

A 43: Ultrafast Dynamics III and High-harmonic Generation (joint session MO/A)

Time: Friday 14:30–16:30

Location: HS 3044

A 43.1 Fri 14:30 HS 3044

Absolute photoemission timing in neon — ●MAXIMILIAN FORSTER, MAXIMILIAN POLLANKA, CHRISTIAN SCHRÖDER, and REINHARD KIENBERGER — Chair for laser and x-ray physics, E11, Technische Universität München, Germany

We measure the relative photoemission time delay between the Ne2p, Ne2s and the Iodine 4d states in iodomethane utilizing attosecond streaking. This allows us to experimentally determine the absolute time delay of neon 2s and 2p photoelectrons for the first time. The delay of neon, being the first ever evidence of atomic delay, has received repeated attention by both experimental and theoretical investigations due to the large cross section and convenient properties of neon. While helium has been the gold standard for absolute time delay measurements, enabled by remarkable theoretical agreement, due to spectral overlap helium cannot be used to reference neon. Recent developments, namely the availability of different chronoscopes, enable measuring the absolute time delay of neon. We take the path via iodomethane and the I4d core state, which has been timed on an absolute scale, and use it to reference neon. The delay between Ne2s and Ne2p can be extracted simultaneously, allowing for a positive consistency check with previous experiments conducted only with neon. Timing neon on an absolute scale allows an assignment of absolute values to these experiments in retrospect and establishes neon as a chronoscope species.

A 43.2 Fri 14:45 HS 3044

Isosteric molecules in the time-domain — ●MAXIMILIAN POLLANKA, CHRISTIAN SCHRÖDER, MAXIMILIAN FORSTER, and REINHARD KIENBERGER — Physik Department, Technische Universität München, James-Frank-Str. 1, 85748 Garching, Germany

We report on absolute photoemission timing measurements on isosteric molecules in the gas phase. Photoemission time delays are accessed via streaking spectroscopy on attosecond timescales. To be able to (directly) access absolute photoemission times of the respective outer and inner valence states of N₂O and CO₂ we are using iodomethane (I4d) as a timing reference. In a complementary study He was used as reference to cross-check the results as well as to verify the usability of the respective chronoscope species. Due to the similarities in molecular structure (isostericity) and electronic configurations (isoelectronicity) between these investigated molecules, the pure effect of the specific molecular/orbital characteristics is expected to be probed. Additionally, N₂ and CO is studied in the same way on the basis of their isosteric behavior. The experimental data show great similar tendencies but also differences between the compared molecular orbitals, which are determined but not completely understood up to now. Nonetheless, recent theoretical calculations hint towards an additional channel coupling photoemission time delay contribution that can be assigned to electron correlations responsible for re-disturbing the excitation among different final photoionization channels.

A 43.3 Fri 15:00 HS 3044

Attosecond time-resolved coincidence spectroscopy of ethylene — ●BARBARA MERZUK¹, DAVID BUSTO^{1,2}, IOANNIS MAKOS¹, DOMINIK ERTEL¹, MARVIN SCHMOLL¹, BENJAMIN STEINER¹, FABIO FRASSETTO³, LUCA POLETTI³, ROBERT MOSHAMMER⁴, CLAUS DIETER SCHRÖTER⁴, THOMAS PFEIFER⁴, SERGUEI PATCHKOVSKI⁵, JAKUB BENDA⁶, ZDENĚK MAŠÍN⁶, and GIUSEPPE SANSONE¹ — ¹Albert-Ludwigs-Universität Freiburg, Germany — ²Lund University, Sweden — ³CNR, Padova, Italy — ⁴MPIK, Heidelberg, Germany — ⁵MBI Berlin, Germany — ⁶Charles University, Prague, Czech Republic

