A 9: Strong-field Ionization and Imaging (joint session MO/A)

Time: Monday 17:00–18:30   Location: HS 3044

A 9.1   Mon 17:00   HS 3044

Strong-Field Ionization of Nitrous Oxide Molecule by Short Femtosecond Laser Pulses — •FERAS AFANEH — Physics Department, The Hashemite University, P.O. Box 150459, Zarqa 13115, Jordan.

The dissociative photoionization of nitrous oxide molecules, an important atmospheric trace gas, induced by circularly and elliptically polarized laser pulses has been studied by photoelectron photoion coincidence (PEPICO) spectroscopy. PEPICO spectra were used to identify different dissociative photoionization channels. It is observed that the ionized N_2O and its fragments have different correlation trends at different polarization schemes. The relative contributions of different double and triple dissociative ionization channels to the total fragment ion yield are also deduced from the coincident spectra of these channels. The results show that the double dissociative photoionization channels: the denitrogenation ($N_2O^{2+} \rightarrow N^+ + NO^+$) and the deoxygenation ($N_2O^{2+} \rightarrow O^+ + N_2^+$). Furthermore, a considerable contribution of the triple dissociative ionization channels to the total fragment ion yield is also observed. The channels "$N^+ + NO^+$" and "$O^+ + N_2^+$" can be explained by dissociation via the $X^3\Sigma^-$ and $1^3\Pi$ states of N_2O^{2+} as the major peaks in the measured kinetic energy release spectra suggested.

A 9.2   Mon 17:15   HS 3044

Theoretical semiclassical modelling of Laser Induced Electron Diffraction (LIED) — •ÁLVARO FERNÁNDEZ[1,2], ARMIN ISKE[3], ANDREY YACHMENEV[1,4], and JOCHEN KÜPPER[1,2,4] — [1]Deutsches Elektronen-Synchrotron DESY — [2]Department of Physics, Universität Hamburg — [3]Department of Mathematics, Universität Hamburg — [4]Center for Ultrafast Imaging, Universität Hamburg

Experimental techniques for molecular imaging underwent a very fast development in the past decades. From a broad range of novel techniques, laser induced electron diffraction (LIED) [1] stands out because of its high spatiotemporal resolution, high cross section, and lack of structural damage compared to other modern techniques. However, the complexity of this technique causes the necessity of its own theory to understand the results. A general and accurate quantum simulation of the experiment is, to this date, unfeasible and, for this reason, semiclassical models [2] have arised as useful predicting algorithms.

In this talk, a comprehensive analysis of the LIED experiment using a semiclassical model will be given. With this model, we can obtain efficient simulations of the outcome for flexible configurations of molecular geometries. The quality of the results will depend on several factors such as the choice of ionisation theory or electrostatic potential model. An study of the relevance of these factors in the computation of effective cross section for high energy electrons will be presented during the talk.

[1] Karamatskos, E. T, *et al.*, *J. Chem. Phys.*, **150**, 24 (2019)
[2] Wiese, J., *et al.*, *Phys. Rev. Research*, **3**, 013089, (2021)

A 9.3   Mon 17:30   HS 3044

Wavefunction Reconstruction of Excitonic Edge States using Machine Learning — •ARITRA MISHRA and ALEXANDER EISFELD — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

A typical problem in quantum mechanics is to reconstruct the eigenstate wave functions from measured data. In the case of molecular aggregates, the information about the excitonic eigenstates is important to understand the optical and transport properties [1]. It has been shown for a linear and a 2D arrangement of the aggregate molecules that such a reconstruction is possible from the spatially resolved near field absorption spectra [2].

Here, we consider the aggregates arranged in two sublattices in a 2D arrangement, each sub lattice having a particular orientation of the molecules as described in [3]. Interestingly, such an arrangement can lead to the formation of topological excitonic edge states. We study the reconstruction of the excitonic wave function of such a system from the near field absorption spectra. The reconstruction is further investigated in the presence of disorder in the Hamiltonian and noise added to the spectra.

