MO 2: X-ray FELs (joint session MO/A)

Time: Monday 14:00–16:15 Location: MO-H5

MO 2.1 Mon 14:00 MO-H5

Following excited-state chemical shifts in molecular ultrafast x-ray photoelectron spectroscopy — •Dennis Mayer[1], Fabiano Lever[1], David Picconi[1], Jan Metje[1], Skirmantas Alisauskas[2], Francesca Calegari[3], Stefan Düsterer[2], Christopher Ehlert[4], Raimund Feifel[5], Mario Niebuhr[1], Bastian Manschwetus[2], Marion Kuhlmann[2], Tomaso Mazza[6], Matthew S. Robinson[1,3], Richard J. Squibb[5], Andrea Trabattoni[3], Mans Wallner[5], Peter Saalfrank[1], Thomas J.A. Wolf[7], and Markus Gühr[1] — [1]University of Potsdam, Germany — [2]DESY, Hamburg, Germany — [3]CFEL, Hamburg, Germany — [4]HITS gGmbH, Heidelberg, Germany — [5]University of Gothenburg, Sweden — [6]European XFEL GmbH, Hamburg, Germany — [7]Stanford PULSE Institute, Menlo Park, USA

We demonstrate the capabilities of time-resolved x-ray photoelectron spectroscopy with a study of the UV-excited dynamics of 2-thiouracil conducted at the FLASH free electron laser in Hamburg, Germany. By probing sulfur 2p core electrons, we discover that a significant part of the excited-state population relaxes to the ground state within 220-250fs. Observed spectral shifts can be directly attributed to a charge redistribution over the molecule during the relaxation process. Additionally, we observe a 250fs oscillation in the kinetic energy of the excited-state population which reveals a coherent population exchange among electronic states.

MO 2.2 Mon 14:15 MO-H5

How to produce nuclear-polarized hydrogen molecules and for what they can be used — •Ralf Engels — Institut für Kernphysik, FZ Jülich/GSI Darmstadt

In accelerator experiments polarized proton/deuteron beams and hydrogen/deuterium targets are an important tool to investigate the spin dependence of the nuclear forces. Both can be made with a polarized atomic beam source, a modern version of a Rabi apparatus. By recombination of these atoms hyper-polarized H_2, D_2 and HD molecules in many hyperfine substates are produced and can be used for further applications. For example, the recombination process itself and his dependence on the electron spin, surface materials or external radiation can be investigated as well as the coupling of the nuclear spins with the rotational magnetic moment. In nuclear physics the polarized molecules allow to increase the target density and with polarized molecular ions a better stripping injection into storage rings is possible. Further applications may be the use as polarized fuel for fusion reactors or the search for an electric dipole moment of the nucleons.

MO 2.3 Mon 14:30 MO-H5

Correlation fingerprints in the x-ray induced Coulomb explosion of iodopyridine — •Benoît Richard[1,2,3], Julia Schäfer[1,4], Zoltan Jurek[1], Robin Santra[1,2,3,4], and Ludger Inhester[1,2] — [1]Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany — [2]The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany — [3]Department of Physics, Universität Hamburg, Notkestr. 9-11, 22607 Hamburg, Germany — [4]Department of Chemistry, Universität Hamburg, Martin-Luther-King-Platz 6, 20146 Hamburg, Germany

Coulomb explosion induced by XFEL radiation is a promising experimental tool to image individual molecules. However, the amount of information about the original molecule geometry that can be inferred from the measured final momenta of the produced ions is presently unknown. In particular, the data acquired by state of the art multi-coincidence measurement techniques contains information about correlations between the different measured ions, but how to exploit this extra information for geometry reconstruction is currently unclear. In this work we propose a first step in this direction. To this end we analyze simulation data for the x-ray induced Coulomb explosion of 2-iodopyridine and describe its fragmentation dynamics. Crucially, we show that a collision between two ions during the Coulomb explosion causes strong and possibly measurable correlations between their final momenta.

