POSTER Session WEDNESDAY 6th:

1. Femtosecond laser induced periodic surface structures as substrate patterns for the growth of Zr-doped ZnO structures, Rocio Ariza, Belén Sotillo, Ana Urbieta, Jan Siegel, Javier Solís, and Paloma Fernández, Complutense University of Madrid

2. Electron angular distribution in double ionization of H₂, Kilian Arteaga, Johannes Feist, Fernando Martín, and Alicia Palacios, Universidad Autónoma de Madrid

3. Deepening into the nucleation and fission processes of nano-hydrated ammonia clusters - a combined theoretical and experimental study, D. Barreiro, B. Oostenrijk, N. Walsh, A. Sankari, E. P. Mansson, S. Maclot, S. Sørensen, S. Díaz-Tenero, and M. Gieselbrecht, Universidad Autónoma de Madrid

4. Ultrafast Fiber Lasers for High-Resolution MultiPhoton Microscopy, Pascal Dupriez and Victor Blanco, Spark Lasers


6. Differences in the Photobehavior of Two Chemically Identical but Structurally Distinct MOFs, Elena Caballero-Mancebo, Boyko Cohem, Simon Smolders, Dirk de Vois, and Abderrazzak Douhal, Universidad de Castilla-La Mancha

7. On the problem of achieving low-threshold yellow-green polymer lasing in energy transfer blends, Juan Cabanillas-González, Qi Zhang, Chen Sun, Larry Lüer, Jingguang Liu, Xiangru Guo, Qi Wei, Ruidong Xia, Yan Qian, Donal C D Bradley, and Wei Huang, IMDEA Nanociencia

8. Laser-induced periodic surface structures on germanium by femtosecond irradiations, Noemí Casquero, Yasser Fuentes-Edluf, Javier Solís, and Jan Siegel, Instituto de Óptica (IO-CSIC)

9. The Influence of β-Phase Conformation on Transient Absorption and Light Amplifying Properties of Polydianilinfluorene, Chen Sun, Jinyi Lin, Jaime J. Hernández Rueda, Xingyuan Shi, Aleksandar Perevedentsev, and Juan Cabanillas-González, IMDEA Nanociencia

10. High harmonic generation assisted by single excited multielectron states, Alba de las Heras, Luis Plaja and Carlos Hernández-García, University of Salamanca

11. Attosecond spectroscopy of small organic molecules, Jorge Delgado, A. Palacios, M. Lara-Astiaso, J. González-Vázquez, P. Decleva, and F. Martín, Universidad Autónoma de Madrid

12. Polarization Effects on Laser-Driven Electronic Transfer Processes, Francisco Fernández-Villoria, Antonio Garzón-Ramírez, and Ignacio Franco, Universidad Autónoma de Madrid

13. Ionization from a coherent superposition in CCl₂F₂, Pedro Fernández-Milán, Markus Klinker, Jesús González-Vázquez, and Fernando Martín, Universidad Autónoma de Madrid

14. Time delays from one-photon transitions in the continuum, Jaco Fuchs, Nicolas Douquet, Stefan Donsa, Fernando Martín, Joachim Burgdörfer, Luca Argenti, Laura Cattaneo, and Ursula Keller, ETH Zürich

15. The role of surface roughness in the formation of LIPSS on metals, Yasser Fuentes-Edluf, José A. Sánchez-Gil, Marina Garcia-Pardo, Rosalía Serna, Vincenzo Giannini, Javier Solís, and Jan Siegel, Instituto de Óptica (IO-CSIC)

16. Signatures of matter Talbot effect in the high-order harmonic generation from periodic systems, Ana García-Cabrera, Carlos Hernández-García, and Luis Plaja, University of Salamanca

17. Deciphering the ultrafast events in a new HOF with remarkable responsiveness to acids, Eduardo Gomez, Yuto Suzuki, Ichiro Hisaki, and Abderrazzak Douhal, Universidad de Castilla-La Mancha

18. THz spectroscopic imaging at kHz pixel rates, Albrecht Bartels and Matthias Beck, Laser Quantum GmbH


20. Excited state dynamics of pyrrole-containing clusters, Iker Lamas, Raúl Montero, and Asier Longarte, University of the Basque Country

21. Ultrafast x-ray scattering from molecular wavepackets, Andrés Moreno Carrascosa, Mats Simmenmacher, and Adam Kirrander, Brown University

22. Time-resolved photodissociation dynamics of vinyl iodide in the UV at 199.2 and 200, M. L. Murillo-Sánchez, I. Mondejar and L. Bañares, Universidad Complutense de Madrid
Femtosecond laser induced periodic surface structures as substrate patterns for the growth of Zr-doped ZnO structures

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ZnO has a huge variety of applications in the optoelectronic field due to its excellent properties. Additionally, doped ZnO nanostructures have been extensively investigated in order to modify the optical and electrical properties. Zr was chosen as dopant due to Zr⁴⁺ ionic radius being comparable with Zn²⁺, meaning the lattice distortion is minimized¹, as well as when Zn is substituted for Zr two extra free electrons are donated and the carrier density should increase². Furthermore, this material presents a high thermal and chemical stability and it has been seen that Zr does not bind with Zn, avoiding intermetallic phase formation. For all these reasons, structures based on Zr-doped ZnO offer potential applications as an alternative to the most used transparent conducting oxides (TCO) working at high temperature¹ among other applications in photocatalysis³.

