SYAD 1: SAMOP Dissertation Prize Symposium

Time: Monday 14:30–16:30 Location: HS 1+2

Invited Talk SYAD 1.1 Mon 14:30 HS 1+2 A simple method to separate single- from multi-particle dynamics in time-resolved spectroscopy — \bullet JULIAN LÜTTIG — University of Michigan, Ann Arbor, USA

The interpretation of time-resolved spectroscopy such as transient absorption (TA) relies on the isolation of a specific nonlinear order, typically the third order, of the sample response. Usually the excitation power is chosen low enough to suppress unwanted higher orders such as exciton–exciton annihilation. However, measurements at low excitation power often exhibit poor signal-to-noise ratio and the contamination by higher-order contributions cannot be quantified. In the opposite case of too high powers, higher-order signals connected to multi-particle dynamics are present and have to be considered in the analysis. We developed a technique using linear combinations of TA spectra at selective powers to extract uncontaminated nonlinear signal contributions. Thus, we access single-particle signals with high signalto-noise ratio and obtain separate multi-particle dynamics [1,2]. Our technique can be easily implemented in any TA experiment and is applicable to any type of quantum system. We measured a broad variety of samples such as quantum dots, photosynthetic complexes, silicon nanocrystals, and polymers. The method provides direct access to the various terms of the perturbative expansion of light–matter interaction allowing one to systematically increase the number of interacting particles, infer their interaction energies and reconstruct their dynamics. [1] P. Malý et al., Nature 2023, 616, 280.

[2] J. Lüttig et al., J. Phys. Chem. Lett. 2023, 14, 7556–7573.

Invited Talk $SYAD 1.2$ Mon $15:00$ HS $1+2$ Time-resolving quantum dynamics in atoms and molecules with intense x-ray lasers and neural networks — • ALEXANDER Magunia — Max-Planck-Institut für Kernphysik

The dynamics of electrons in atoms and nuclei in molecules are essential to the properties of matter. While the electronic processes can be incredibly fast, on the order of a femtosecond or faster, laser pulses even shorter in time allow to resolve them nevertheless. The two main avenues for achieving such short pulses, High-order Harmonic Generation (HHG) and Free-Electron Lasers (FELs), typically imply going to the extreme-ultraviolet (XUV) and x-ray spectral regimes as well.

In this talk, the very first experiment combining FEL and HHG pulses is presented. A prototypical photochemical reaction, the coupling of nuclear and electronic responses during state-selective photodissociation of molecular oxygen, is time resolved. In addition, ultrafast electronic-population transfer mechanisms in the XUV/x-ray regime, in particular Rabi oscillations studied with absorption spectroscopy, will be addressed. This further enables machine-learning applications for reconstructing time-dependent quantum properties, such as the electronic-state populations.

Invited Talk $SYAD 1.3$ Mon 15:30 HS $1+2$

How rotation shapes the decay of diatomic carbon anions — ∙Viviane C. Schmidt — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Molecules that are internally highly excited play an important role in a range of fields from atmospheric to plasma physics. Modelling such systems requires a detailed understanding of the molecules' behaviour under these extreme conditions. However, this is a non-trivial task due to the high density of excited states as well as the variety of competing decay mechanisms available. The diatomic carbon anion C_2^- presents an excellent benchmark to understand the interplay of different decay channels at high internal excitation. The system is arguably the most extensively studied molecular anion in history. Yet, its decay behaviour at high internal excitation has long remained a puzzle for physicists. When produced in a hot ion source, a subset of the resulting anions spontaneously eject their excess electron with a very narrow lifetime span of about 3 ms. While this autodetachment phenomenon has been known since the 1990s, the responsible anionic excited states and their decay mechanism have long remained elusive. Based on our measurements of autodecay of highly excited \mathcal{C}_2^- at the Cryogenic Storage Ring (CSR) facility in Heidelberg, we carried out detailed calculations of the excited states and their decay behaviour. Here, we were able to uncover the profound effect rotational excitation has on the system's electronic landscape. This in turn alters the available decay channels at high excitations and enabled the discovery of a new autodetachment mechanism, which explains the measured feature.

Invited Talk $SYAD 1.4$ Mon $16:00$ HS $1+2$ Interstellar stardust from stellar explosions recorded in a deep-ocean ferromanganese crust within the last 10 million years — ∙Dominik Koll — Australian National University, Canberra, Australia — TUD Dresden University of Technology, Dresden, Germany — Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany

Stars are the element factories in the universe. Stellar explosions eject freshly synthesized radionuclides into interstellar space which subsequently can be accreted by Earth. These radionuclides are the fingerprints of stellar nucleosynthesis.

In this work, I characterized a 10-million year old ferromanganese crust from the bottom of the Central Pacific and chemically extracted single atoms of live interstellar radionuclides ${}^{60}Fe$, ${}^{244}Pu$ and ${}^{247}Cm$ for accelerator mass spectrometry. The dating of the crust with cosmogenic ¹⁰Be led to the discovery of an unexpected anomaly during the late Miocene, which has the potential to be used as a new dating anchor on the Myr timescale for deep-ocean archives.

Highly time-resolved profiles of supernova 60 Fe and r-process 244 Pu and ²⁴⁷Cm revealed two influxes of supernova-produced interstellar stardust within the last 10 Myr. The influx profile of ²⁴⁴Pu and ²⁴⁷Cm constrain the last r-process event in the Milky Way.