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CUI THE HAMBURG CENTRE FOR ULTRAFAST IMAGING

NEWSLETTER

CUI – Graduate School

No.15, August 2017

Main topics

- Research highlights
- Personalia
- Important dates

Editorial

While waiting for the outcome of the evaluation of our preproposal for the next excellence initiative, we would like to inform you also this summer about recent scientific results obtained by our graduate school members. Once again the choice for the highlights was rather difficult, given the quality and abundance of the research papers. Nonetheless, this indicates once again the very high level of the research conducted within our cluster.

Antonio Negretti and Peter Schmelcher

Research highlights

<u>Ultrafast decoherence caused by nuclear motion:</u> Recently, attosecond pulses became experimentally available. This allows for studying ultrafast molecular dynamics through pump-probe experiments with unprecedented time resolution, where ultrashort, broadband pulses are used to create coherent superpositions of cationic electronic states in molecules through photoionization. This triggers both nuclear and electronic dynamics, leading eventually to the loss of electronic coherence.

Since the heavy nuclei are much slower than the electrons, their motion is often neglected in the interpretation of the experiments. This results in the view that electronic dynamics remain coherent for up to 100 femtoseconds. However, a thorough understanding of ultrafast processes in molecules requires investigating the influence of nuclear motion, even at such short timescales.

To this end, the CUI PhD student C. Arnold, the former CUI young researcher Dr. O. Vendrell, and Prof. R. Santra investigated the nuclear motion at short times in an ab initio, fully quantum-dynamical framework. This implies that they included all nuclear degrees of freedom and represented the nuclei as wavepackets moving on adiabatic potential energy surfaces that were expanded harmonically at the molecular equilibrium geometry.

Electronic coherence, defined as the spatial overlap of nuclear wavepackets across different electronic states, is shown to vanish within few femtoseconds in applying the model to paraxylene and phenylalanine. Surprisingly, it is not the fast modes, e.g. C-H vibrations, but rather the interplay of a large number of slower atomic displacements that add up to destroy electronic coherence.

The researchers found that the topology of the potential energy surfaces is more relevant for electronic decoherence than the actual speed of the nuclear motion. It is thus conceivable that the speed of decoherence is comparable in other molecules (see also Fig. 1).

These conclusions challenge the concept of long-lived electronic coherences in molecules ionized by ultrashort pulses. Purely electronic dynamics that may be described in terms of a coherent electronic wave packet exist only for subfemtosecond time scales, and nuclear motion cannot be neglected.

The work has been published in Phys. Rev. A **95**, 033425 (2017).

<u>Attosecond interferometry:</u> The CUI postdoc S. Usenko together with the CUI PhD student M. Jakob and other colleagues from the DESY X-ray Femtochemistry and Cluster physics group headed by T. Laarmann have realized the phase control of XUV pulses from a free-electron laser (FEL) with attosecond precision.

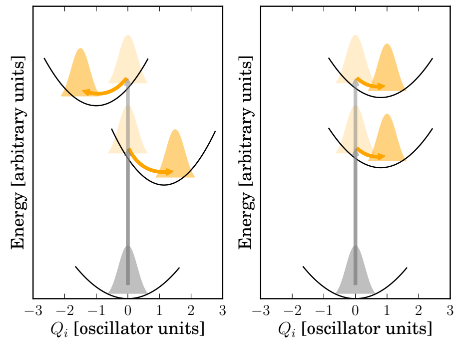

Fig. 1: Motion of nuclear wavepackets on adiabatic potential energy surfaces. Left: different topology leads to diminishing spatial overlap and decoherence. Right: the nuclear wavepackets move synchronously, preserving coherence.

Personalia

Dr. Alexander Britz has obtained recently the doctoral degree in Physics with a thesis on ultrafast X-ray spectroscopies of transition metal complexes relevant for catalysis under the supervision of Prof. Christian Bressler and Prof. Wilfried Wurth.

We congratulate him for his achievements!

Alexander Britz

A reflective split-and-delay unit (SDU) developed in collaboration with Prof. D. Kip from the Helmut Schmidt University in Hamburg and C. Becker from Institute of Laser Physics on the DESY campus plays the key role in the experiment. The SDU splits the wavefront of the incoming FEL pulse uniformly across the beam profile by two interleaved lamellar gratings and provides two pulse replicas with a variable delay. In contrast to a conventional half-mirror SDU, the lamellar geometry provides collinear propagation of both pulse replicas, which ensures that their phase difference dependents only on the delay precisely measured in the experiment.