In this work, various substrates were irradiated by a femtosecond laser operating at 1030 nm with a pulse duration of 350 fs. Under certain conditions it is possible to generate laser induced periodic surface structures (LIPSS) in different materials⁴,⁵ as is shown in Figure 1a, which allows to explore the influence of different types of LIPSS on the growth process of nanostructures. To this end, Zr-doped ZnO structures were grown by a vapour-solid method under a constant Argon flux (Figure 1b). In order to determine the critical parameter in the process, mixtures of ZnS and ZrO₂ in different percentages were used as precursor, in conjunction with changes made in parameters such as the thermal treatment duration, orientation and type of LIPSS, crystalline orientation and substrate material. Characterization of the structures was carried out by means of Scanning Electron Microscopy (SEM) and Photoluminescence (PL), providing information about the morphology and variations in luminescent defects as a result of growing on patterned substrates.

Figure 1. a) Optical image of amorphous-crystalline LIPSS in silicon <100> and b) secondary electron image of Zr-doped structures grown on laser patterned silicon substrate.

Notes and References

1 Herodotou, S., Treharne, R., Durose, K., Tatlock, G., & Potter, R., Materials, 2015, vol. 8, no 10, p. 7230-7240
**POSTER**

**Electron angular distribution in double ionization of H$_2$**

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Pump-probe experiments are nowadays able to unravel the role of nuclear motion and electron correlation in excitation and ionization processes in molecules on the ultrashort time scale. The interaction of intense short light pulses with molecular systems necessarily implies the solution of the time-dependent Schrödinger equation in a configuration space that comprises all break-up channels. The resolution of the time-dependent Schrödinger equation in full dimensionality for the case of molecular systems with two electrons, such as H$_2$, represents a non-trivial task. Its main difficulty relies on the fact that, for solving the problem of molecular double ionization, it is mandatory to solve the four-body coulomb break-up to include all the possible paths and be able to extract the observables of the double ionization from the out-going wave packets. On this work, we have implemented a new computational tool allowing us to describe, for the first time, the four-body coulomb break-up of H$_2$ including nuclear motion. This methodology gives access to two main unresolved problems, the multiphoton double ionization of the molecule and its dissociation into neutrals after double excitation. The implemented software tools employ a description of the wave function written in a basis set of FE-DVR (finite elements combined with a discrete variable representation), which makes it easier to handle and converges faster than existing spectral methods employed for single excitation and single ionization problems. Moreover, we have used an exterior complex scaling procedure to impose the appropriate many-body Coulomb boundary conditions in a defined volume. Finally, the existence of particular cases of molecular double ionization of H$_2$ where the nuclear motion does not play an important role, has allowed us to validate the accuracy and applicability of our implementation against the existing results without nuclear motion.

**Fig:** Comparison of the one-photon double ionization: triply differential probability (TDP) for H$_2$ in fixed nuclei approximation.

**Notes and References**

8. CW McCurdy and TN Rescigno. “Practical calculations of quantum breakupcross sections”. In: Physical
Deepening into the nucleation and fission processes of nano-hydrated ammonia clusters - a combined theoretical and experimental study

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The detailed mechanisms occurring at the molecular level in nucleation and fission processes have been reported to be relevant for the formation of atmospheric aerosols [1]. In the troposphere, cosmic radiation background is responsible for molecular ionization and have shown to be also crucial on the formation of these particles. As the concentration of anthropogenic ammonia in earth’s atmosphere has increased in last years, our study model has consisted of multiply charge nano-sized clusters of hydrated ammonia, which may act as nucleation enhancers and therefore explain the particle formation rates of atmospheric aerosols containing, for example, sulfuric acid [2].

Advanced 3D momentum imaging mass spectrometry techniques have been implemented to uncover fundamental mechanisms related to charge and proton dynamics [3]. The aggregates absorb a single soft X-Ray photon, resulting in the formation of multiple charged species. Thanks to our experimental set-up and selection of energies, we can focus on doubly charged clusters. In our calculations, different cluster sizes of the type (NH\textsubscript{3})\textsubscript{m}(H\textsubscript{2}O)\textsubscript{n} have been taken into consideration. In order to find the most stable neutral conformers for these clusters, semi-classical calculations using Self-Consistent Charge Density Functional Tight Binding based calculations (DFTB-SCC) were carried out [4] applying the Parallel Tempering Molecular Dynamics (PTMD) formalism [5]. DFTB in combination with Parallel Tempering Molecular Dynamics has demonstrated to contribute as an excellent tool to successfully explore the Potential Energy Surface (PES). These calculations were performed using specific water-ammonia interaction potentials developed by Cuny et al. [6]. To replicate the experimental conditions, a certain excitation energy have been randomly redistributed among the nuclear degrees of freedom. Then, ab initio molecular dynamics calculations have been perform for the doubly-charged clusters using the atom-centered Density Matrix Propagation (ADMP) method at a DFT/M062X level of theory, see Figure 1.

