MO 4: Molecular Spectroscopy of Liquid Jets I

Time: Monday 17:00–18:00 Location: HS XVI

MO 4.1 Mon 17:00 HS XVI

Development and implementation of a flat jet device for mesophase-dependent High Harmonic Generation experiments in thermotropic liquid crystals — •Marta Luisa Murillo-Sánchez, Natalia Copete-Plazas, and Laura Cattaneo — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Deutschland

High harmonic generation (HHG) spectroscopy aims for the study of ultrafast molecular dynamics, offering insights into electronic and nuclear motion. While early work in gas-phase systems can be fully understood by the three-step model, the focus has shifted to condensed matter, where collective interactions make HHG mechanisms more complex to unravel. To explore these mechanisms, thermotropic liquid crystals (LCs) are promising due to their tunable mesophases*intermediate states between liquid and crystalline phases*achieved via temperature changes. Using a custom flat jet device adapted for the high viscosity and non-Newtonian behavior of LCs, we generated stable, down to 2 μm-thin sheets for substrate-free HHG experiments with a precise temperature control. Preliminary HHG spectra were obtained for isotropic (liquid) and nematic phases of 4-cyano-4'-pentylbiphenyl (5CB) under a mid-infrared driving field (4 *m). Results highlighted the nematic phase*s anisotropic structure and birefringence, underscoring the need for molecular alignment control. These findings pave the way for optimizing HHG in LCs and deepening our understanding of nonlinear optical phenomena in soft condensed matter.

MO 4.2 Mon 17:15 HS XVI

Exploring the origin of multiple plateaus in liquid high-harmonic generation — •Angana Mondal[1], Ofer Neufeld[2], Tadas Balciunas[1], Zhong Yin[1], Benedikt Wasser[1], Serge Müller[1], Angel Rubio[3,4,5], Nicolas Tancogne-Dejean3[3,4], and Hans Jakob Wörner[1] — [1]Laboratorium für Physikalische Chemie, ETH Zürich, Switzerland — [2]Technion Israel Institute of Technology, Israel — [3]Max Planck Institute for the Structure and Dynamics of Matter, Germany — [4]Center for Free-Electron Laser Science CFEL, DESY, Germany — [5]Physics Department, University of Hamburg, Germany

Recent studies of liquid high-harmonic generation highlight scattering-limited electron trajectories as the key mechanism, with on-site recombination as the primary process[1]. However, this framework left unexplained the absence of higher-order nonlinearities with increasing laser power. Here we report, the observation of a second plateau in the liquid HHG spectrum, attributed to electron recombination with neighboring molecules, dominated by second solvation shell contributions via hole delocalization [2,3]. The plateau exhibits weak scaling with laser wavelength, intensity, and distinct ellipticity dependence, confirmed experimentally and theoretically. Our results predict the existence of higher-order plateaus linked to successive recombination events, establishing a pathway for attosecond-scale probing of electron dynamics in liquids and solutions. Reference 1. A Mondal et al. Nat. Phys. 19, 1813-1820 (2023) 2. I Jordan et al. Science 369, 974-979 (2020) 3. X Gong et al. Nature 609, 507-511 (2022)

MO 4.3 Mon 17:30 HS XVI

Delocalized electrons in aqueous jets — •Fabio Novelli[1], Adrian Buchmann[1], Iqra Yousaf[1], Lion-Luca Stiewe[1], Wibke Bronsch[2], Federico Cilento[2], Claudius Hoberg[1], and Martina Havenith[1] — [1]Ruhr University Bochum, Bochum, Germany — [2]Elettra - Sincrotrone Trieste S.C.p.A., Strada Statale 14, km 163.5, Trieste I-34149, Italy

The photoexcitation of iodide solutions serves as a model for generating electrons in liquid water. We used transient absorption spectroscopy across terahertz, near-infrared, and visible frequencies on a 10-micron thick liquid jet operating at normal temperature and pressure conditions (20 C, 1 atm). We demonstrate that the two-photon absorption of 400 nm pulses can impulsively generate short-lived (250 fs) electrons that are delocalized tens of angstroms away from the parent anion. These electron states are associated with 5p-6p transitions, similar to frustrated excitons with a large radius. Our transient terahertz spectroscopy findings reveal that delocalized electrons exhibit an electronic mobility of 1 cm2/(Vs). This is significantly higher, by approximately 500 times, than that of fully relaxed or hydrated electrons, and roughly comparable to that found in amorphous silicon or conductive conjugated polymers. This work highlights the effectiveness of transient terahertz spectroscopy in investigating low-energy, intra-band electronic transitions in soft condensed matter systems and can assist in the development of liquid-based optoelectronic devices.

MO 4.4 Mon 17:45 HS XVI

Optical-Pump THz-Probe Spectroscopy of Myoglobin in Water — •Adrian Buchmann[1], Sebastian Jung[1], Lion-Luca Stiewe[1], Luigi Caminiti[2], and Martina Havenith[1] — [1]Ruhr Universität Bochum, Bochum, Germany — [2]European Laboratory for Non-Linear Spectroscopy, Florence, Italy

Enzymes (functional proteins) are known to be the most effective catalysts. In the search for more and more effective catalysts and energy storage devices, chemists look for solutions to copy nature and attempt to design proteins from scratch. However, so far most fail to reproduce the catalytic activity of their evolved siblings. A factor suggested to contribute to this shortcoming is the unknown mechanism of energy flow through the proteins. We employ nonlinear THz spectroscopy to follow this energy flow from an excited heme through its surrounding myoglobin protein into the surrounding solvent. This enables us to determine the energy flow properties from the vibrational relaxation time of the protein. Optical Pump THz probe spectroscopy in water requires a windowless approach making the liquid jet essential for the experiment.

We observe a transient signal which reaches an equilibrium around 50 ps. The final difference spectrum resembles the spectrum of heated water with a temperature difference of 0.1 K. We can determine a rise time of 7.9 +/- 0.5 ps which is consistent with the vibrational relaxation time observed in the IR frequency range.