

MO 20: Experimental Techniques I

Time: Thursday 10:30–12:30

Location: F 142

Prize Talk

MO 20.1 Th 10:30 F 142

Speckle-Reduktion bei der Infrarotlaser-gestützten abbildenden Ferndetektion von TNT-Oberflächenkontaminationen — ●JAN KASTER — Ludwig-Maximilians-Universität München – Träger des Georg-Simon-Ohm-Preises

Das Ziel der Diplomarbeit war die Speckle-Reduktion bei der Infrarotlaser-gestützten abbildenden Ferndetektion von TNT-Oberflächenkontaminationen. Für die Detektion wird die Probenoberfläche mit einem abstimmbaren Quantenkaskadenlaser bei verschiedenen Wellenzahlen im Spektralbereich von 1340cm^{-1} bis 1380cm^{-1} bestrahlt, während eine Infrarot-Kamera Bilder der bestrahlten Fläche aufzeichnet. Diese Bilder werden dann so verarbeitet, dass im Ausgabebild die chemisch spezifische TNT-Absorption durch eine Änderung der Helligkeit örtlich aufgelöst erkennbar wird.

Da kohärente Strahlung, welche an optisch rauen Oberflächen reflektiert und mit einem abbildenden System beobachtet wird aufgrund der stochastischen Verteilung der Oberflächenrautiefe eine ebenso lateral stochastische Phasenverteilung der Wellenfront aufweist, bildet sich eine entsprechende Interferenz der Punktspreizfunktionen auf dem bildgebenden Detektor aus – es entsteht ein „Rauschen“ in der aufzeichneten Strahlungsleistungsverteilung, das sog. Speckle-Muster. Die Empfindlichkeit der oben skizzierten lasergestützten Messtechnik wird im Wesentlichen durch diesen Effekt limitiert.

Um die durch Laser-Speckles verursachten Rauschanteile zu reduzieren, gibt es verschiedene Möglichkeiten. Durch Reduktion von zeitlicher und räumlicher Kohärenz der Strahlung wird deren Interferenzfähigkeit reduziert und durch Variieren des Speckle-Musters während einer Bildaufzeichnung führen Mittelungen unkorrelierter Rauschanteile zu einer weiteren Speckle- Kontrastreduktion. Letztere Möglichkeit resultiert daraus, dass Laser-Speckles statistisch durch die Familie der Gammaverteilungen beschrieben werden können und sich das Signal-zu-Rausch-Verhältnis demnach mit der Anzahl gemittelter, unkorrelierter Rauschmuster proportional nach $S/N \propto \sqrt{N}$ verhält, wobei N der Anzahl unkorrelierter Speckle-Muster entspricht.

Ein im MIR-Spektralbereich umsetzbarer Ansatz, der all diese Möglichkeiten berücksichtigt, wurde bereits 1971 von Lowenthal und Joyeux (Lowenthal, S. und Joyeux, D. Speckle Removal by a Slowly Moving Diffuser Associated with a Motionless Diffuser. *Journal of the Optical Society of America*. 1971, Bd. 61, 7, S. 847-851) vorgeschlagen und im Rahmen der Arbeit für den infraroten Spektralbereich adaptiert. Dazu wird die Strahlung mit zwei polykristallinen CVD-Diamant-Wafern moduliert; ein Wafer ist dabei stationär und der andere rotiert mit ca. 70Hz.

In der Diplomarbeit konnte mit diesem Ansatz eine Steigerung des Signal-zu-Rausch-Verhältnisses in der betrachteten Strahlungsleistungsverteilung von ursprünglich 3 auf etwa 45 erzielt werden. So konnten nach unserem Wissen erstmals TNT-Kontaminationen von nur $10\mu\text{g}/\text{cm}^2$ in einem Abstand von 5m abbildend nachgewiesen werden. Dies entspricht der Kontamination einer Oberfläche durch einen leicht mit TNT verunreinigten Fingerabdruck.