Our results give an insight into the structure and charge location before and after ionization and also the mechanisms of the most probable fragmentation pathways. They support the role of ammonium as a charge carrier in the solution, preferentially bound to surrounding ammonia molecules, which could influence the atmospheric nucleation process.

1 Kulmala M et al, 2013, Science 339, 943
6 Cuny J et al, 2018, Molecular Simulation, 0, 1

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Snapshots of molecular dynamics simulations of the doubly-charged (NH\textsubscript{3})\textsubscript{2}(H\textsubscript{2}O)\textsubscript{8} cluster showing two different reaction paths.}
\end{figure}
POSTER

Ultrafast Fiber Lasers for High-Resolution MultiPhoton Microscopy

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Biological and Biomedical Imaging has benefited from dramatic technological advances in nonlinear microscopy techniques over the past three decades. Solid-state femtosecond lasers have been the dominant technology over many years, becoming a standard in scientific applications. Recent developments in high performance ultrafast fiber lasers offer new attractive features, challenging traditional solid-state lasers in most applications, and especially in the field of 2-photon microscopy.

Ultrafast fiber lasers provide significant advantages over solid-state lasers, and are becoming the ideal tool for multiphoton microscopy and other imaging applications thanks to their robustness, stability, high performance, small size, beam quality and reliability.

New lasers specifically designed by Spark Lasers for microscopy and in-vivo excitation offer two simultaneous wavelengths (920 and 1040 or 1064nm) exiting from two compact laser heads individually controlled, offering optimal flexibility and ease of use: 920 nm is used for neural response detection and imaging, and a longer wavelength (1040-1064 nm) provides photo-activation in optogenetics applications. Individual control of GDD pre-compensation ensures optimal performance even under the most demanding conditions. The Alcor laser series developed by Spark Lasers is a mode-locked fiber laser with an M2 of < 1.2, producing ultrashort pulses (< 100 fs) with 80 MHz repetition rate, and is capable of producing up to 2 W average power both at 920nm and 1064 nm.

The new generation of ultrafast fiber lasers will expand the boundaries of bioimaging, and already constitutes a significant breakthrough in the field of two-photon imaging and neural excitation.

References

1 Dupriez, P. Photonics Views 2019, Vol.4, 70-73
Using Broadband Time-Resolved THz Spectroscopy in the Study of Hot Carrier Dynamics in Lead Halide Perovskites

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Ultrafast time-resolved terahertz spectroscopy (TRTS) allows for the direct probing of charge carriers and quasiparticles in semiconductors. The sensitivity of the technique to both the carrier mobility and its density can help elucidate the mechanisms of their temporal evolution. The use of gas photonics provides short, ultra-broadband THz pulses and, thus, offers an improved time-resolution.

Here, we present a study of the early steps of the charge carrier dynamics in lead halide perovskite thin films from the point of view of the THz mobility. Taking advantage of a 200 fs time-resolution, we were able to temporally follow the cooling of hot carriers through the observation of the change of their mobility. This change is understood as resulting from a modification of the carrier effective mass.

Our results are compatible with a hot carrier cooling mechanism implying LO phonon emission. This is subjected to a phonon bottleneck, and competes with a dynamic screening process, which time-constant was identified as being due to polaron formation. While the screened hot carriers take longer to cool down, the dynamic screening process does not produce a change in mobility when only cold carriers are involved.

This technique is then applied to 2D perovskite quantum wells where the evolution between carriers and excitons can be observed. This gives an opportunity to exploit the broadband range of frequencies in our probe in order to study the spectral signatures of the different particles and quasiparticles.

Notes and References

POSTER
Differences in the Photobehavior of Two Chemically Identical but Structurally Distinct MOFs

Elena Caballero-Mancebo, Boyko Cohem, Simon Smolders, Dirk de Vois and Abderrazzak Douhal

Metal-organic frameworks (MOFs), a class of hybrid materials made from organic linkers and inorganic nodes, have emerged as smart materials that are gaining relevance in different scientific fields due to their wide range of applications.1 Here, we show how two Ce-MOFs (Ce-NU-1000 and Ce-CAU-24-TBAPy), both having the same linkers and metal clusters, but different structures differ on their photoproperties (Figure 1).2,3 The Ce-NU-1000 exhibits excimer formation, contrary to the Ce-CAU-24-TBAPy which lacks these events. This behaviour reflects the different structural arrangement of the linkers within these MOFs. Furthermore, after a ligand-to-metal charge transfer reaction, the e-h+ recombination times in Ce-CAU-24-TBAPy (1.59 and 13.43 µs) is twice of those of Ce-NU-1000 (0.64 and 4.91 µs), which suggests different structural configuration of the e-acceptor metal clusters. We will also discuss the results of single crystals photobehaviour using time-resolved fluorescence microscopy. Our findings shed new light on the photodynamics of two structurally different Ce-MOFs, and open the door to further research on this kind of hybrid materials.

