

MO 19: Interaction with Strong or Short Laser Pulses I (joint session A/MO)

Time: Wednesday 14:30–16:15

Location: GrHS Mathe

Invited Talk

MO 19.1 Wed 14:30 GrHS Mathe

Time Resolved Diffractive Imaging of Laser Induced Dynamics in Materials — •TOM BÖTTCHER, RICHARD ALTENKIRCH, STEFAN LOCHBRUNNER, CHRISTIAN PELTZ, THOMAS FENNEL, and FRANZISKA FENNEL — Institute of Physics and Department of Life, Light and Matter, University of Rostock, 18051 Rostock, Germany

Micromachining with ultrashort laser pulses is widely used for industrial applications. In contrast to picosecond and nanosecond lasers, ultrashort laser pulses allow precise material modifications due to local electronic excitation on timescales well below electron-ion equilibration times and thermal dissipation. However, the underlying processes leading to target modification and ablation after ultrashort laser pulse excitation are still insufficiently understood.

We present an experimental method to study the excitation and relaxation processes in thin gold films using femtosecond to nanosecond single-shot pump probe coherent diffractive imaging. The target is a 30 nm-thick, free-standing gold foil, which is excited using an 800 nm femtosecond pump pulse. The dynamics in the excited foil are imaged after a variable time delay using a 400 nm femtosecond probe pulse which creates a diffraction image that is captured by a CMOS camera. A phase retrieval algorithm is used to reconstruct the 2D spatial and time resolved exit field at the target position from the captured diffraction images. Dynamics are monitored up to 2 ns, providing access to ultrafast excitation (fs-ps regime) as well as melting and ablation dynamics (ps-ns regime).

MO 19.2 Wed 15:00 GrHS Mathe

Ionization and Fragmentation of Polyatomic Molecules in Intense Laser Fields using a Reaction Microscope — •MARTIN GARRO, NARAYAN KUNDU, HORST ROTTKE, ARNE SENFTLEBEN, and JOCHEN MIKOSCH — Institut für Physik, Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel, Germany

As attosecond science advances into the study of polyatomic molecules, the many-electron and non-adiabatic phenomena in strong-field ionization (SFI) become relevant. A fundamental question concerns the population of electronically excited states of the cation in the intense field, in particular whether these processes are direct or sequential. We study ionization and fragmentation of polyatomic molecules in intense laser fields experimentally with a Reaction Microscope. Coincidence detection of electron and ion momenta reveal detailed insights into the underlying physics. This presentation highlights our recent work on SFI of 1,3-butadiene, n-butane, and 1-butene, in which the intensities and wavelengths were varied. We observe qualitative changes of experimental observables as a function of these parameters, which we interpret as transition between non-sequential and sequential excitation processes.

MO 19.3 Wed 15:15 GrHS Mathe

Machine learning for retrieval of the time-dependent internuclear distance in a molecule from photoelectron momentum distributions: fully quantum mechanical approach — •NIKOLAY SHVETSOV-SHILOVSKI and MANFRED LEIN — Leibniz Universität Hannover

We use a neural network for retrieval of the time-varying bond length in a dissociating one-dimensional H_2^+ molecule based on photoelectron momentum distributions (PMDs) from strong-field ionization. In contrast to our previous study [1], the motion of the atomic nuclei is treated fully quantum mechanically, i.e., PMDs are obtained from the solution of the time-dependent Schrödinger equation for the wavefunction depending on both the electron coordinate and the internuclear distance. We show that the neural network can recognize the time-dependent bond length with a good accuracy. Therefore, machine learning can be applied for time-resolved molecular imaging.

[1] N. I. Shvetsov-Shilovski and M. Lein, *J. Phys. B: At. Mol. Opt. Phys.* 57, 06LT01 (2024).