MO 20.2 Th 11:15 F 142

Target preparation for diffractive X-ray imaging of gas-phase molecules — ●FRANK FILSINGER¹, JOCHEN KÜPPER¹, GERARD MEIJER¹, LOTTE HOLMEGAARD², JOCHEN MAURER², JAN THØGENSEN², and HENRIK STAPELFELDT² — ¹Fritz-Haber-Institut der MPG, Berlin, Germany — ²University of Aarhus, Denmark

In 2009 the Linac Coherent Light Source (LCLS), a novel hard X-ray free-electron laser, became operational. One envisioned application is the diffractive imaging of isolated, non-crystallizable bio-molecules in the gas phase [1]. Because a single X-ray laser pulse will completely destroy the molecule, its diffraction pattern needs to be recorded in a single shot. The (electronic) structure of the molecule must not significantly change on the timescale of the laser pulse (<100 fs) in order to obtain unperturbed diffraction images. In order to benchmark radiation damage and the structure retrieval algorithms, we will perform first studies on small molecules (e. g., diiodobenzene) at LCLS in spring 2010. In order to observe the diffraction image, ensembles of identical molecules must be prepared. We have demonstrated the spatial separation of individual structural isomers [2, 3] and very strong laser-alignment and mixed field orientation of such molecules [4]. Here, we will discuss how such targets can be prepared, using quantum-state-

selection techniques, and applied for X-ray diffraction experiments.

- [1] R. Neutze et al., *Nature* **406** (2000), 752
- [2] F. Filsinger et al., *Phys. Rev. Lett.* **100**, (2008), 133003
- [3] F. Filsinger et al., *Angew. Chem. Int. Ed.* **48**, (2009), 6900
- [3] L. Holmegaard et al., *Phys. Rev. Lett.* **102**, (2009), 023001

MO 20.3 Th 11:30 F 142

Quantum state effects on alignment and orientation of OCS — ●JENS HEDEGAARD NIELSEN¹, JOCHEN MAURER², SOFIE LOUISE KRAGH², HENRIK STAPELFELDT^{2,4}, FRANK FILSINGER³, JOCHEN KÜPPER³, and GERARD MEIJER³ — ¹Department of Physics and Astronomy, Aarhus University, 8000 Aarhus C, Denmark — ²Department of Chemistry, Aarhus University, 8000 Aarhus C, Denmark — ³Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ⁴Interdisciplinary Nanoscience Center (iNANO), Aarhus University, 8000 Aarhus C, Denmark

We present new experiments and simulations of laser induced adiabatic alignment and orientation of Quantum-State-Selected carbonyl sulfide (OCS). We systematically investigated the effect of rotational quantum levels on the degree of alignment and orientation.

A molecular beam is formed in a supersonic expansion and deflected in a strong inhomogeneous electric field. The deflection strength is determined by the effective dipole moment and thus the molecular rotational quantum state. The molecular beam is crossed by two focused laser beams. One nanosecond alignment beam and one femtosecond Coulomb explosion probe beam. The adiabatic laser alignment and mixed static field and laser orientation is investigated as a function of position in the molecular beam and thus as a function of the selected rotational states. We also present simulations of the alignment and mixed field orientation for individual quantum states as well as the states present in the deflected beam as extracted from Monte-Carlo trajectory simulations of the deflection process.