Figure 1. Scanning Electron Microscopy (SEM) images of A) Ce-NU-1000 and B) Ce-CAU-24-TBAPy. The insets show a close-up of the crystals structure. Structures of C) H4TBAPy and D) Ce cluster that form both MOFs.

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References:
POSTER

On the problem of achieving low-threshold yellow-green polymer lasing in energy transfer blends

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Efficient yellow-green polymer lasing cannot be achieved from energy-transfer blends based on blue-emitting polymer hosts due to spectral overlap between the guest stimulated emission and characteristically long-lived polymer host photoinduced absorption. We report a solution to this problem wherein the photoinduced absorption can be suppressed or delayed by using selected short conjugation length oligomers as host media.[1,2] Transient absorption experiments performed in blend films demonstrate a 2-4 ps build-up of guest-based stimulated emission without substantial competition from residual host photoinduced absorption. We attribute this behavior to a stronger Coulomb confinement and consequently lower photogeneration yield of polaron-pairs in the selected oligomers. This results in an absence or delayed formation of harmful long-lived photoinduced absorption. As a consequence we have achieved a four times increase in net optical gain and a two orders of magnitude threshold reduction for amplified spontaneous emission in the yellow-green emission blends of poly(9,9-dioctylfluorene-co-benzothiadiazole) blends with the short oligomer hosts.

Notes and References


Laser induced ripples that are also known as Laser Induced Periodic Surface Structures (LIPSS) have gained considerable attention by researchers and industry due to their surface functionalization applications. Surface modification with LIPSS takes place in metals, semiconductors or dielectrics [1]. Their morphology depends strongly on many parameters, both on material properties and irradiation conditions (pulse fluence, wavelength, light polarization, pulse repetition rate, and processing speed). Two types of LIPSS are distinguished by their spatial periodicity, namely high-spatial frequency LIPSS (HSFL) having a periodicity significantly smaller than the laser wavelength and low-spatial frequency LIPSS (LSFL) having a periodicity close to the laser wavelength.

In this work, we have investigated the LIPSS formation on germanium by using a Ti:Sapphire (800nm, 120 fs) laser. The results show different scenarios depending on the irradiation conditions. One of these scenarios is presented in Figure 1, showing images of LIPSS formed by the same fluence but different number of pulses. For a low number of pulses, LIPSS of type HSFL were formed parallel to polarization, whereas for larger number of pulses they were of type LSFL and perpendicular to polarization. This evolution of spatial periodicity by increasing the number of pulses is fundamentally different regarding the behaviour in silicon [2], which will be discussed for comparison. On the other hand, since the spatial intensity distribution of the laser beam is Gaussian, the central disk of the irradiated spot has been exposed to the peak fluence. Surprisingly, this central disk did not present ablation but LIPSS appeared on its surroundings, as can be seen in the images. This suggests that a phase change from the crystalline phase into the amorphous one could be the origin of these LIPSS.

References
The Influence of β-Phase Conformation on Transient Absorption and Light Amplifying Properties of Polydiarylfuorene

Chen Sun, a Jinyi Lin, b Jaime J. Hernández Rueda, a Xingyuan Shi, c Aleksandr Perevedentsev, a and Juan Cabanillas-Gonzalez * a

During the past decades, π-conjugated polymers (CPs) and oligomers have attracted extensive attentions on account of their peculiar optical properties, high conductivities and low-cost fabrication process. Poly(9,9-dioctylfluorene) (PFO) is a typical blue light emitting polymer which can form β-phase conformation, resulting in an enhanced effective conjugation length, close molecular chain packing, and larger Förster radius of excitation transfer. However, PFO is not very stable and easy to form green band by photo-oxidation process. We use PODPF as a building block to investigate the relation between photophysical properties and chain conformation in solid state. Absorption and photoluminescence measurements are performed to estimate the β-phase content in films. Two-dimensional Grazing-Incidence Wide-Angle X-ray Scattering (2D-GIWAXS) diffractograms and Raman spectra are employed as a function of annealing temperature to monitor the structural properties of PODPF films. Continuous-wave photoinduced absorption (CW-PIA) and Raman imaging shows that β-phase domains of PODPF are isolated with each other at a long distance at relative low annealing temperature. The photophysics of annealed PODPF films are explained in terms of self-doped mixtures of α and β-phases where singlet excitons and polaron-pairs coexist as confirmed by femtosecond transient absorption (fs-TA) experiments. PODPF films thermally annealed at 220 ºC display outstanding dual ASE peaks ascribed to the α- and β-phase.

Figure 1. Differential transmission (ΔT/T) spectra of PODPF films: (a) untreated, (b) annealed at 210ºC, (c) annealed at 220ºC, (d) annealed at 230ºC, (e) annealed at 240ºC and (f) annealed at 250ºC at different pump-probe delays, (from dark to light colour): 2, 20 and 40 ps. The characteristic peaks ascribed to the α- and β-phase are indicated by the dotted and dashed lines, respectively.

