

MO 24: Experimental Techniques

Time: Friday 10:30–12:15

Location: PA 2.150

MO 24.1 Fri 10:30 PA 2.150

Investigations of Molecular Dynamics using Gas-Phase Ultrafast Electron Diffraction — THOMAS WOLF^{1,2}, JIE YANG¹, ●MATTHEW S. ROBINSON³, J. PEDRO F. NUNES⁴, THEODORE VECCHIONE¹, RENAKI LI¹, NICK HARTMANN¹, XIAOZHE SHEN¹, RYAN COFFEE¹, JEFF CORBETT¹, JAMES CRYAN^{1,2}, ALAN FRY¹, KELLY GAFFNEY¹, TAIS GORKHOVER¹, KARSTEN HAST¹, KAREEM HEGAZY¹, KEITH JOBE¹, IGOR MAKASYUK¹, ALEXANDER REID¹, JOSEPH ROBINSON¹, SHARON VETTER¹, FENGLIN WANG¹, STEPHEN WEATHERSBY¹, KYLE WILKIN⁵, CHARLES YONEDA¹, QIANG ZEHNG¹, MARTIN CENTURION⁵, MARKUS GÜHR^{1,2,3}, and XIJIE WANG¹ — ¹SLAC, Menlo Park, USA — ²PULSE, Menlo Park, USA — ³Physik und Astronomie, Potsdam, Germany — ⁴Chemistry, York, UK — ⁵Physics and Astronomy, Lincoln-Ne, USA.

With its ability to provide direct structural information of molecules and how they change in an excited state, Ultrafast Electron Diffraction (UED) is becoming one of the most useful techniques in ultrafast science.[1] At the forefront of this are the MeV gas-phase UED experiments carried out at SLAC, which have observed molecular wavepackets using diffraction methods with unprecedented sub-Ångstrom spatial, and 100 fs temporal, resolution.[2,3] In this talk I will discuss several UED experiments, including the study of N₂ rotational wavepackets, I₂ vibrational wavepackets, and the recent investigation of the ring-opening process of stilbene oxide. 1 - Sciaini, G. et al. Rep. Prog. Phys. 74, 96101, 2011. 2 - Yang, J. et al., Nat. Commun. 7, 11232, 2016. 3 - Yang, J. et al., Phys. Rev. Lett. 117, 153002, 2016.

MO 24.2 Fri 10:45 PA 2.150

Electronic wave packet interferometry in the UV - first steps towards coherent time-domain XUV spectroscopy — ●ANDREAS WITUSCHEK¹, LUKAS BRUDER¹, ULRICH BANGERT¹, MARCEL BINZ¹, LARS-STEPHAN KLEIN¹, TIM LAARMANN^{2,3}, and FRANK STIENKEMEIER¹ — ¹Universität Freiburg, Physikalisches Institut, Hermann-Herder-Str. 3, 79104 Freiburg, Germany — ²Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany — ³The Hamburg Centre for Ultrafast Imaging CUI, Luruper Chaussee 149, 22761 Hamburg, Germany

Coherent time-resolved spectroscopy is a powerful tool to study ultrafast dynamics in complex systems. Extending this to the XUV spectral region is on the frontier of nonlinear spectroscopy. However, demands on interferometric stability increase when going to short wavelengths and advanced pulse manipulation in the XUV is challenging. In seeded free electron lasers (FEL) the emitted XUV pulses inherit the coherence properties of the seed pulses [1]. This motivates our approach based on a stable and transportable platform performing acousto-optical phase modulation on the seed laser with subsequent seeding of the FEL and lock-in detection at the harmonics of the seed modulation [2]. This reduces demands on interferometric stability and signals are efficiently isolated and amplified [3]. The platform has been characterized on dilute sodium vapor, which allowed us to record electronic quantum beats at 268nm.

[1] Gauthier et al., PRL 116, 024801 (2016), [2] Bruder et al., Opt. Express 25, 5302-5315 (2017), [3] Bruder et al., PRA 92, 053412 (2015)

MO 24.3 Fri 11:00 PA 2.150

Electron cooling of bunched ion beams and experimental program of the CSR electron merged beam setup — ●PATRICK WILHELM, ARNO BECKER, CLAUDE KRANTZ, JORRIT LION, SVENJA LOHMANN, OLDŘICH NOVOTNÝ, MARIUS RIMMLER, SUNNY SAURABH, STEPHEN VOGEL, and ANDREAS WOLF — Max Planck Institute for Nuclear Physics, Heidelberg, Germany

In the Cryogenic Storage Ring (CSR) molecular ions are stored for hundreds to thousands of seconds in a low-temperature radiation field given by the black-body radiation of the beamline wall at less than 10 K. By this stored molecular and cluster ions can undergo radiative relaxation toward their ro-vibrational ground-state. Measurements probing fundamental molecular and cluster dynamics can be performed with good internal state definition. For ion-beam phase-space cooling a low-energy electron cooler was set up for the CSR. Cold electrons are produced by a LN₂-cooled GaAs photocathode. Merging of the electrons with the stored ion beam is realized by a magnetic guiding field which is created by a set of high-temperature superconducting

solenoids, toroids and racetrack coils. The CSR electron cooler is designed to achieve an average kinetic electron-beam energy as low as 1 eV at a rest-frame energy spread of about 1 meV. It was successfully taken into operation in June 2017. A ~40 eV electron beam was merged with a 1200 keV F⁶⁺ ion beam and bunched-beam electron cooling has been demonstrated. Upcoming beamtimes will aim at phase-space cooling and high-resolution studies of electron-ion reactions.

