Lightwave electronics: sampling optical fields with attosecond resolution

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Time-domain sampling of arbitrary electric fields with sub-cycle resolution enables complementary time-frequency analyses of a system's electromagnetic response. However, scaling such techniques to the visible and near-IR spectral regimes has remained challenging, and seemingly demand high-energy optical sources and complicated optical apparatuses. We have developed and demonstrated an all-on-chip, optoelectronic device capable of sampling arbitrary, low-energy, near-infrared waveforms under ambient conditions with sub-optical-cycle resolution. Our detector uses field-driven photoemission from resonant nanoantennas to create attosecond electron bursts that probe the electric field of sampled waveforms. Additionally, our measurements directly reveal the localized plasmonic dynamics of the emitting nanoantennas *in situ*.

Time Resolved THz Dynamics in Thermotropic Liquid Crystals

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Liquid crystals (LCs) exhibit a meta-phase behavior, combining solid-like order and fluid-like flow. So far, dynamics in LCs occurring on the picosecond timescale remain largely unexplored. They show, in fact, resonant features in the THz spectral region, specifically above 3 THz, but a clear understanding of these signatures has yet to be developed. Here, we use nearly-single cycle THz pulses, generated via optical rectification, to perform both time-domain spectroscopy (TDS) and resonant, time-resolved birefringence measurements on 5CB and 8CB thermotropic LC, presenting nematic and smectic A phase, respectively.

Advances in the simulation of molecular ionization processes

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Complex processes due to interaction of a molecule with electromagnetic radiation are currently investigated in a vast domain of photon ranges, field intensities and time domain. Most processes involve photoionization and in fact electron detection is one of the most common and powerful probe.

Analysis of the results is not easy and often sophisticated models are developed and prove essential for interpretation, at various levels of accuracy. In principle it is always possible to resort to a full Quantum mechanics calculation, which could include the full complexity of the phenomenon, although for larger systems unavoidable approximations have to be introduced, which must be carefully considered. A full simulation may also be the only way to extract vital information from the experiment. A case is the determination of absolute configuration of chiral molecules from spectroscopic signatures, like circular dichroism, optical rotation or more recently photoelectron circular dichroism.

In the presentation I shall broadly review the main problems and tools available for electronic structure calculations in molecules, and go into more detail about the description of continuum states, and the use of B-spline bases. Examples of molecular photoionization will be presented to illustrate current capabilities and open problems.

Ultrafast charge migration in molecules initiated by ionization and excitation

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Exposing molecules to ultrashort pulses results typically in a coherent population of several and even many electronic states. This triggers ultrafast electron dynamics, which may lead to a redistribution of the charge even before the nuclei start to move and adjust – a process referred to as charge migration. The non-adiabatic coupling between the electronic and nuclear motion will eventually destroy the initial electronic coherence and trap the migrating charge. The time during which the electron coherence is maintained is an important parameter, as it can be used to modulate the charge migration process and thus predetermine the succeeding nuclear rearrangement in the molecule – the paradigm of the emerging field of attochemistry. In this talk, fully quantum simulations of ultrafast charge dynamics initiated by both ionization and excitation of polyatomic molecules will be presented and discussed.

High-energy multidimensional solitary states in hollow-core fibers

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Using sub-picosecond pulses, we report the first time observation of the formation of highly-stable multidimensional solitary states (MDSS) in molecular-filled hollow-core fibers. The MDSS have broadband red-shifted spectra with an uncommon negative quadratic spectral phase at output, originating from strong intermodal interactions. This approach paves the route to compress sub-picosecond Ytterbium laser systems to few-cycle pulse duration using a compact setup.

Solid state spectroscopy with attosecond all-optical techniques

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The possibility to manipulate the electrical properties of matter with very short optical pulses is a fascinating field of research with expected far reaching implications for many relevant technological fields. In the last decade, attosecond all-optical techniques have been established as powerful tools to investigate the primary physical processes at the basis of light matter interaction in atoms, molecules and condensed targets, moving the first step towards optical control. In my talk I will introduce

attosecond transient absorption and reflection spectroscopies, reviewing the most important experiments performed with these techniques and discussing non-equilibrium ultrafast electron and exciton dynamics in dielectrics driven by few-cycle PHz pulses.

Ultrafast dynamics probed by X-ray scattering

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The fundamental understanding of ultrafast events in atoms and molecules has undergone significant advancements thanks to time-resolved studies. Modern ultrafast X-ray techniques are particularly suited for such investigations. Yet, the analysis and interpretation of the experimental outcomes need support from detailed atomistic simulations. I will highlight key elements in our contribution to the theory of ultrafast X-ray scattering [1,2] and provide examples of applications to atoms and molecules in the atto- to femtosecond regime [1-5].

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[5] Kunnus, K. et al. Nat. Commun. 2020, **11**, 634.

Real time and local observation of ultrafast electronic and nuclear rearrangements using time-resolved X-ray spectroscopy

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The possibility to generate ultrashort (femtosecond down to attosecond) and intense pulses in the X-ray domain at the new X-ray Free Electron Laser (XFEL) facilities open the door to the investigation of the coupled electron and nuclear dynamics in molecules, which marks the fate of chemical and biological processes. In addition to this extremely high temporal resolution X-ray pulses offer atomic spatial resolution, which allows us to trigger and probe ultrafast dynamics at a particular site in the molecule, and to create localized electronic wavepacket and coherent dynamics via nonlinear X-ray excitation [1]. In this talk, we will present recent joint theoretical and experimental studies, in which we demonstrate the potential of time-resolved X-ray spectroscopy to sensitively probe the rich X-ray induced dynamics in molecules. With such a tool, we are able to follow in real time and site-selectively ultrafast changes in electron density [2], the rapid decay of transient states driven by electron correlations, as well as the formation and breaking of chemical bonds [3]. This work offers ample perspectives to resolve ultrafast evolution of excited states of complex systems in space and time.

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[2] A. Al-Haddad, S. Oberli et al., Submitted (2021)
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Control the spatial orientation of molecules – isolated or in helium nanodroplets –by strong laser pulses

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I will discuss how moderately intense laser pulses can be used to sharply align and orient gas phase molecules – both in the adiabatic limit where a laser pulse slowly brings molecules to strong alignment at its peak and in the impulsive limit where femtosecond pulses set molecules into coherent rotation by launching rotational wave packets. Time-resolved twisting of a chiral molecule and attosecond charge migration are used to illustrate applications of aligned molecules. Finally, I will discuss recent work where molecules or molecular oligomers are embedded inside or on the surface of nanometer-sized, 0.37 K droplets of superfluid liquid helium.

Relativistic and non-adiabatic developments for molecular quantum theory

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High and ultra-high resolution spectroscopy experiments of small molecular systems make it possible to test and further develop the fundamental theory of molecular matter. I will speak about complementary, perturbative and variational approaches to the non-adiabatic and relativistic problems of molecular quantum theory [1–5]. I will sketch theoretical, algorithmic, and computational challenges that were recently met and that are still open for an increasingly complete and accurate description of small atomic and molecular systems.

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Exploiting electronic coherences for steering selective bond formation in molecules pumped by ultrashort optical pulses

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Few-cycle short optical pulses allow pumping coherently several coupled electronic states towards steering nuclear motions in neutral molecules and cations. I will discuss two recent examples for which we demonstrate such a control by fully quantum dynamical computations: the selective bond formation in a 4C ring closure occurring upon excitation of norbornadiene by a few cycle, 2 fs, deep UV pulse and the strong isotope effect occurring in the ultrafast structural Jahn-Teller rearrangement in the methane cation suddenly ionized.