Notes and References

High harmonic generation (HHG) represents a reliable mechanism to generate coherent high frequency radiation up to the extreme-ultraviolet or even soft x-rays. In atoms or molecules, HHG is based on the ionization-acceleration-recombination process of an electronic wavefunction driven by an intense laser field. This process is typically understood considering a single-active electron occupying outermost valence orbital of the atom/molecule under study. However, despite of the success of the single-active electron approximation (SAE), multi-electron effects can influence the ionization or recombination processes leading to HHG\textsuperscript{1,2}. In this work we describe a novel HHG mechanism based on electron correlation that has an unequivocal signature in the harmonic spectrum. We find that electron-correlated excitation to single-excited multielectron states of the neutral atom leads to the appearance of a high-energy secondary plateau in the HHG spectrum, beyond the single-active electron cutoff.

We consider two active electrons (TAE) in the numerical integration of the one-dimensional time-dependent Schrödinger equation in He. The emergence of a secondary plateau is evident both in the HHG spectrum (Fig. 1a, solid black line) and the time-frequency analysis (Fig. 1b). This high-energy plateau is absent when considering the SAE approximation (yellow solid line in Fig. 1a, and Fig 1c), and it is substantially more intense than the spectrum obtained from the contribution of the cation He\textsuperscript{+} (purple dashed line in Fig. 1a). This suggests that the secondary plateau originates from a multi-electron response. In fact, the double structure in the HHG spectrum is even preserved if one of the electrons is artificially disconnected from the electric field, which is a distinctive element compared to other previously reported mechanisms\textsuperscript{1,2}, and unequivocally demonstrates the fundamental role of electron correlation.

![Figure 1](image_url)

**Figure 1:** (a) HHG spectrum obtained in He considering two active electrons (He TAE, black solid line), a single-active electron (He SAE, yellow solid line) and (c) the exact description of He\textsuperscript{+} (purple dashed line). The incident laser pulse is modelled with a trapezoidal envelope of 8 cycles, 400 nm in wavelength and peak intensity of $5 \times 10^{14}$ W/cm\textsuperscript{2}. The secondary plateau originated from the multi-electron correlation is indicated. Time-frequencies analysis in TAE (b) and SAE (c) calculations are also shown.

**Notes and References**

The advances in laser technology have opened the way to perform time-resolved experiments with attosecond resolution (1 as = 10^{-18} seconds), enabling us to access ultrafast processes in matter and to obtain images of electron dynamics at its intrinsic time scale. Among the numerous applications of attosecond spectroscopic tools, the investigation of ultrafast charge migration occurring in biomolecules upon ionization has become a topic of increasing interest. The existence of a charge migration process occurring in an organic molecule was originally discussed in pioneer nanosecond pump-probe experiments performed by Weinkauf and collaborators [1]. Subsequent theoretical works involving several biomolecules, lead by Cederbaum and collaborators, demonstrated the occurrence of sub-femtosecond charge fluctuations after sudden ionization of a molecule as the result of electron correlation in the ion [2]. The first experimental evidence of charge migration was obtained using an attosecond UV pump/femtosecond IR probe scheme in phenylaniline, proving the existence of a sub-femtosecond ultrafast dynamics, which in contrast with previous predictions, was associated to a highly delocalized hole density involving one-hole states [3].

Within this context, we present our ongoing theoretical investigation exploring ultrafast electron dynamics in ionized glycine using an attosecond XUV-pump/XUV-probe scheme. We explicitly account for the pump and probe pulse interactions and include the nuclear degrees of freedom using a semi-classical approach. Most existing theoretical studies on charge migration on large molecules were performed within the fixed nuclei approximation, which implies that the description of the dynamics was only valid within the first few femtoseconds [4]. We here propose the use of a Surface Hopping approach [5], combined with a multi-configurational self-consistent field method to compute the electronic structure, to trace the electron-nuclear coupled dynamics of the molecule after ionization. A thorough study of the electronic populations and charge distribution of the cationic and dicationic species allows us to observe and identify the different fragmentation paths that arise after the action of the pump and the probe pulses.

Notes and References
The electronic transport across nanojunctions due to the Stark effect induced by short laser pulses was recently studied by Franco and Garzón. In this work, we analyse how the polarization of the materials forming the nanojunction, which had previously been neglected, affects this electronic transport. As we can see in the figure below, our results show that the charge transfer mechanism is robust to polarization, even if the total electronic transport is reduced.

FIGURE
Results for the non-polarized (dashed line) and polarized (solid line) systems. The effective electric field felt by the nanojunction is shown on the top. The total charge transferred from one side of the junction (A) to the other (B) is presented on the bottom.

Notes and References
POSTER

Ionization from a coherent superposition in CCl$_2$F$_2$

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The recent XCHEM$^1$ code allows the study of photoionization considering the effect of electron correlation in the configuration interaction picture and taking into account the coupling between several continua. It is especially appropriate for the study of low energy ionization, when the electron still feels the ionic core and electron correlation still plays a role; as recently shown in our study of the Hopfield resonant series in the nitrogen molecule$^2$.

