MO 9: Poster – Novel Approaches

Time: Tuesday 14:00–16:00 Location: Tent

MO 9.1 Tue 14:00 Tent

Spectroelectrochemical cell designs for ultrafast spectroscopy — •Rebecca Fröhlich and Tobias Brixner — Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

Charged molecules play essential roles in many photophysical and photochemical processes. Therefore, the identification of chemical redox species and the kinetic evolution of their photoexcited states is highly desirable. However, unequivocal identification of species remains challenging when reaction intermediates are chemically unstable and the dynamics take place on an ultrafast timescale. The ultrafast dynamics of specific redox species can be accessed by combining electrochemical methods with transient absorption spectroscopy [1,2]. Here, we present and compare several spectroelectrochemical cells of our own design for application in time-resolved spectroelectrochemistry. The cell designs feature flow options as well as optically transparent thin-layer geometries with the aim of realizing a simple setup and reliable performance. The advantages and drawbacks of each cell design are illustrated by performing cyclic voltammetry, absorption spectroelectrochemistry, and ultrafast transient absorption spectroscopy experiments.

[1] S. Bold et al., Chem. Commun. **54**, 10594 (2018).
[2] R. Fröhlich et al., J. Chem. Phys. **160**, 234201 (2024).

MO 9.2 Tue 14:00 Tent

Optimized Velocity Map Imaging Spectrometer for Deep-UV Measurements — •Fabian Westmeier, Nicolas Ladda, Jochen Mikosch, Thomas Baumert, and Arne Senftleben — Institute of Physics and CINSaT, University of Kassel, 34132 Kassel, Germany

Velocity Map Imaging spectroscopy [1] is a powerful method for investigating photoionization processes, by projecting the photoelectron angular distribution onto a position-sensitive detector. We present a spectrometer that is used to study the dynamics of chiral molecules via time-resolved photoelectron circular dichroism [2] with deep-UV photons. Such experiments often experience a high level of background signals due to photoelectrons generated at the spectrometer electrodes from scattered photons. Here we present our successful approaches to reduce this background. We achieved the biggest improvement by using thin electrodes, which minimize the surface area exposed to scattered light. A large hole in the repeller plate combined with an additional high-voltage electrode underneath results in the photoelectrons emitted from the repeller plate being captured by the electrode above. Furthermore, we designed light baffles exhibiting high UV absorption [3], which confine the opening angle for scattered light. To minimize overall scattering, we used thin single-crystal calcium fluoride (CaF2) windows.

[1]: A. T. J. B. Eppink, D. H. Parker, Rev. Sci. Instrum. 68, 3477-3484 (1997)
[2]: C. Lux et al., Angew. Chem. Int. Ed. 51, 5001-5005 (2012)
[3]: O. J. Clarkin, Dissertation, Queen's University, Canada (2012)

MO 9.3 Tue 14:00 Tent

Generalized energy gap law: An open system dynamics approach to non-adiabatic phenomena in molecules — •Nico Bassler[1,2], Michael Reitz[3], Raphael Holzinger[4], Agnes Vibók[5,6], Gábor Halász[7], Burak Gurlek[8], and Claudiu Genes[1,2] — [1]Max Planck Institute for the Science of Light, D-91058 Erlangen, Germany — [2]Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), D-91058 Erlangen, Germany — [3]Department of Chemistry and Biochemistry, University of California San Diego, La Jolla, California 92093, USA — [4]Institut für Theoretische Physik, Universität Innsbruck, A-6020 Innsbruck, Austria — [5]Department of Theoretical Physics, University of Debrecen, H-4002 Debrecen, Hungary — [6]ELI-ALPS, ELI-HU Non-Profit Ltd, H-6720 Szeged, Hungary — [7]Department of Information Technology, University of Debrecen, H-4002 Debrecen, Hungary — [8]Max Planck Institute for the Structure and Dynamics of Matter and Center for Free-Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg, Germany

Non-adiabatic phenomena, resulting from the breakdown of the Born-Oppenheimer approximation, influence most photo-physical and photo-chemical processes, limiting molecular quantum efficiency. The energy gap law, established five decades ago, predicts non-radiative decay with an exponential dependence on the energy gap. Here, we revisit and extend this theory to incorporate vibrational relaxation, dephasing, and radiative loss with a focus on the structure of the nonadiabatic coupling.

MO 9.4 Tue 14:00 Tent

Generation of broad-bandwidth deep ultraviolet pulses with achromatic second harmonic generation — •Nils-Oliver Schütz, Lukas Bruder, Ferdinand Bergmeier, and Ulrich Bangert — University of Freiburg, Institute of Physics, Hermann-Herder-Straße 3, 79104 Freiburg, Germany

The generation of deep ultraviolet optical pulses featuring broad spectral bandwidth and short pulse durations is a challenging task, especially when using high repetition rate ($>$ 100kHz) laser systems that provide low pulse energies to drive the nonlinear conversion processes. We present a scheme based on second harmonic generation of the output of a non-collinear optical parametric amplifier. To increase the bandwidth and efficiency of the second harmonic generation we employ achromatic phase matching [1]. First results will be presented.

MO 9.5 Tue 14:00 Tent

High-repetition-rate ultrafast electron diffraction with direct electron detection — Fernando Rodriguez Diaz, Andrey Ryabov, Mark Mero, and •Kasra Amini — Max-Born-Institut, Max-Born-Straße 2A, 12489, Berlin, Germany

We present a novel ultrafast electron diffraction (UED) instrument that operates at high repetition rates and utilizes direct electron detection, enabling the measurement of time-resolved electron scattering signals with single-electron pulses at 30 kHz. With this state-of-the-art setup, we achieved time-resolved measurements from thin-film solid samples, demonstrating a difference contrast signal, $\Delta I/I_0$, as low as 10^{-5} and an instrument response function of 184 fs (FWHM) without temporal compression and a 1-metre cathode-sample distance. This significant advancement, combined with ongoing developments in RF-compressed and THz-streaked electron pulses, lays the groundwork for investigating ultrafast photochemical reaction dynamics in gas-phase molecules with sub-100-fs total temporal resolution.