MO 18: Experimental Techniques

Time: Wednesday 14:00–16:00 Location: F 102

MO 18.1 Wed 14:00 F 102

Photon economy considerations for pump-probe spectroscopy — •Gerald Ryseck and Peter Gilch — HHU Düsseldorf, AG Femtosekundenspektroskopie, Universitätsstr. 1, 40225 Düsseldorf

Pump-probe techniques are commonly applied to achieve high time resolution in ultrafast spectroscopy of physical and chemical processes. The time evolution of the derived signals can for the most part be described by a superposition of exponential decays. One aim is to estimate the involved rate constants with a defined accuracy. One fundamental result of parameter estimation theory is the Cramér-Rao lower bound [1] giving a limit for the lowest variance of the estimator. Even though applied for photon counting measurements [2] similar consideration for pump probe setups are lacking. We here present our findings for this case and discuss the choice of time delays giving the highest information on the decay.

[1] H-O Georgii, "Stochastik", Walter de Gruyter, Berlin, 2004
[2] M Köllner, J Wolfrum, Chem Phys Lett, (1992) 200, 1-2, 199-204

MO 18.2 Wed 14:15 F 102

On the Background Problem of Fluorescence Kerr Gating — •Ramona Mundt, Gerald Ryseck, and Peter Gilch — Institut für Physikalische Chemie, HHU Düsseldorf, Universitätsstr.1, 40225 Düsseldorf

Fluorescence decays in the femto- to picosecond time regime are usually traced by upconversion or optical Kerr gating [1].

Though Kerr gating commonly offers a larger spectral coverage than upconversion, it suffers from background light leaking through the gate. It consists of a pair of crossed polarisers through which a portion of fluorescence light leaks in the closed state. This becomes particularly problematic when studying long-lived fluorophores with lifetimes up to some hundred picoseconds or longer.

When employing large aperture optics to collect the fluorescence light, the leakage is predominantly due to the intrinsic depolarising properties of the optics. These properties will be presented for a gate consisting of Cassegrainian microscope objectives and wire grid polarisers [2]. Since the depolarisation is not uniform along the beam cross section, suitable apertures can reduce the background. As shown in a recent study [3] with such an aperture, measurements on long-lived fluorophores are possible. Further refinements will open the route to fluorophores with nanosecond lifetimes keeping 100 fs time resolution.

[1] G. Ryseck and P. Gilch, "Ultrafast Fluorescence Spectroscopy", Walter de Gruyter, in press [2] B. Schmidt et al., Appl. Phys. B, 2003, 76/8, 809-814 [3] T. Cordes et al., J. Photochem. Photobiol. A, 2009, 206/1, 10-17

MO 18.3 Wed 14:30 F 102

Elimination of strong two-photon-excited fluorescence using a single-beam-CARS-setup — •Alexander Wipfler, Rehbinder Jean, Buckup Tiago, and Motzkus Marcus — Physikalisch-Chemisches Institut, Ruprecht-Karls-Universität Heidelberg, D-69120 Heidelberg, Germany

The absence of fluorescence is often claimed to be the major advantage of Coherent Anti-Stokes Raman Scattering (CARS) experiments compared to spontaneous Raman measurements. Using ultra-short laser pulses and therefore high peak intensities may cause a strong contribution of two-photon-excited fluorescence (2PEF), which overlays the CARS signal. Especially in single-beam-CARS experiments the use of short pulses with durations in the range of 10 fs is mandatory. In our talk we will present strategies to extract the CARS contribution from measurements where strong 2PEF is present. We harness the interference of the CARS field with a local oscillator that is intrinsically generated in single-beam-CARS experiments. A double-quadrature spectral interferometry (DQSI) scheme is used to extract Raman equivalent information by subtracting CARS signals for different excitation spectra. However, the phase-shaping necessary for single-beam-CARS also affects the two-photon-absorption and therefore 2PEF is not necessarily canceled out in the DQSI scheme although it is an incoherent process. We present two phase-shaping schemes that keep the 2PEF contribution constant and allow for the complete extraction of Raman-equivalent spectra. [1] Wipfler et al. J. Raman Spec. 2012 (accepted)

MO 18.4 Wed 14:45 F 102

Biological samples imaged by multimodal nonlinear microscopy with 10fs pulses. — Jean Rehbinder, •Lukas Brückner, Tiago Buckup, and Marcus Motzkus — Physikalisch-Chemisches Institut, Ruprecht-Karls-Universität Heidelberg, D-69120 Heidelberg, Germany

In Multimodal Nonlinear Optical Microscopy (NLOM), several multiphoton signals are detected simultaneously providing complementary, label-free contrast mechanisms based on chemical and structural properties of the sample. Usually, pulses with durations down to 100fs are used. In this work, imaging with 10fs NIR pulses is demonstrated. A 10 fold increase in two-photon signals is predicted and 100 fold for three-photon effects. Such broadband pulses are strongly affected by the big amount of dispersion introduced by microscope objectives. A pulse shaper is used to compensate arbitrary phase distortions. Once the pulse is compressed, the shaper can be used to tailor it with high flexibility and increase contrast and selectivity of the excitation. These advantages are illustrated using Second Harmonic Generation (SHG) in combination with Two-Photon Excited Fluorescence (TPEF) and Coherent Anti-Stokes Raman Scattering (CARS) for imaging of skin biopsies. Phase and amplitude shaping schemes are demonstrated on the basis of moss leaves using CARS and TPEF. Polarization control gives access to the tensorial nature of given nonlinear effects. The potential to determine orientation of collagen fibrils in a tendon from a rat-tail is shown and illustrates the versatility of pulse shaping for nonlinear microscopy.