At the core of the XCHEM method lies the hybrid Gaussian-B-spline basis$^3$ (GABS), used to represent the bound and continuum states of hydrogen with essentially exact results. It consists of a set of monocentric gaussians (MC) centered at the center of mass (CoM) of the molecule and a box of B-splines that ranges from a distance to the CoM $R_0$ to a sufficiently far distance $R_\infty$. The XCHEM model couples this basis with the polycentric gaussian (PC) basis that results from a standard quantum chemistry calculation (QCP). The $R_0$ distance is chosen so that the overlap between PC gaussians and B-splines is negligible. The MC gaussians serve as a bridge communicating B-splines and PC gaussians.

This model has been applied up to now to small systems: atoms like hydrogen, helium and neon$^4$ and diatomics such as hydrogen and nitrogen. In this communication we present the surface hopping dynamics of a coherent superposition in CCl$_2$F$_2$, which will be ionized with the XCHEM package.

Notes and References

POSTER

Time delays from one-photon transitions in the continuum

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Free electrons can interact with light only via Compton scattering. In a coulomb continuum, however, electrons can absorb or emit photons. Such continuum-continuum (cc) transitions [1] are commonly exploited in a variety of attosecond photoionization experiments to extract delays originating from the propagation of the liberated electron wave packet across the ionic potential [2].

We present an experimental study, which allows us to quantify the delay arising from different cc-transitions. To this end, we developed a novel experimental procedure which gives access to both amplitudes and phases of multiple interfering quantum pathways in a photoionization experiment.

(a) Schematic of the photoionization pathways contributing to a sideband in a RABBITT experiment in Helium. (b)- (d) Experimental anisotropy parameters ($\beta_0$, $\beta_2$, $\beta_4$) of the angle resolved RABBITT spectra. (e) Integrated photoelectron yield and retrieved phase difference between continuum-continuum transitions to s- and d-waves involving absorption.

We perform angle resolved photoelectron spectroscopy in Helium, following the RABBITT scheme [3]. As illustrated in Figure 1a, four quantum pathways contribute to each sideband, comprising the pathways with angular quantum numbers $s\rightarrow p\rightarrow s$ and $s\rightarrow p\rightarrow d$ for absorption and emission. This allows to reduce the angular dependence of the measured RABBITT spectra to a set of three anisotropy parameters (Figure 1b-d), for which analytic expression can be calculated within second order perturbation theory. A simultaneous fit of the anisotropy parameters then retrieves the amplitudes and relative phases of the four quantum pathways and resolves a delay between the different cc-transitions.

We measured a time delay between electrons arising from cc-transitions to a d-wave with respect to a s-wave in excellent agreement with the theoretical results from two independent methods: a single-active-electron [4] and an ab-initio [5] calculation. Moreover, simulations in atomic hydrogen suggest that the found angular momentum contribution to the photoionisation time delay has a universal character across different species.

This work not only serves as a proof-of-principle demonstration how to disentangle multiple interfering quantum pathways in photoionization experiments but also reveals new physical insight. To the best of our knowledge, we have for the first time directly measured time delays between one-photon transitions in the continuum.

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POSTER

The role of surface roughness in the formation of LIPSS on metals

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The present work addresses the controlled formation of Laser Induced Periodic Surface Structures (LIPSS) using femtosecond laser pulses in metals: copper and steel. In the former, we have identified a processing regime which produces two different periods formed at different local fluences1. Given that the simple plasmonic model2 overestimates the periods experimentally observed at oblique angles of incidence, we have introduced a novel predictive model. This model takes into account the influence of the surface’s roughness on the surface plasmon polariton (SPP) formation. The model results fit adequately the ripples periods so obtained2.

We have also investigated the formation of LIPSS in steel and discovered a new effect when processing the surface at oblique angle while scanning the sample, featuring a dependence of the ripple period on the scan direction. This phenomenon, coined quill writing or non-reciprocal writing3, allows us to select with high precision the ripples period just by inverting the scan direction4. The underlying mechanism is related to the asymmetry the roughness induced upon exposure at oblique incidence. As a consequence, different roughness at different positions generate different wavevectors of the surface plasmon polariton when scanning in different directions. In addition, we have adapted the roughness-plasmonic model to absorbent materials such as steel, using an effective dielectric constant.

Notes and References

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Signatures of matter Talbot effect in the high-order harmonic generation from periodic systems

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High harmonic generation (HHG) stands as a robust technique to generate extreme-ultraviolet/soft x-ray radiation in the form of attosecond pulses [1]. The highly coherent nature of the HHG process, based on electronic wavepacket ionization, acceleration and recombination, allows a detailed control of the emitted high frequency radiation. Recently, the possibility of HHG in crystalline solids has attracted the attention of the community [2,3]. In this work, we exploit the coherence effects of the ionized electronic wave packets to enhance the HHG emission. We explore the appearance of the Talbot effect [4] in the electronic wavepacket and monitor its spectral signature in the HHG. Following an analogy between the electromagnetic wave equation in the Fresnel approximation and the time-dependent Schrödinger equation (TDSE), we demonstrate that the electronic Talbot effect forms perfect images of the wavepacket distribution at different instants of time. Synchronizing this Talbot time with the wavefunction rescattering, we optimize the recombination probability, and thus, we enhance the high-order harmonic emission.