This enables recording of interferometric autocorrelation signals with maximum phase contrast. In the experiment, an in-vacuum white light interferometer monitors the topography of the SDU in real time thus providing information about the pulse-replica delay with attosecond precision for every laser shot.

The scientists demonstrated the attosecond phase control of XUV light waves by generating two phase-locked replicas of self- phase-locked replicas of self- amplified spontaneous emission pulses from DESY's free-electron laser FLASH with the grating SDU and observing their interference directly in the time domain (Fig. 2). This study opens up the door for novel phase-sensitive spectroscopic studies with attosecond resolution even at partially coherent XUV and soft X-ray FEL sources. Full control over the relative temporal phase in FEL pulse replicas provides opportunities to trace energy and charge migration in systems of increasing complexity with unprecedented spatial and temporal resolution. It makes the local electronic structure and dynamics accessible and controllable.

Further information can be found in Nat. Commun. **8**, 15626 (2017) and Appl. Sci. **7**, 544 (2017).

<u>**Coherent generation of THz spin currents:**</u> When the magnetisation is propagating in a ferromagnetic material, a spin current can be generated in a non-magnetic material. Such a spin-pumping effect is particularly large when a magnetic material is resonantly excited via an external magnetic field, whereas such resonance frequencies are typically in the GHz range.

The CUI postdoc L. Bocklage of the group of Prof. R. Röhlsberger has theoretically investigated the generation of ultrafast spin currents and showed that even for frequencies larger than the resonance a spin current can be observed. This occurs even though the magnetisation has a smaller dependence on external magnetic fields. He founds that the reduction of magnetisation is compensated by its fast time evolution, which induces a prolonged spin current at high frequencies. In this way it is possible to generate spin currents up to the THz-range with conventional ferromagnetic materials. Besides, the calculations show that the spin current behaves at high frequencies coherently, proving thus that external fields can be used to control such currents (see Fig. 3).

The work is published in Phys. Rev. Lett. **118**, 257202 (2017).

Important dates

We draw your attention to the following CUI events: October 11-13 CUI Annual Meeting in the Hotel Hohe Wacht in Hohwacht; November 9 CUI International Symposium in conjunction with the Hamburg Prize for Theoretical Physics. Finally, the next graduate days of CUI will take place during March 19-22, 2018.

You are welcome to: ... send us suggestions of topics, which you would like to be mentioned in the next newsletter (anegrett@physnet.uni-hamburg.de).

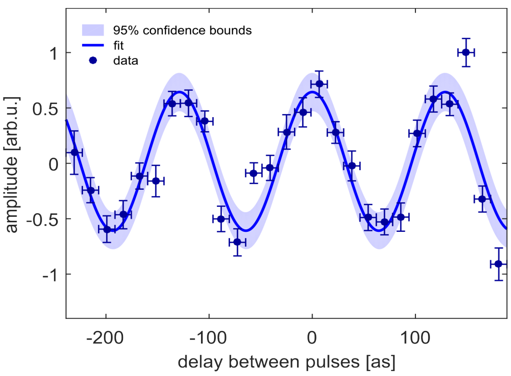

Fig.2: The oscillation of the FEL carrier light wave is monitored by plotting the intensity resulting from the interference of the two pulse replicas as a function of their relative phase delay, e.g. linear autocorrelation. The measured thus optical cycle at the FEL wavelength of 38 nm is 129 as.

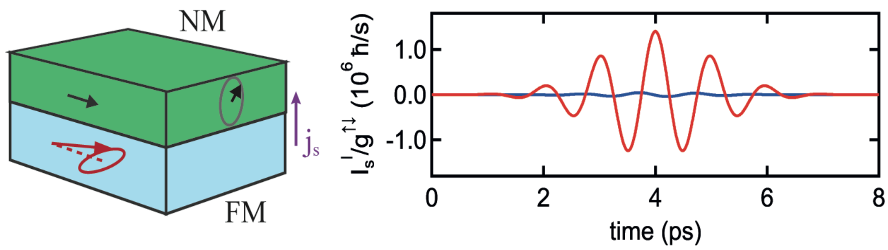

Fig.3: On the left a scheme of the ferromagnetic/non-magnetic (FM/NM) bilayer, which is used for spin pumping experiments, is shown. A spin current j_s is pumped into the non-magnetic layer when the magnetization (red) precesses. Its spin polarization is constant in one direction and oscillates in the other directions. On the right the components of the spin current pulses are shown in different directions at THz frequencies.