MO 5: Ultrafast Electronic Dynamics

Time: Monday 17:00–18:45 Location: P 204

Invited Talk MO 5.1 Mon 17:00 P 204

Charge migration in aromatic amino acids — •Sabine Rockenstein for the AMINO-Collaboration — Deutsches Elektronen-Synchrotron DESY

Aromatic amino acids play pivotal roles in protein structure and function. Their prompt ionization can trigger an ultrafast charge motion between the various functional groups. At the Linac Coherent Light Source (LCLS), the enhanced self-amplified spontaneous emission (eSASE, XLEAP) configuration allows attosecond-resolved x-ray absorption spectroscopy of such processes with unprecedented time resolution. Site-specific measurements in a $\omega/2\omega$ XLEAP scheme at the oxygen and nitrogen K edge, exploiting the high data rate available from LCLSII, enable studies of electronic dynamics and electron-nuclear coupling. This technique has been applied to phenylalanine and tryptophan, which, in previous studies with table-top attosecond sources, showed remarkably different dephasing times despite structural similarity. We discuss the experimental methods and the preliminary analysis of the collected data.

MO 5.2 Mon 17:30 P 204

Ultrafast charge transfer dynamics in phosphorylated amino acids in aqueous solution — •Nicolas Velasquez[1], Juliette Leroux[2], and Florian Trinter[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6 14195, Berlin, Germany — [2]Department of Physics, Universität Hamburg, 22607 Hamburg, Germany

Charge transfer (CT) plays a fundamental role in chemistry and biology, particularly in aqueous environments where most reactions occur. Post-translational modifications such as phosphorylation strongly alter local charge distribution and intermolecular interactions in proteins. In this study, we employ Auger–Meitner spectroscopy to address ultrafast CT dynamics in phosphorylated amino acids in aqueous solution, focusing on the role of the phosphate group at the biomolecule–water interface. Our results indicate that the phosphate group dramatically enhances charge transfer to the solvent (CTTS). This finding highlights the role of solvation and, in particular, hydrogen-bond network effects in facilitating CTTS. Furthermore, the core-hole lifetime provides a natural timescale for electron dynamics, allowing quantification of CT rates. Our observations suggest that CT pathways are governed by the interplay between the phosphate moiety and the amino-acid backbone in the hydrated environment. Understanding CT in biomolecules is crucial for fields such as radiation chemistry, protein chemistry, and biophysics. Our findings contribute to a deeper understanding of early electronic processes in complex systems, paving the way for future studies of radiation-induced damage in biologically relevant conditions.

MO 5.3 Mon 17:45 P 204

Low dispersion, phase-modulated rapid-scanning interferometry in the deep UV spectral region — •Felix Selz, Ulrich Bangert, Fabian Richter, Felix Riedel, Nils-Oliver Schütz, and Lukas Bruder — Institute of Physics, University of Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

Stable interferometer schemes for the deep UV spectral region and ultrashort pulse applications remain limited. Conventional spectrometers typically lack the required phase stability, whereas common-path interferometers introduce substantial temporal dispersion. We have recently developed a low-dispersion interferometric approach that combines high phase stability with excellent sensitivity using an efficient rapid scanning lock-in detection scheme [1]. We are now extending this scheme into the deep UV spectral region with particular emphasis on minimizing material dispersion to ensure compatibility with sub 10-fs laser pulses. This opens up Fourier transform and coherent multidimensional spectroscopy with high temporal and spectral resolution in the deep UV domain.

[1] F. Richter et al., Opt. Lett. 50, 3668-3671 (2025)

MO 5.4 Mon 18:00 P 204

New insight into photoionization by attosecond spectroscopy of isosteric molecules — •Maximilian Pollanka[1], Maximilian Forster[1], Sven-Joachim Paul[1], Zdenek Masin[2], Jakub Benda[2], and Reinhard Kienberger[1] — [1]School of Natural Sciences, Chair for Laser- and X-ray physics, Technische Universität München, Garching, Germany — [2]Institute of Theoretical Physics, Faculty of Mathematics and Physics, Charles University, Prague, Czechia

Our work addresses time-resolved photoemission in molecules. To this end, the four isosteric di-and tri-atomic molecules CO, N2, CO2, and N2O are investigated in complementary studies and compared with each other. By using suitable chronoscopes in combination with the attosecond streaking spectroscopy method, it is possible not only to determine relative photoemission times between different electronic states but also to access absolute photoemission delays. Together with theoretical considerations and simulations, the experimentally obtained results can be discussed and explained. This measurement method is applied to four molecules that are almost identical in their electronic states, molecular orbitals, and number of outer valence electrons, which is described by the term isosterism. The chronoscopes iodomethane (I4d state) and helium (He1s state) are not only used as a reference to gain access to the absolute photoemission times, but the results obtained in separate experiments are also used to verify the validity of the unrestricted use of both. Together with theoretical support, the physical processes underlying photoemission in these isosteric molecules could finally be deciphered and interpreted.

MO 5.5 Mon 18:15 P 204

Investigating higher-excited-state properties through nonlinear order separation in two-dimensional electronic spectroscopy — •Katja Mayershofer[1], Peter A. Rose[2], Julian Lüttig[2], Luisa Brenneis[1], Simon Büttner[1], Jacob J. Krich[2,3], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, 97074 Würzburg, Germany — [2]Department of Physics, University of Ottawa, Ottawa, Ontario, Canada — [3]Nexus for Quantum Technologies, University of Ottawa, Ottawa, Ontario, Canada

Recently, we developed an implementation of coherent two-dimensional (2D) electronic spectroscopy that enables us to separate signals of different pertubative orders through intensity variation and inversion with a Vandermonde matrix [1]. We applied this new technique to investigate a squaraine dimer. This procedure allowed us to obtain signals of different nonlinear orders that are uncontaminated from overlapping higher orders. We observe changes in the lineshapes from third to higher orders. To gain further insight into the higher-excited states of our sample, we performed simulations using the Ultrafast Spectroscopy Suite toolbox [2,3]. Through the combination of uncontaminated higher-order 2D signals and simulations, we retrieve transition dipole moments and energy levels of higher-excited states, giving us a new way to verify and improve theoretical models.

[1] J. J. Krich et al., J. Phys. Chem. Lett. **2025**, *16*, 5897.
[2] P. A. Rose & J. J. Krich, J. Chem. Phys **2021**, *154*, 034108.
[3] P. A. Rose & J. J. Krich, J. Chem. Phys **2021**, *154*, 034109.

MO 5.6 Mon 18:30 P 204

Efficient Two-Dimensional Spectroscopy Simulations including Realistic Pulse Shapes, Overlapping, and Time-Ordering Effects — •Rémi Gilliot[1,2], Matteo Russo[1], Alexander Blech[1], Manuel Joffre[2], Christiane Koch[1], and Hélène Seiler[1] — [1]Freie Universität Berlin, Berlin, Germany — [2]Institut Polytechnique de Paris, Palaiseau, France

Two-dimensional (2D) spectroscopy is a powerful pump-pump-probe method to reveal coupling between quantum states and disentangle optical response contributions. We present an efficient approach for simulating 2D spectra with arbitrary pulse shapes including overlapping and time-ordering. Under the assumption of simple dephasing, we show that the emitted signal expression reduces to three independent nested integrals, that can be computed in linear time. We utilize this approach to study the impact of spectral phase distortions and strongly non-Gaussian pulse shapes - such as those produced experimentally by hollow-core fibers.