MO 2.4 Mon 14:45 MO-H5

Universal Reconstruction of Nanoclusters from Wide-Angle X-Ray Diffraction Patterns with Physics-Informed Neural Networks — •Thomas Stielow and Stefan Scheel — Institut für Physik, Universität Rostock, Albert-Einstein-Straße 23, 18059 Rostock

Single-shot diffraction imaging by soft X-ray laser pulses is a valuable tool for structural analysis of unsupported and short-lived nanosystems, while the exact inversion of the scattering patterns still proves challenging [1]. Deep learning, on the other hand, is widely used in data sciences for the extraction of information from images and has recently been used to accelerate parameter reconstructions from wide-angle scattering patterns [2]. Here, we show how a deep neural network can be used to reconstruct complete three-dimensional object models of uniform, convex particles from single two-dimensional wide-angle scattering patterns. Through physics-informed training the reconstructions achieve unprecedented levels of detail on real-world experimental data [3].

[1] I. Barke *et al.* Nat. Commun. **6**, 6187 (2015).
[2] T. Stielow *et al.* Mach. Learn.: Sci. Technol. **1**, 045007 (2020).
[3] T. Stielow and S. Scheel, Phys. Rev. E **103**, 053312 (2021).

MO 2.5 Mon 15:00 MO-H5

Ultrafast Auger spectroscopy of 2-thiouracil — •F. Lever[1], D. Mayer[1], D. Picconi[1], J. Metje[1], S. Alisauskas[2], F. Calegari[2], S. Düsterer[2], C. Ehlert[3], R. Feifel[4], M. Niebuhr[1], B. Manschwetus[2], M. Kuhlmann[2], T. Mazza[6], M. S. Robinson[5], R. J. Squibb[4], A. Trabattoni[5], M. Wallner[4], P. Saalfrank[1], T. J. A. Wolf[7], and M. Gühr[1] — [1]Universität Potsdam — [2]Deutsches Elektronen Synchroton (DESY) — [3]Heidelberg Institute for Theoretical Studies — [4]Department of Physics, Gothenburg University — [5]Center for Free-Electron Laser Science (CFEL) — [6]European XFEL — [7]SLAC, Stanford

Investigating the effects of UV exposure in thionucleobases can shed light on the mechanisms that cause the formation of DNA lesions. In this talk, we show how ultrafast x-ray spectroscopy can be used to gain information on such processes. We study the sulfur 2p Auger spectrum of 2-thiouracil in a uv-pump, x-ray probe experiment at the free electron laser FLASH. We observe ultrafast dynamics in the electron kinetic energy spectrum, happening on time scales of 100fs to 1ps. Using a simple coulomb model for the electron binding energies, aided by quantum chemical calculations of the electronic states energy, we deduce an elongation of the C-S bond on a 100fs time scale. The geometric changes trigger internal conversion from the initally excited S2 state to the S1 state. For longer pump-probe delays, the observerd timescales provide evidence for inter system crossing from the S1 state to the triplet manifold [1].

[1] F Lever et al 2020 J. Phys. B: At. Mol. Opt. Phys. 54 014002

MO 2.6 Mon 15:15 MO-H5

Control of bionanoparticles with electrical fields — •Jannik Lübke[1,2,3], Lena Worbs[1,2], Armando Estillore[1], Amit Samanta[1], and Jochen Küpper[1,2,3] — [1]Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [2]Department of Physics, Universität Hamburg, Hamburg, Germany — [3]Center for Ultrafast Imaging, Universität Hamburg, Hamburg, Germany

Single-particle imaging (SPI) experiments at free-electron lasers (FELs) promise high-resolution imaging of the structure and dynamics of nanoparticles and macromolecules. Guiding sample particles into the 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. Optimized delivery of identical nanoparticles is key to efficient and successful SPI experiments. Here, we present an approach for the production of purified high-density beams of a broad variety of biological nanoparticles, demonstrated on a large protein. We establish control through electric fields, aiming at charge state or conformational state selectivity. This is especially relevant for soft biological samples, such as proteins or protein complexes, which in uncontrolled environment are prone to structural instability.

