H_2 mit CO auf einer Pd(100) — •Daniel Wolpert[1], Patrick Nuernberger[1], Horst Weiss[2] und Gustav Gerber[1] — [1]Physikalisches Institut, Universität Würzburg, Am Hubland, — [2]BASF AG, Polymer Research Division, 67056 Ludwigshafen, Germany

Wir untersuchen die Reaktion von H_2 und CO Molekülen auf einer Pd(100) Einkristalloberfläche mit Femtosekunden-Laserimpulsen. Mittels Flugzeitmassenspektroskopie werden verschiedene Produktmoleküle, darunter auch solche (z.B. CH_3^+), bei deren Bildung mindestens drei Reaktionspartner mit der Oberfläche und dem Laserfeld wechselwirken müssen, beobachtet. Durch die Anwendung von optimal geformten Femtosekunden-Laserimpulsen können die Verhältnisse der entstehenden Produktionen gezielt beeinflusst werden. Im Gegensatz zu früheren Quantenkontrollexperimenten, in denen Bindungen in Molekülen selektiv gebrochen wurden, wird hier die Bildung von Molekülbindungen kontrolliert.

MO 61.4 Fr 11:15 6F

Coherent Control despite a dominant spectral Continuum: — •Heide Ibrahim[1], Mónika Héjjas[1], Mizuho Fushitani[2], Markus Gühr[3], and Nikolaus Schwentner[1] — [1]FU Berlin, Institut für Experimentalphysik, Arnimallee 14, 14195 Berlin — [2]Institute for Molecular Science, Okazaki, Aichi 444-8585, Japan — [3]Stanford PULSE Center, Varian Physics Bldg., 382 Via Pueblo Mall, Stanford CA 94305-4060, USA

The interaction strength of a vibrational mode with a multidimensional (crystalline) environment is reflected in frequency domain spectroscopy in the lineshape. Weak coupling leads to sharp zero-phononlines (ZPL) which represent an exclusive chromophore vibrational excitation. Phonon side bands (PSB) display the simultaneous excitation of bath modes which merge to a continuum with increasing strength. Concerning coherent control of molecular dynamics it is desirable to generate analoguos weakly and strongly coupled vibrational wave packets composed of ZPL or PSB respectively by appropriate femtosecond pulse sequences. The X-A and X-B transitions of Br_2 in an Ar matrix provide these properties and are used to develop the means to achieve this separation with phase locked pulses. Firstly we show that the powerful A continuum and the faint B progression can be sorted out by either state-selective fluorescence or an additional probe step. Secondly we illustrate how variably coupled wave packets are generated by optimising timing and phases of pulse sequences. Pros and cons of sequence generation with spatial light modulators or interferometers are analysed based on a Fouriertransformation.

MO 61.5 Fr 11:30 6F

Analyse von Mechanismen der Quantenkontrolle durch farbige Femtosekunden-Doppelimpulse — Gerhard Vogt, Patrick Nuernberger, Reimer Selle, •Frank Dimler, Tobias Brixner und Gustav Gerber — Physikalisches Institut, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Die einer adaptiven Quantenkontrolle mit Femtosekunden-Laserimpulsen zugrunde liegenden Mechanismen können mit Hilfe von sogenannten Fitnesslandkarten, die nur von einer beschränkten Anzahl von Impulsformparametern abhängen, gefunden werden [1]. Bei der Fitness handelt es sich hierbei beispielsweise um die Effizienz einer Photoreaktion.

In einem ersten Experiment werden farbige Femtosekunden-Doppelimpulse verwendet, um eine solche Fitness als Funktion der zeitlichen Separation und der relativen Amplitude der beiden Laserimpulse aufzunehmen. Der Vergleich mit den Ergebnissen einer freien Optimierung zeigt die Übereinstimmung mit der Fitnesskarte.

In einem weiteren Experiment wird geklärt, ob die Anregungseffizienz des Laserfarbstoffes IR140 bei linear gechirpten Laserimpulsen äquivalent ist zur Anregung mit farbigen Doppelimpulsen bei entsprechendem Zeit- und Frequenzabstand.

[1] G. Vogt et al., Phys. Rev. A **74,** 033413 (2006)

MO 61.6 Fr 11:45 6F

Selective bond breaking in model dipeptides with temporally shaped laser pulses — •Tim Laarmann, Ihar Shchatsinin, Nick Zhavoronkov, Claus Peter Schulz, and Ingolf Volker Hertel — Max Born Institute, Max-Born-Str. 2A, 12489 Berlin

The control of photophysical processes with judiciously tailored femtosecond laser pulses is a cutting edge topic in modern laser science and might pave the way to optically controlled organic chemistry [1]. In this contribution we report on pulse-shaping experiments using closed-loop, optimal control feedback for selective bond breaking in Ac-Phe-NHMe. This molecule contains a -CO-NH-CHR-CO- moiety, the key structural element of peptides. Moreover, due to the two amino groups this system can be regarded as a model dipeptide. Perspectives for "laser-induced protein sequencing (LIPS)" using temporally shaped laser pulses will be discussed.

[1] T. Laarmann, I. Shchatsinin, A. Stalmashonak, M. Boyle, N. Zhavoronkov, J. Handt, R. Schmidt, C. P. Schulz, and I.V. Hertel, Physical Review Letters, in press (2006)

MO 61.7 Fr 12:00 6F

Quantum Information Transfer through Molecular Chains — •Caroline Gollub, Ulrike Troppmann, and Regina de Vivie-Riedle — LMU Department Chemie, Butenandt-Str. 11, 81377 München, Germany

Molecular bridge systems consisting of $(C{\equiv}C-)_n$ units, such as octatetrayne, offer an innovative possibility to construct quantum networks, within our approach of molecular quantum computing [1].

The vibrational qubits are encoded in selected normal modes of molecular subunits. These multiple nodes are connected by molecular chains to larger quantum registers. Ultrashort, shaped laser pulses, act as quantum gates on the multi-qubit systems. Independent control of the nodes is possible. The bridging molecules represent the

units, essential for quantum information transfer. The communication between the qubit systems is realized by specially shaped, ultrashort laser pulses, calculated with Optimal Control Theory. In this context we present shaped pulses inducing selective quantum state transfer through different quantum channels of the bridging molecules. Additionally, the generation of inter-node superpositions has to be required for quantum multicomputers and we demonstrate the implemenation of entanglement generation in our approach of scalable molecular quantum computing with vibrational qubits.

[1] C. M. Tesch, R. de Vivie-Riedle, Phys. Rev. Lett., **89**, (2002).

MO 61.8 Fr 12:15 6F

Single-beam Heterodyne CARS for Sensitive Chemically Selective Microscopy — •Bernhard von Vacano, Tiago Buckup, and Marcus Motzkus — Physikalische Chemie, Philipps-Universität Marburg, D-35032 Marburg, Germany

With shaped femtosecond laser pulses, coherent anti-Stokes Raman-scattering (CARS) microspectroscopy can be accomplished in a straight-forward single-beam approach. To increase the sensitivity, we introduce interferometric detection in a very simple experimental extension: in addition to the optical fields driving the CARS process, a controllable local oscillator is derived by pulse-shaping of a single femtosecond laser beam. The heterodyne detected signal can be amplified by more than three orders of magnitude and shows linear concentration dependence. Like this, the sensitivity of chemically selective CARS detection can be increased dramatically, without adding complexity to the single-beam setup. As an example, we show sensitive and chemically selective detection of down to 8×10^6 analyte molecules.