MO 19.4 Wed 15:30 GrHS Mathe

Harmonic generation with topological edge states and electron-electron interaction — •SIAMAK POOYAN and DIETER

BAUER — Institute of Physics, Rostock University, 18051 Rostock, Germany

It has been found previously that the presence or absence of topological edge states in the Su-Schrieffer-Heeger (SSH) model has a huge impact on harmonic generation spectra. More specifically, the yield of harmonics for harmonic orders that correspond to photon energies below the band gap is many orders of magnitude different in the trivial and topological phase. It is shown in this work that this effect is still present if electron-electron interaction is taken into account, i.e., if a Hubbard term is added to the SSH Hamiltonian. To that end, finite SSH-Hubbard chains at half filling are considered that are short enough to be accessible to exact diagonalization but already showing edge states in the topological phase. We show that the huge difference in the harmonic yield between the trivial and the topological phase can be reproduced with few-level models employing only the many-body ground state and a few excited many-body states.

MO 19.5 Wed 15:45 GrHS Mathe

High-harmonic generation in weakly coupled organic molecular systems — •FALK-ERIK WIECHMANN^{1,2}, SAMUEL SCHÖPA¹, LINA MARIE BIELKE¹, FELIPE MORALES³, SERGUEI PATCHKOVSKI³, MARIA RICHTER³, DIETER BAUER¹, and FRANZISKA FENNEL^{1,2} — ¹Institute of physics, University of Rostock, 18059 Rostock, Germany — ²Department of Life, Light and Matter, University of Rostock, 18059 Rostock — ³Max Born Institute (MBI) for Nonlinear Optics and Short Pulse Spectroscopy, 12489 Berlin, Germany

We introduce organic molecular crystals (OMCs) as a novel target class for high-harmonic Generation (HHG), bridging the gap between gas phase and solid state high-harmonic spectroscopy. In OMCs, neighboring molecules experience a weak van-der-Waals coupling, considerably smaller compared to the covalent or ionic bonds in previous solid-state target. However, this finite coupling leads to *solid like* features, e.g. a delocalization of the electronic states over several unit cells. Additionally, the perfect inherent alignment of all molecules makes OMCs an ideal target class for HH spectroscopy of large organic molecules, as it avoids the need for extremely challenging alignment techniques that have so far prevented corresponding measurements in the gas phase. With a fundamental 4000 nm mid-IR beam reaching 0.67 TW/cm² we demonstrate that HHG from Pentacene crystals is possible without imposing physical damage. We find that the harmonic-generation process is driven by collective intermolecular effects and not by the response of non-interacting aligned molecules.

MO 19.6 Wed 16:00 GrHS Mathe

A theoretical perspective on high-harmonic generation in organic molecular crystals — •SAMUEL SCHÖPA¹, LINA BIELKE¹, FALK-ERIK WIECHMANN¹, FELIPE MORALES², SERGUEI PATCHKOVSKI², MARIA RICHTER², FRANZISKA FENNEL¹, and DIETER BAUER¹ — ¹Institute of physics, University of Rostock, 18059 Rostock — ²Max Born Institute (MBI) for Nonlinear Optics and Short Pulse Spectroscopy, 12489 Berlin

We investigate the underlying mechanism of high-harmonic generation (HHG) in the novel target class of organic molecular crystals (OMCs). Compared to covalent and ionic-bonded solids, the molecules that bond to form OMCs are much more weakly coupled, which is reflected in an energy band structure dominated by single-molecule excitations and charge-transfer states of neighbouring molecules. But does the intramolecular response of the aligned molecules dominate the HHG process? Or can we exploit HH spectroscopy to study the solid-state properties of OMCs, which are characterized by the intermolecular couplings? We addressed this by simulating the HHG process using full time-dependent density-functional theory (TD-DFT) for different polarizations of the driving field and compared it with experimental results. We find in both, that the rotation of the driver polarization reveals maxima in the harmonic yield when the polarization is aligned with the axes connecting neighbouring molecules. A simple tight-binding model shows, that lower harmonic orders are primarily governed by the intramolecular response, while higher orders depend mainly on the intermolecular coupling.