

MO 20: Control (joint session MO/Q)

Time: Friday 14:00–16:00

Location: f102

MO 20.1 Fri 14:00 f102

Control of molecular alignment using tailored picosecond laser pulses — ●STEFANIE KERBSTADT¹, EMIL ZAK¹, ANDREY YACHMENEV¹, SEBASTIAN TRIPPEL^{1,2}, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Center for Ultrafast Imaging, Universität Hamburg — ³Department of Physics, Universität Hamburg

The control of molecular axes is essential for fundamental studies of molecular structure and of chemical reactions. Strong non-resonant laser fields have been established as powerful tool to control and to fix molecular axes in space, providing access to molecular-frame spectroscopy and imaging experiments. Here, we present experimental as well as theoretical results on the refined control of field-free molecular alignment and angular-momentum alignment by employing shaped picosecond pulses to a ground state-selected cold molecular beam of carbonylsulfid (OCS).

MO 20.2 Fri 14:15 f102

A comparative study on ionization-induced dissociation of hydrogen, irradiated by 800 nm and 400 nm laser fields — ●RENÉ WAGNER¹, SABA ARIFE BOZPOLAT², PATRIK GRYCHTOL¹, ILHAN YAVUZ², and MICHAEL MEYER¹ — ¹Small Quantum Systems Group, European XFEL GmbH, 22869 Schenefeld, Germany — ²Physics Department, Marmara University, 34722 Ziverbey, Istanbul, Turkey

We present a two color investigation of the hydrogen molecule to benchmark our tabletop experiment dedicated to ultrafast investigations of electronic correlations in atoms and molecules from the near infrared (NIR) to the extreme ultraviolet (EUV) range. Our setup focuses on the study of dissociating molecules by different pump-probe techniques aiming to obtain invaluable and novel insights into atomic and molecular dynamics. For this purpose, we have built a femtosecond laser driven EUV source based on high harmonic generation (HHG) combining it with a pulsed molecular jet, a delay-line based velocity map imaging (VMI) detector and a time-of-flight (TOF) spectrometer. We are going to show first experimental as well as theoretical results quantifying the performance of our apparatus having captured and analysed the angular ion momentum distribution of the photo-induced dissociation process of hydrogen, irradiated by ultrafast 800 nm and 400 nm laser fields.

[1] Ibrahim *et al.*, J. Phys B: At. Mol. Opt. Phys. **51** (2018) 042002.

MO 20.3 Fri 14:30 f102

Electron-vibrational coupling dynamics in SF₆ — ●PATRICK RUPPRECHT¹, LENNART AUFLEGER¹, ALEXANDER MAGUNIA¹, SIMON HEINZE², THOMAS DING¹, MARC REBHOLZ¹, STEFANO AMBERG¹, NIKOLA MOLLOV¹, FELIX HENRICH¹, MAURITS HAVERKORT², CHRISTIAN OTT¹, and THOMAS PFEIFER¹ — ¹Max-Planck-Institut für Kernphysik, Heidelberg — ²Universität Heidelberg, Institut für Theoretische Physik

Visible and infrared vibrational spectroscopy, probing non-localized electronic molecular states, is commonly used in chemistry and biology. In combination with core-level spectroscopy, studies of coherently coupled electronic and vibrational dynamics with site and element specificity are possible. We report on the investigation of sulfur hexafluoride (SF₆) using attosecond transient absorption spectroscopy driven by mJ-level, few-cycle 15 fs FWHM pulses centered at 1550 nm in the short-wave infrared (SWIR) spectral region. The excited 6a_{1g}, 2t_{2g} and 4e_g molecular states related to the sulfur L_{2,3} absorption edge were probed. First, altering the absorption spectrum in the 160 eV to 200 eV soft X-ray (SXR) region under the presence of a strong SWIR field was demonstrated. Furthermore, varying the delay between the SXR and SWIR pulses resulted in an oscillatory behavior of the resonance lines' intensities, with a leading SWIR pulse. The extracted oscillation period of (773 ± 16) cm⁻¹ matches the Raman-active symmetric breathing mode $\nu_1 = 775$ cm⁻¹. This result implies sensitivity to nonresonant impulsive stimulated Raman scattering via probing electronic transitions to states localized near the sulfur atom.

MO 20.4 Fri 14:45 f102

Laser-induced alignment of nanoparticles — ●HAKAN AKARSU^{1,2}, MUHAMED AMIN¹, LENA WORBS^{1,2}, JANNIK LÜBKE^{1,2,3},

ARMANDO ESTILLORE¹, AMIT K. SAMANTA¹, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen Synchrotron DESY, Hamburg — ²Fachbereich Physik, Universität Hamburg — ³Center for Ultrafast Imaging, Universität Hamburg

Single-particle imaging (SPI) experiments at free-electron lasers (FELs) promise high-resolution-imaging of the structure and dynamics of nanoparticles. By guiding sample molecules in the gas phase into the x-ray focus of an FEL, diffraction patterns of individual particles can be collected. Sufficient amounts of patterns of identical nanoparticles are needed to overcome the inherently small signal-to-noise ratio and reconstruct the underlying 3D structure [1]. Laser-induced alignment of nanoparticles has potential to improve the achievable resolution and to push it toward the atomic scale [2,3].

Here, we present simulation results on laser-induced alignment of gold nanorods by integrating the Euler equation of rotational motion. We have also tested different laser pulse profiles for improvement of the alignment. In addition, we will present first experimental investigations of the laser-induced alignment of gold nanorods.