MO 2.7 Mon 15:30 MO-H5

Tracing Inner-Shell-Ionization-Induced Dynamics of Water Molecules Using an X-ray Free-Electron Laser and Ab-

Initio Simulations — •Ludger Inhester[1], Till Jahnke[2], Renaud Guillemin[3], and Maria Novella Piancastelli[3,4] — [1]Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — [2]European XFEL, Schenefeld — [3]Sorbonne Université, CNRS, LCPMR, Paris — [4]Uppsala University, Uppsala

The response of molecules to ionizing radiation is of utmost relevance to many research areas. Multi-coincidence signals from experiments at x-ray free-electrons lasers provide us new opportunities to study the dynamics of molecules upon inner-shell ionization. In a recent experiment at the European XFEL, water vapor has been exposed to intense x-ray pulses and all the resulting ion fragments have been recorded in coincidence. In this talk, I will discuss how through ab-initio simulations of the multiphoton multiple ionization and fragmentation dynamics we could identify distinct signatures in the ion momentum data with different break-up patterns. By combining experimental results and theoretical modeling, we were able to image the dissociation dynamics of water after core-shell ionization and subsequent Auger decay in unprecedented detail and uncover fundamental dynamical patterns relevant for the radiation damage in aqueous environments. [1]

[1] T. Jahnke et al., Phys. Rev. X 11, 041044 (2021)

MO 2.8 Mon 15:45 MO-H5

Competition of interatomic Coulombic decay and autoionization in doubly excited helium nanodroplets — •Björn Bastian, Jakob D. Asmussen, Ltaief B. Ltaief, Akgash Sundaralingam, Catharina I. Vandekerckhove, and Marcel Mudrich — Department of Physics and Astronomy, Aarhus University, DK

Double-excitation states in helium atoms are an important model system to study electron-electron correlation. Doubly excited atoms can autoionize and the interference with the direct ionization pathway gives rise to characteristic Fano peaks in the photoexcitation spectrum [1] which has also been observed in helium nanodroplets [2]. In dimers or clusters, the de-excitation energy can instead be transferred and cause ionization of the environment. Theory has shown, that this interatomic Coulombic decay (ICD) pathway becomes fast at small interatomic distances and competes with autoionization especially in large environments [3].

We present photoion-photoelectron coincidence spectra around the Fano resonance below the N=2 ionization threshold in helium nanodroplets that have been recorded at our new endstation at the AMO-Line of the ASTRID2 synchrotron at Aarhus. Slow electrons reveal ICD or secondary inelastic scattering. Highly resolved electron spectra recorded at various photon energies across the Fano resonance reveal the details of the decay process.

[1] Domke et al. *Phys. Rev. Lett.* **66**, 1306 (1991). [2] LaForge et al. *Phys. Rev. A* **93**, 050502 (2016). [3] Jabbari et al. *Chem. Phys. Lett.* **754**, 137571 (2020).

MO 2.9 Mon 16:00 MO-H5

Simulating Molecular Diffraction Patterns using CMIdiffract — •Nidin Vadassery[1,3], Sebastian Trippel[1,2], and Jochen Küpper[1,2,3] — [1]Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — [2]Department of Physics, Universität Hamburg — [3]Department of Chemistry, Universität Hamburg

The structure and time-dependent dynamics of molecules in the gas phase reveal a plethora of information about fundamental processes in nature. X-rays and electrons are typically used to image the molecular structure using diffraction techniques. In that respect, x-ray pulses provided by XFELs have the potential to study the chemical dynamics of gaseous molecules on the ultrafast time scale with sub-picometer spatial resolution. Here, we present our computational results using CMIdiffract, an in-house software package developed to compare experimental diffraction images with theory. The package incorporates various aspects of x-ray diffraction experiments, e.g., angular distributions of molecular samples.