We solve the TDSE in a linear crystal in a two-dimensional space, considering a linearly polarized laser field, perpendicular to the direction of the array of atoms. In Figure 1(a-f) we show the evolution in time of the ionized wavepacket. Interestingly, we observe at t=0.48T the inverse Talbot image of the original wavefunction and, at t=0.72T, a perfect Talbot image, where the maxima of the wavepacket density coincide with the atomic positions in the crystal. Translated to HHG, the radiation efficiency depends on the electronic wavepacket excursion. It is possible to define the Talbot time (τT) as the excursion time needed by the electronic wavepacket to form a perfect image. In Figure 1(g), we show the HHG spectra versus the corresponding time of excursion (t_ex) of the classic trajectories associated to each harmonic, in units of τT, for the linear crystal (dashed blue), in comparison with the single-atom (solid orange). In the crystal case, we clearly observe a minimum for harmonics corresponding to excursion times near τT/2 (inverse Talbot image), while an enhancement for the emission corresponding to excursion times of τT (perfect Talbot image).

Figure 1. (a-f) Typical evolution of the electronic density of probability (logarithmic scale) in the X-Y plane for a linear crystal with a separation of 20 a.u. between atoms. Time is expressed in units of the period of the electric field (T). (g) HHG intensity for an incident frequency of 0.0328 a.u. and an amplitude of 0.067 a.u. as a function of the excursion time for the classic trajectories for a linear crystal (dashed blue) and a single atom (solid orange).

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Deciphering the ultrafast events in a new HOF with remarkable responsiveness to acids

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Hydrogen-bonded organic frameworks (HOFs) are highly crystalline materials for possible applications in science and technology. In this contribution, we will show results of ultrafast studies and single crystal fluorescence microscopy of an acid-responsive, extremely thermostable HOF (CPHATN-1a), with permanent porosity, by using hexaazatrinaphtylene derivative. The fluorescence decays of the HOF show three emission lifetimes. Moreover, the crystals exhibit a rich photochemistry as a result of intramolecular charge-transfer and interunit proton-transfer reactions, which we can elucidate with femtosecond (fs) experiments, demonstrating that these events occur in ≤200 fs and 1.2 ps, respectively. Fluorescence microscopy at the level of single crystals reveals a high anisotropic behavior and the importance of defects and crystallinity in their photodynamic. Remarkably, CPHATN-1a changes its color reversibly from yellow to reddish-brown when exposed to acids (Figure 1). Moreover, the fluorescence properties was also changed upon the exposure. This is, to the best of our knowledge, the first example of HOFs with external-stimuli responsiveness in color. The present results would open a door to develop new porous materials with stimuli responsiveness.

Figure 1. The background of the figure shows the the fundamental units of the framework. The central image illustrates the fluorescence color of several crystals. Change of HOF crystals color and their fluorescence after exposing to acid.

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References
THz spectroscopic imaging at kHz pixel rates

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The fastest imaging technique on the market. Combining High-Speed ASOPS and efficient THz generation antenna allows for spectroscopic imaging at 10 kpixels / second acquisition speed with an excellent signal to noise ratio. Being able to measure the phase and amplitude information at this high speed is unmatched by any other device on the market.

FIGURE: Comparison of the final image on the right consisting of 130 000 pixels to the photograph of the coin on the left.

Notes and References

References:
Commissioning of the pump-probe laser infrastructure of the Small Quantum System instrument at the European XFEL

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This contribution presents the pump-probe laser infrastructure of the Small Quantum System (SQS) instrument, which is one of six experimental end stations at the European x-ray free electron laser (XFEL) facility. This scientific platform is designed for investigations of atomic and molecular systems, as well as clusters, nanoparticles and small bio-molecules. It is located behind the SASE3 soft x-ray undulator, which provides horizontally polarized FEL radiation in a photon energy range between 260eV and 3000eV (4.8\text{nm} to 0.4\text{nm}) with up to 2\times10^{14} photons per pulse and up to 27000 pulses per second. Two high-quality elliptical mirrors in Kirkpatrick-Baez (KB) configuration focus the FEL beam to a spot size of approximately 1\text{um} in diameter. This results in an intensity of more than 10^{19}\text{W/cm}^2 within the interaction region that allows for studying non-linear multi-photon processes. The short FEL pulse duration between 2fs and 100fs in combination with a synchronized optical femtosecond laser are going to enable time-resolved studies of dynamic processes, thereby capturing the motion of electrons and nuclei with unprecedented resolution in space on ultrafast time scales.