Studying photoionization dynamics and characterising the time delays associated with the photoemission of an electron wave packet can unveil important characteristics of coupled electronic-nuclear dynamics in molecular systems. Attosecond photoelectron spectroscopy in combination with electron-ion coincidence detection is beneficial since this allows disentangling the different photoionization and dissociation channels. Additionally, it may give access to the orientation of the molecule at the instant of photoionization. Using our experimental setup that consists of an attosecond beamline, based on high-order harmonic generation operating at 50 kHz repetition rate, we investigate the photoionization dynamics in ethylene molecules by performing RABBIT (Reconstruction of Attosecond Beating By Interference of Two-photon transitions) measurements while detecting photoelectrons

and photoions in coincidence. The experimental results are interpreted with the help of multi-electron R-matrix calculations of two-photon ionization.

A 43.4 Fri 15:15 HS 3044

Probing well aligned molecular environments on surfaces via attosecond streaking — ●PASCAL SCIGALLA¹, SVEN PAUL¹, CHRISTIAN SCHRÖDER¹, PETER FEULNER², and REINHARD KIENBERGER¹ — ¹Chair for laser and x-ray physics, E11, Technische Universität München, Germany — ²Surface and Interface Physics, E20, Technische Universität München, Germany

We report on the photoemission timing measurements of well-aligned iodomethane and -ethane molecules on a Pt111 surface. In this set of experiments, we clock the I4d photoemission of iodine against the Platinum valence photoemission using the attosecond streak camera technique, allowing the extraction of a relative photoemission delay. As the I4d photoemission in the selected energy range is dominated by a giant resonance in the I4d → ef channel, its photoemission time is mostly unaffected by its chemical environment; thus, any observed change in the photoemission delay can be attributed to the traversed potential landscape of the molecule. By carefully selecting the detection angle and crystal surface coverage we can reliably choose whether only parts of the molecule or its entirety was traversed by the detected photoelectron wavepackets. It is furthermore possible to investigate the influence of slight coverage variations onto the observed photoemission delay. Planned, complementary scattering simulations will be used to gain deeper insight into the observations with the goal to establish photoemission timing experiments as an efficient and accurate means to study molecular environments on surfaces.

A 43.5 Fri 15:30 HS 3044

Automatic optimization of intense high-harmonic pulses — ●JOSÉ GÓMEZ TORRES, FREDERIC USSLING, SIMON WÄCHTER, ALESSANDRO COLOMBO, LINOS HECHT, KATHARINA KOLATZKI, ALEXANDRE ROSILLO VORSIN, MARIO SAUPPE, and DANIELA RUPP — ETH Zurich, Laboratory for Solid State Physics, John-von-Neumann-Weg 9, 8093 Zurich, Switzerland

High harmonic generation (HHG) allows the production of extreme ultraviolet pulses ranging from picosecond up to attosecond timescales from intense infrared (IR) pulses, making it an invaluable tool for the study of ultrafast phenomena. It has been recently demonstrated that HHG is capable of producing pulses intense enough for diffraction experiments like coherent diffraction imaging of isolated nanoparticles [1]. Very intense pulses of short time duration in a stable delivery over hours are necessary for this, requiring a time-consuming optimization of the experimental parameters. We developed a tool for the automatic optimization of HHG parameters, sweeping different geometric parameters of the setup and measuring for each step the pulse energy achieved. Due to the complexity of simulating the specific conditions of the experiment, this trial and error approach is a necessary final step to achieve the highest pulse energy. In order to optimize the XUV peak focal intensity, we perform electron spectroscopy on a diffuse gas in the focus region. Via IR-XUV pump probe, RABBIT measurements can be carried out for the temporal characterization of pulses.