MO 24.4 Fri 11:15 PA 2.150

Optimization of field-free alignment of molecules for imaging experiments — ●EVANGELOS THOMAS KARAMATSKOS^{1,2}, SEBASTIAN RAABE³, PHILIPP STAMMER³, ANDREA TRABATTONI¹, TERENCE MULLINS¹, GILDAS GOLDSZTEJN³, SEBASTIAN TRIPPEL^{1,4}, ARNAUD ROUZÉE³, and JOCHEN KÜPPER^{1,2,4,5} — ¹CFEL, DESY, Hamburg, Germany — ²Department of Physics, Universität Hamburg, Germany — ³Max-Born Institute, Berlin, Germany — ⁴CUI, Universität Hamburg, Germany — ⁵Department of Chemistry, Universität Hamburg, Germany

Field-free alignment and orientation of molecules is an important prerequisite for many gas phase imaging experiments. It allows to access information directly in the molecular frame and extract observables such as internuclear distances, angles and the electronic density through a measurement of the photoelectron angular distribution. We present new experimental results and simulations on strongly field-free aligned molecules, ranging from linear to complex asymmetric rotor molecules. Additionally, photoelectron momentum distributions of strongly aligned OCS molecules, recorded experimentally at different angles between the laser polarization axis and the molecular axis will be presented. The photoelectron angular distributions display large modifications with the molecular axis distribution, both at low and high kinetic energies, which encode the molecular structure and electronic density of the molecule. A discussion of the experimental results will be presented.

MO 24.5 Fri 11:30 PA 2.150

Coherent anti-stokes Raman spectroscopy in gas-filled hollow-core PCF — ●RINAT TYUMENEV¹, BARBARA TRABOLD¹, AMIR ABDOLVAND², LUISA SPAETH¹, and PHILIP ST J RUSSEL¹ — ¹Max-Planck-Institut für die Physik des Lichts Staudtstraße 2 91058 Erlangen — ²Nanyang Technological University

Gas sensing and composition analysis are of great importance in numerous fields, for example environmental studies of greenhouse gases and toxic substances, medicine for detection of marker gases in human breath, chemical and industrial applications. Raman spectroscopy provides quantitative molecule-specific measurement of analytes. However, some analytes, for example gases, have relatively weak Raman gain and at low concentrations free space Raman spectroscopy yields tiny signal levels, which require very powerful pump lasers to be detectable. We present an approach that relaxes the need for high laser power and provides broadband Raman spectroscopy of gas mixtures. The approach is based on coherent anti-Stokes Raman spectroscopy in gas filled hollow-core photonic crystal fibre (PCF). The hollow core provides long interaction lengths allied to core diameters of a few tens of micrometers, dramatically increasing the CARS signal. The ability to change the dispersion of the waveguide by altering the gas pressure or (less practically) the PCF geometry permits CARS phase-matching over a broad wavelength range, so that the Raman signal can build up coherently over the whole fiber length. We report broadband (4000 1/cm) single-shot CARS with a concentration detection limit of 300 ppm.

MO 24.6 Fri 11:45 PA 2.150

Generation, Shaping, and Complete Characterization of Complex Mid-Infrared-fs Laser Pulses — ●MARKUS ALEXANDER JAKOB^{1,2}, MARK PRANDOLINI³, ROBERT RIEDEL⁴, GIULIO MARIA ROSSI^{2,5}, MICHAEL SCHULZ⁴, and TIM LAARMANN^{1,2} — ¹The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, 22761 Hamburg — ²Deutsches Elektronen-Synchrotron, 22607 Hamburg — ³Institut für Experimentalphysik, Universität Hamburg — ⁴Class 5 Photonics GmbH, 22607 Hamburg — ⁵Center for Free-Electron Laser Science, 22607 Hamburg

Shape-optimized Mid-IR laser pulses at a central wavelength of around $10\ \mu\text{m}$ represent a genuine tool to study coupling of different vibrational modes in complex molecular systems, as well as to manipulate charge and energy transport mechanisms and might even be used to influence catalytic behaviour. A scheme will be presented, that enables for generation of wavelength tunable and CEP stable Mid-IR laser pulses, their arbitrary shaping by means of an acousto-optical modulator, and their full characterization. These provide an excellent toolbox to address specific vibrational modes in large molecular systems and to influence and steer associated dynamics in the electronic ground state. This is where chemistry typically takes place.

MO 24.7 Fri 12:00 PA 2.150

Molecular frame 3D momentum imaged using TimePix3 —
•RUTH LIVINGSTONE¹, MILIJA SARAJLIG², DAVID PENNICARD², MARTIN VAN BEUZekom³, BAS VAN DER HEIJDEN³, JAN VISSER³, HEINZ GRAAFSMA², ANDREI NOMEROTSKI⁴, SEBASTIAN TRIPPEL^{1,2}, and JOCHEN KÜPPER^{1,2,5} — ¹The Hamburg Center for Ultrafast Imaging, Luruper Chaussee 149, Universität Hamburg, DE — ²Hamburg

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Timepix3 is a powerful new detector for imaging photons, ions, or electrons with 1.8 ns time resolution [1]. Here, we demonstrate the capabilities of the detector by measuring carbonyl sulphide (OCS) fragment momentum vectors after strong field ionization. The full 3D momenta of the $\text{OCS}^{++} \rightarrow \text{OC}^+ + \text{S}^+$ fragmentation channel were measured in coincidence mode, allowing for the molecular frame distribution to be reconstructed. The Timepix detector was integrated into the current spectrometer setup without modification to the vacuum chamber. The resulting temporal resolution was under 3 ns, as a result of the phosphor used. The advantages of this detector over the current time resolved detection methods will be discussed, along with ways to achieve the optimum temporal resolution.

[1] A. Zhao et. al., Rev. Sci. Instrum. 88, 113104 (2017)