[1] X. Gao and A. Eisfeld, J. Phys. Chem. Lett. 9, 6003 (2018)
[2] F. Zheng, X. Gao and A. Eisfeld, Phys. Rev. Lett. 123, 163202

(2019)
[3] J. Yuen-Zhou, S. K. Saikin, N. Y. Yao and A. Aspuru-Guzik, Nature Materials 13, 1026 (2014)

A 9.4   Mon 17:45   HS 3044

Molecular self-probing for the visualisation of vibrational wave-packet dynamics and its laser-induced modification — •GERGANA D. BORISOVA[1], PAULA BARBER BELDA[1], SHUYUAN HU[1], PAUL BIRK[1], VEIT STOOSS[1], MAXIMILIAN HARTMANN[1], ROBERT MOSHAMMER[1], ALEJANDRO SAENZ[2], CHRISTIAN OTT[1], and THOMAS PFEIFER[1] — [1]Max-Planck-Institut für Kernphysik, 69117 Heidelberg — [2]Institut für Physik, Humboldt-Universität zu Berlin, 12489 Berlin

We present an all-optical pump-control scheme for molecular wave-packet (WP) visualisation and control, where the molecular ground state acts as an intrinsic self-probe of the system, imprinting the evolution of an excited wave packet onto the coherent dipole emission [1]. In a proof-of-principle experiment, coherent extreme ultraviolet (XUV) light creates a vibrational wave packet in the electronically excited $D\,^1\Pi_u 3p\pi$ state of neutral H_2. Measured XUV absorption spectra of the D-state vibronic resonances provide access to the WP dynamics after reconstruction of the time-dependent dipole response [2], which probes the vibrating wave packet through the molecular ground state. An intense near-infrared (NIR) pulse, applied shortly after the WP excitation, is used to control the wave-packet evolution and through this its revival. With increasing NIR intensity the WP revival shifts to earlier times. We identify state-specific NIR-induced phase shifts as the origin of the observed time shifts, which can be applied even to complex molecular systems to coherently steer the recovery of vibrational wave packets on electronically excited potential-energy curves at a desired time. [1] arXiv:2301.03908 [2] PRL 121 (2018) 173005

A 9.5   Mon 18:00   HS 3044

Ultrafast imaging of rare-gas clusters from their formation to their ionization dynamics — •ALESSANDRO COLOMBO for the RareGas Clusters at SwissFEL-Collaboration — ETH Zurich, 8049 Zürich, Switzerland

Coherent Diffraction Imaging (CDI) experiments performed at Free-Electron Lasers (FELs) allow to capture femtosecond snapshots of isolated nanosamples, and are a unique tool for spatially and temporally resolve ultrafast electron dynamics at the nanoscale. Isolated atomic clusters represent the perfect prototypical system for such investigations, thanks to their simple electronic structure and their highly tunable size and shape [1]. We present imaging studies performed at Swiss-FEL on mixed Ar/Xe nanoclusters produced by supersonic expansion into vacuum. Imaging results at 1 keV photon energy reveal fascinating and unexpected shapes at a spatial resolution of few nanometers, which stimulate further research about the thermodynamics and kinematics of these systems. Additionally, the FEL was tuned to photon energies around 0.67 keV, corresponding to the xenon 3d electronic resonance. Fluctuations of the scattering cross-section of Xe can be identified in the CDI reconstructions even several tens of eV away from the 3d edge. The observed behavior can be interpreted as the footprint of ultrafast ionization dynamics happening within the FEL pulse duration, giving insights into the evolution of high charge states, their optical properties and the contribution of transient electronic resonances.

[1] A. Colombo and D. Rupp. (2023) in *Structural Dynamics with X-ray and Electron Scattering*, Royal Society of Chemistry, *in press*

A 9.6   Mon 18:15   HS 3044

High repetition rate ultrafast electron diffraction with direct electron detection — FERNANDO RODRIGUEZ DIAZ, MARK MERÖ, and •KASRA AMINI — Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany

Ultrafast electron diffraction (UED) is a power tool that can monitor the nuclear dynamics of photo-induced gas-phase reactions in real-time with picometre and <250-fs spatiotemporal resolution. However, the temporal resolution of state-of-the-art gas-phase UED setups, often operating at <1-kHz, is insufficient to time-resolve rapidly evolving photo-induced processes (e.g., <350-fs predicted timescale of photoisomerization which plays a crucial role in vision). The limited temporal resolution is due to the severe space-charge dispersion experienced in electron pulses containing 10^4 to 10^5 electrons.

Here, we present a new 30-kHz 100-keV UED setup employing direct electron detection that will be capable of performing time-resolved measurements of photochemical reactions in gas-phase molecules with <100-fs temporal resolution, going beyond the current state-of-the-art in keV and MeV gas-phase UED. This is made possible by operating below the severe space-charge dispersion regime using electron pulses containing very few electrons ($<10^2$) but with sufficient electron flux ($>10^6$ electrons/s) thanks to the high repetition rate of our system. Latest results from the commissioning of our pump-probe UED instrument is presented with details of the current implementation of radiofrequency-compressed electron pulses and the correction of time-of-arrival jitter issues.