[1] D. Rupp et al., Nature Communication 8, 493 (2017)

A 43.6 Fri 15:45 HS 3044

Orbital interference effects in low-order harmonic generation in benzene — ●SAMUEL SCHÖPA, FALK-ERIK WIECHMANN, FRANZISKA FENNEL, and DIETER BAUER — Universität Rostock, Rostock, Germany

We explore the impact of the driving laser's ellipticity and polarization on the low-order harmonic spectrum of benzene and find a strong interference in the 5th harmonic between emission originating from transitions between π orbitals and emission from σ orbitals. The contribution of the π orbitals entirely vanishes due to interference for driving with a laser polarized along a σ_v mirror axis. However, the π orbital's contribution takes over for elliptic polarization while being fundamentally different from the σ orbital emission, i.e., having the opposite helicity and a perpendicular major polarization axis. The resulting interference yields a complex dependence of the low-order harmonic spectrum of benzene on the ellipticity and the polarization

of the driving field.

A 43.7 Fri 16:00 HS 3044

Observation of HHG from organic molecular crystals
— ●FALK-ERIK WIECHMANN¹, SAMUEL SCHÖPA¹, ALEXANDER VILLINGER², DIETER BAUER¹, and FRANZISKA FENNEL¹ — ¹Institute of Physics, Rostock, Germany — ²Institute of Chemistry, Rostock, Germany

This project aims at a detailed understanding of the harmonic generation process in large organic molecules in the crystalline phase. Unlike previous studies, which were limited to small molecules in the gas phase, we introduce organic molecular crystals as a novel target for HH spectroscopy, taking advantage of the inherent molecular alignment. Unlike in gas phase experiments, neighboring molecules in organic crystals experience a weak but finite coupling, leading to 'solid like' features, e.g. a delocalization of the electronic states over several unit cells. With a fundamental 4000 nm mid-IR beam reaching 6 TW/cm² we demonstrate that HHG up to the order of 17 is possible without imposing physical damage. When the fundamental driving polarization is rotated, maxima of harmonic emission occur at polarization directions parallel to connecting axes between neighboring molecules, reflecting the crystal structure. Despite the linearly polarized driving field, the emitted harmonics exhibit elliptical polarization with a main axis different from the fundamental polarization direction.

A 43.8 Fri 16:15 HS 3044

High-order Harmonic Generation (HHG) in the nonadiabatic regime over a sub-mm glass chip — ●SABINE ROCKENSTEIN^{1,2}, AGATA AZZOLIN^{1,2}, GAIA GIOVANETTI², GUANGYU FAN^{2,3}, MD SABBIR AHSAN^{2,4}, OLIVIERO CANNELLI², LORENZO COLAIZZI^{1,2,5}, ERIK P MÅNSSON², DAVIDE FACCIALÀ⁴, FABIO FRASSETTO⁴, DARIO W LODI⁵, CRISTIAN MANZONI⁴, REBECA M VÁZQUEZ⁴, MICHELE DEVATTA⁴, ROBERTO OSELLAME⁴, LUCA POLETTA⁴, SALVATORE STAGIRA^{4,5}, CATERINA VOZZI⁴, VINCENT WANIE², ANDREA TRABATTONI^{2,6}, and FRANCESCA CALEGARI¹ — ¹UHH (DE) — ²DESY (DE) — ³CUI (DE) — ⁴CNR (IT) — ⁵Politecnico di Milano (IT) — ⁶Uni. Hannover (DE)

HHG-based sources are nowadays operating up to the soft-x spectral region. One of the main challenges remains to extend the cut-off frequency while retaining high-photon flux. Approaches based on the so-called nonadiabatic regime have allowed to overcome phase matching limitations and achieve substantial cut-off extension [1]. We present a new HHG source, operating with high driver laser intensities (up to 1E16 W/cm²) and a laser-micromachined glass cell allowing for highly efficient gas confinement over 900 μm, to achieve nonadiabatic phase matching. The setup was operated with both 800-nm and 1500-nm sub-35-fs driving pulses. With the 800-nm driver, the HHG energy cutoff was extended to 100 eV in Argon and 180 eV in Neon, 160 eV were reached using the 1500-nm driver in Argon. Our results highlight the potential of optimizing the nonadiabatic regime for covering the water-window spectral region. [1] Johnson et al., Sci. Adv. 4(5), 2018