[1] M. M. Seibert, *et al.*, Nature **470**, 78 (2011)

[2] J. C. H. Spence, *et al.*, Phys. Rev. Lett. **92**, 198102 (2004)

[3] J. Küpper, *et al.*, Phys. Rev. Lett. **112**, 083002 (2014)

MO 20.5 Fri 15:00 f102

Rotational spectroscopy of molecular superrotors: probing high rotational states of O₂ via REMPI technique combined with an optical centrifuge. — ●AUDREY SCOGNAMIGLIO¹, JORDAN FORDYCE², IAN MACPHAIL-BARTLEY², KATRIN DULITZ¹, FRANK STIENKEMEIER¹, and VALERY MILNER² — ¹Institut of Physics, University of Freiburg, Germany — ²Department of Physics and Astronomy, University of British Columbia, Canada

To access highly excited rotational states (e.g. N=100 in O₂), the use of an optical centrifuge has been demonstrated to be a powerful and unique experimental tool.

To start from a well-defined initial ro-vibrational state, oxygen molecules are cooled to 10K by means of supersonic expansion. Due to the relatively low density, spectroscopic techniques combining high frequency resolution and high sensitivity are required. For this purpose, resonance enhanced multiphoton ionization of oxygen is employed in a "2+1" scheme with a two-photons transition from the ground state $X^3\Sigma_g^- (\nu = 0)$ to the excited state $C^3\Pi_g (\nu = 2)$ from which another photon ionizes the molecule.

In this contribution, experimental results of REMPI spectroscopy of oxygen superrotors, as well as their numerical analysis, will be presented.

MO 20.6 Fri 15:15 f102

Setting the basis for a good Carrier Envelope Phase control — ●FRANZISKA SCHÜPPEL, THOMAS SCHNAPPINGER, and REGINA DE VIVIE-RIEDEL — Department Chemie, Ludwig-Maximilians-Universität München

Controlling the process of photo reactions is a major goal in chemistry. For synthetic application, for example, it is significant to have the desired product in the best possible yield. To achieve this, one approach is to apply a few-cycle IR laser pulse to a system in the vicinity of a conical intersection (CoIn).[1] This influences the system in a way to build a superposition between the states of interest. By changing the CEP (Carrier Envelope Phase) of the laser pulse, the superposition can be controlled to lead to the preferred transfer through the CoIn and by that to the preferred product.

In this theoretical work, we want to investigate the effectiveness of a CEP control on the basis of a dissociative model system. For that, we use quantum dynamic simulations with two coupled states in the adiabatic representation. We want to describe the interaction of the laser pulse, the transition dipole moment and the non-adiabatic coupling on the CEP control and find the basis needed for an effective control of a system. By changing the shape of the CoIn of the model system, we want to test the influence of a different topography on the CEP control.

[1] P. von den Hoff *et al.*, IEEE J SEL TOP QUANT, **18** (2012), 119-129.

MO 20.7 Fri 15:30 f102

Controlling the nuclear- and electron-dynamics at a conical intersection — •THOMAS SCHNAPPINGER, FRANZISKA SCHÜPPEL, and REGINA DE VIVIE-RIEDLE — Department of Chemistry, LMU Munich, Germany

The combination of ultra-fast optical techniques with quantum dynamics simulations give extensive insights into the nuclear and electronic dynamics of molecules and give rise to the possibility of modifying or even controlling the dynamics.

In this theoretical work we aim to control the coupled nuclear- and electron-dynamics in the vicinity of a conical intersection (CoIn). The control scheme relies on the carrier envelope phase (CEP) of a few-cycle IR pulse. The laser interaction creates an electronic superposition of the involved states before the wavepacket reaches the CoIn and influences the population transfer through the CoIn. To simulate the coupled nuclear- and electron-motion of this process we are using the NEMol (coupled nuclear- and electron-dynamics in molecules) ansatz developed in our group. In this purely quantum mechanical ansatz the quantum-dynamical description of the nuclear motion is combined with the calculation of the electron-dynamics in the eigenfunction basis.

We want to show two examples the molecule NO₂ and the nucleobase uracil. Both systems show relaxation dynamics back in the ground state via a CoIn after photoexcitation. But the circumstances, e.g. the transitional dipole moment and the localization of the wavepacket, differ significantly in both systems. These facts should be reflected in

the controllability of the relaxation.

MO 20.8 Fri 15:45 f102

Laser induced electron diffraction in the molecular frame — •JOSS WIESE^{1,2}, JOLIYN ONVLEE^{1,3}, ANDREA TRABATTONI^{1,3}, EVANGELOS KARAMATSKOS^{1,4}, SEBASTIAN TRIPPEL^{1,3}, and JOCHEN KÜPPER^{1,2,3,4} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Chemistry, Universität Hamburg — ³The Hamburg Center for Ultrafast Imaging, Universität Hamburg — ⁴Department of Physics, Universität Hamburg

Laser-induced electron diffraction (LIED) has the potential to provide time-dependent images of molecules at sub-femtosecond and few-picometer resolution and is therefore ideally suited to record quantum molecular movies. Here we present our work on LIED and strong field ionization off strongly aligned molecular samples. Our samples include simple linear molecules like OCS as well as prototypical biomolecules such as indole and its microsolvated clusters. Effects of the overall strong-field recollision dynamics on the orientation of the molecules will be presented and compared with time dependent density-functional theory (TDDFT) simulations as well as with novel highly efficient semiclassical simulations based on the adiabatic tunneling theory. Our findings have strong impact on the interpretation of self-diffraction experiments, where the photoelectron momentum distribution is used to retrieve molecular structures.