MO 18.5 Wed 15:00 F 102

Non-linear interferometric autocorrelation in the vacuum ultraviolet spectral range — •Sergey Usenko[1], Jörn Bödewadt[2], Andreas Przystawik[1], and Tim Laarmann[1] — [1]Deutsches Elektronen-Synchrotron, DESY Photon Science, Hamburg, Germany — [2]Universität Hamburg, Hamburg, Germany

In this contribution the conceptual design of an interferometric autocorrelator working in the vacuum ultraviolet (VUV) spectral range is presented. A MEMS (microelectromechanical system) based on a micro mirror array is the key component of the planned setup. Simulations of the MEMS performance were made using the ZEMAX software package. The results show that such device can act as an effective beam splitter. An outlook on the planned experiments will be given.

MO 18.6 Wed 15:15 F 102

Stationary Flow Conditions in Pulsed Supersonic Beams — •Wolfgang Christen and Bo-Gaun Chen — Humboldt-Universität zu Berlin, Brook-Taylor-Straße 2, 12489 Berlin, http://c

One of the most relevant characteristics of pulsed supersonic jets is the duration of the steady hydrodynamic flow: Due to the complex collision dynamics at elevated densities current model calculations are based on the assumption of a quasi-continuous flow. Accordingly, a comparison of experimental results with theoretical models requires a sufficiently long opening of the valve. Too short opening times do not allow for the evolution of stationary beam properties and may lead to incorrect conclusions.

We present a comprehensive analysis of this key aspect, using a focused electron beam with a short on-time for tagging a localised portion of the expanding jet, permitting measurements with high spatio-temporal resolution. It also provides an invaluable diagnostic tool for characterising the evolution of beam properties within the very short particle bunches of fast acting valves. In particular, the study reveals that the true opening time of the valve may be strikingly longer than the control pulse applied to the valve driver. Although pulse durations as short as 10 μs are possible, much longer pulse durations are required for an optimised adiabatic cooling. Also, the in-depth analysis of arrival time distributions for different pulse durations of the valve allows for the identification of quasi-continuous flow conditions with respect to number density, translational temperature, and particle speed.

MO 18.7 Wed 15:30 F 102

Optimized Cooling and Transmission of Pulsed High-Density Molecular Beams — •Wolfgang Christen and Bo-Gaun Chen — Humboldt-Universität zu Berlin, Institut für Chemie, Brook-Taylor-Straße 2, 12489 Berlin, http://clusterlab.de

Pulsed jet sources facilitate beam parameters that are hardly accessi-

ble with a continuous expansion. Most notably, they provide a means to substantially increased source densities, allowing for the convenient and reliable generation of supersonic beams including liquid and supercritical stagnation conditions. However, obtaining meaningful values for the mean cluster size, kinetic energy, and temperature at high beam densities also requires a collision-free beam propagation. At increased source densities this is a non-trivial and frequently neglected issue.

We demonstrate the substantial influence of background gas pressure and beam-skimmer interactions on the minimum attainable beam temperature. The thorough analysis of beam properties and the correspondingly optimised choice of expansion parameters result in much colder beams, at the same conditions of source pressure and temperature, allowing to reduce the beam temperature by more than one order of magnitude.

MO 18.8 Wed 15:45 F 102

COATEX: Testing optical coatings for resistance against tritium — •Vera Schäfer and Sebastian Fischer — Karlsruhe Institute of Technology (KIT), Institute of Technical Physics - Tritium Laboratory Karlsruhe (ITEP-TLK), Germany

The Coating Test Experiment COATEX is an experiment to monitor the tritium compatibility of optical windows with different reflection coatings. The monitoring of these samples is based on transmission/reflection measurements, which are carried out with the aid of laser diodes, photodiodes and other optical components. The tritium compatibility of these coatings is important for the Laser Raman spectroscopy (LARA). The main component of LARA is the LARA cell, which is totally floated with tritium and has four windows with such optical coatings. During test measurements with a high tritium concentration these coatings were damaged.

This talk gives a brief overview about the experimental setup and the measurement principle of COATEX. A long term measurement showed problems with the reproducibility of the measurement results, because drifts of 11.5 % occurred. Possible reasons for these drifts like a temperature dependence of the photodiodes, electrical noise and an undefined polarisation of the laser diodes will be presented. In addition, mechanical instabilities of the different optical components caused variations of the measurement results. Although not all planned modifications of the experimental setup have yet been realized, the drift has been reduced to 3 %.