First experiments have successfully been performed at the SQS end station, which also include the commissioning of the designated pump-laser laser system in combination with timing diagnostics based on the principal of spectral encoding, thereby monitoring the temporal jitter between optical and soft x-ray pulses. Centered around a wavelength of 1030\text{nm}, the fiber-based pump-probe laser amplifier delivers sub 40\text{fs} pulses with an energy of up to 1\text{mJ} at a repetition rate of 100kHz. A triggerable acousto-optical modulator (AOM) allows for mimicking the 10Hz bunch pattern of the XFEL, while a fiber link in combination with an optical cross correlator (OXC) enables femtosecond synchronization of the amplifier seed to the master oscillator. To quantify the jitter between the optical and the soft x-ray pulses accumulating along the respective beam paths, a dedicated pulse arrival monitor (PAM) has been placed into the SQS beamline downstream of the experimental chambers. Here, the soft x-ray pulses induce a change in the optical transmission of a few micron thin Si\text{3N}4 membrane, which allows for mapping the relative temporal delay onto the spectral coordinate of the stretched laser pulse also penetrating the sample membrane. Based on our recent measurements (fig. 1), the jitter has been determined to be at the order of tens of femtoseconds from shot-to-shot. Using PAM thereby opens the door for time-resolved single-shot experiments with femtosecond time resolution at the SQS end station of the European XFEL.

FIGURE: LEFT) spectra of stretched optical laser pulses transmitted through a Si\text{3N}4 membrane for temporal calibration, RIGHT) from white dashed line: single shot spectra for quantifying jitter between laser and soft x-ray pulses.

POSTER

Excited state dynamics of pyrrole-containing clusters

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The role of πσ∗ states in the electronic relaxation of photoexcited heteroaromatic molecules has attracted a great deal of attention since the publication of the seminal theoretical studies of Domcke and co-workers,1,2 due to the potential involvement of these states in the relaxation channels of biomolecules and their chromophores. In this sense, pyrrole (Py) has been extensively studied as a benchmark model, since its S1 π3s(N) Rydberg excitation, which acquires πσ∗ valence character as the N-H bond extends, is the lowest energy excited state.3

In this work, aiming to transfer some of the ideas gained about the dynamics in the isolated phase to the photophysical behaviour of Py aggregates and, also, to understand at microscopic level the role of solvation on the photophysical and photochemical properties of Py, the relaxation dynamics of the neutral aggregates Py(N-methylPy) and Py(H2O)1 have been tracked by femtosecond pump-probe ionization. The applied technique overcomes fragmentation issues and permits to obtain IR spectra of molecular complexes by combining high resolution ns tunable sources and high intensity fs pulses.

FIGURE: a) IR spectrum recorded at the Py(H2O)1+ mass channel. b) Transient recorded at the Py(H2O)1+ mass channel with the IR ns laser tuned at 3450 cm−1. The black dots represent the transient obtained, by substracting the IR ns laser on and off decays, by exciting at 244 nm and probing with 800 nm radiation. The solid red line corresponds to the exponential fit.

Notes and References

POSTER

Ultrafast x-ray scattering from molecular wavepackets

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X-rays have been widely exploited to unravel structure of matter since their discovery in 1895. Nowadays, with the emergence of new X-ray sources with higher intensity and very short pulse duration, notably XFELs (X-ray Free Electron Lasers), the number of experiments available in the X-ray regime has increased dramatically, even allowing the characterization of gas phase atoms and molecules in space and time.

Our aim is to study ultrafast X-ray scattering theoretically. Based on electronic structure ab-initio calculations we have developed tools to predict the signatures of atoms and molecules in both elastic, inelastic and total scattering. The combination of these methods with the state-of-the-art quantum dynamic methodologies can be used to accurately reproduce how excited wave packets evolve in time using pump-probe X-ray scattering experiments.4,5,6

The different components of the total scattering signal measured in an optical pump-x-ray probe experiment, i.e. elastic, inelastic and mixed coherent components, have a direct influence in the interpretation of molecular dynamics. In this talk, I will explain in detail how a theoretical framework based on a first-order perturbation theory and quantum electrodynamics can be used to extract these features from the ultrafast x-ray scattering picture. I will apply this theory to the study of H2 molecule dynamics, employing ab-initio and quantum dynamics simulations.7,8

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Time-resolved photodissociation dynamics of vinyl iodide in the UV at 199.2 and 200

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In this work, we have performed novel femtosecond time-resolved velocity map ion imaging experiments of vinyl iodide at 199.2 nm and 200 nm, corresponding to the first absorption maximum of the sharp trident-shaped transitions and the red edge of this broad peak, respectively.1, 2

Time-resolved I*(2P1/2) and I*(2P3/2) fragment images obtained through (2+1) REMPI have allowed us to compare the dynamics at both excitation wavelengths. The present time-resolved results reveal in both cases a fast photodissociation dynamics in the few hundred femtosecond timescale, with 50 fs longer reaction times at 200 nm than at 199.2 nm, and the involvement of different initial excited electronic states, disentangled from the clear change of the angular distribution of the iodine image after photodissociation, despite varying the excitation wavelength by just 0.8 nm. At 200 nm, photodissociation is governed by an initial parallel character transition, while at 199.2 nm a perpendicular transition is responsible for the dissociation process. Nevertheless, the contribution of non-adiabatic routes or of other electronic states upon excitation must be remarkable taking into account the values of the anisotropy parameters obtained at both excitation wavelengths.

Figure 1. Gas phase absorption spectrum of vinyl iodide in the 194-204 nm region. Adapted from Ref. [1]. The excitation wavelengths used in the present work are marked with vertical dashed lines.

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