MO 20.4 Th 11:45 F 142

Photoelectron distributions of fixed-in-space molecules — LOTTE HOLMEGAARD¹, JONAS L. HANSEN¹, LINE KALHØJ¹, SOFIE LOUISE KRAGH¹, HENRIK STAPELFELDT¹, FRANK FILSINGER², ●JOCHEN KÜPPER², GERARD MEIJER², DARKO DIMITROVSKI¹, MAHMOUD ABU-SAMHA¹, CHRISTIAN P. J. MARTINY¹, and LARS BOJER MADSEN¹ — ¹University of Aarhus, Denmark — ²Fritz-Haber-Institut der MPG, Berlin

Currently, many novel investigations, such as ultrafast X-ray or electron diffraction, aim at providing structural and dynamical information directly from the molecular frame. In order to experimentally obtain such data from molecular ensembles, one has to rigidly connect the laboratory and molecular frames. Over the last years, we have developed the necessary techniques to perform such experiments: We have demonstrated the quantum-state and conformer-selection of large molecules using inhomogeneous electric fields. We have successively demonstrated unprecedented degrees of laser alignment and mixed (dc-electric and laser) field orientation of these samples, rigidly linking molecular and laboratory frame. We have now exploited these state-selected and oriented samples to measure photoelectron angular distributions in the molecular frame (MFPADs) from non-resonant femtosecond-laser photoionization. The obtained MFPADs show rich structure which provide information about the charge distribution and electrical properties of the molecule. Moreover, these MFPADs could provide detailed information on ultrafast molecular dynamics in future pump-probe experiments.

MO 20.5 Th 12:00 F 142

Comparison of fluorescence spectra of tryptamine obtained by thermal vaporization and laser desorption — ●BENJAMIN STUHLMANN and KARL KLEINERMANN — Heinrich Heine Universität, Düsseldorf, Institut für Physikalische Chemie

Laser desorption is an alternative to thermal vaporization for experiments in molecular beams.

In this work it is used to probe biomolecules, which can be susceptible to fragmentation at the temperatures needed for thermal vaporization.

The exposition of the probed molecules to heat in a desorption source occurs only for a short time and in a small spatial region determined

by the desorption laser spot. In a thermal source, the whole supply of sample is constantly heated.

We used tryptamine as test-molecule for desorption from a graphite surface by a Nd:YAG laser at 1064 nm.

Tryptamine is a monoamine alkaloid found in plants, fungi and animals. It contains an indole-ring as chromophore like the amino-acid tryptophan. Tryptophan is fluorophore in many folded proteins.

This relation -and its suspected role as a neurotransmitter- makes tryptamine a very interesting model system for spectroscopical investigation of biomolecules.

Spectral data of tryptamine after thermal vaporization are available from our group, thus these results could be compared to the spectra obtained after laser desorption.

MO 20.6 Th 12:15 F 142

Hyperspectral data processing for chemoselective MCARS microscopy using principal component analysis — •CHRISTOPH POHLING^{1,2}, TIAGO BUCKUP^{1,2}, and MARCUS MOTZKUS^{1,2} — ¹Physikalisch-Chemisches Institut, Ruprecht-Karls-Universität, D-69120 Heidelberg, Germany — ²Physikalische Chemie, Philipps-

Universität, D-35043 Marburg, Germany

Multiplex Coherent anti-Stokes Raman Scattering (MCARS) microscopy is a labelling free imaging technique, which has been steadily improved during the last decade [1]. Important for future application in medicine is the capability of providing chemoselective image contrast in case of biological samples. In this context, the Raman lineshape should be retrieved from the coherent CARS signal [2] and the unknown sample components must be labelled clearly. We have implemented a processing tool for MCARS microscopy that applies principal component analysis (PCA) to generate chemical contrast. Initially, the PCA calculates the eigenvectors from the MCARS hyperspectral data set. Later the image is recomposed automatically from the main eigenvectors received from PCA. The last step is realized either by matrix multiplication or by using an evolutionary fitting algorithm. We discuss the sensitivity regarding line width, line separation and sample concentration with simulated data. Furthermore, we show the effect of the extraction of the RAMAN line on the sensitivity of this approach and demonstrate its capability in biological samples. [1] von Vacano, B. et al., *J. Raman Spectr.*, 7 (2007) 916. [2] Liu, Lee, Cicerone, J. *J. Raman Spectr.*, 40 (2009) 726.