



# USTS 2022

ULTRAFAST SCIENCE & TECHNOLOGY **SPAIN**  
2022

16<sup>th</sup> to 18<sup>th</sup> November 2022  
Málaga, Spain

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# WELCOME

Welcome to the fourth edition of the Ultrafast Science and Technology Spain 2022 (USTS 2022) meeting celebrated from 16<sup>th</sup> to 18<sup>th</sup> November 2022 in Málaga and organized by the Grupo Especializado de Láseres Ultrarrápidos (GELUR) of the Spanish Royal Society of Physics (RSEF) and the University of Málaga.



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The USTS 2022 conference has been promoted since its first edition by GELUR whose governing board is constituted by:

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The main objective of this event is to continue a series of meetings aimed at gathering and promoting the whole ultrafast laser community in Spain, providing a space for encounter, discussion and dissemination of the most recent results in ultrafast science, and facilitating synergies among groups with common themes, tools or methodologies. All senior scientists, postdocs and students working in areas related to ultrafast science are strongly encouraged to attend the meeting.

The scope of USTS 2022 is broad and multidisciplinary, covering different topics such as ultrafast laser development, femtosecond laser spectroscopy and microscopy, nonlinear optical phenomena, attosecond physics, ultrafast synchrotron and X-ray free-electron laser (XFEL) studies, and in general, any ultrafast study related to biology, chemistry and condensed matter.

# THE CONFERENCE VENUE

The congress is held at the rectorate of the University of Málaga (Avenida de Cervantes). The rectorate is located next to the park of Málaga, between the Palacio de la Aduana and the Bank of Spain headquarters, and behind the Alcazaba.



The rectorate building was built between 1916 and 1923 in a neo-Mudejar style and used to be the city's main post office (Antiguo Edificio de Correos). Nevertheless, due to space problems, the post company stop using the building in 1986. Years later, in 1993, it was acquired by the University of Málaga, which restored it between 1998 and 2002 and has been using it ever since then as its rectorate.

Under this building, remains of the Phoenician wall of Málaga dating from the third and fourth centuries BC and other elements such as roman garum (fish paste) pools have been found and can be seen from the inside of the building.

# PRACTICAL INFORMATION



The lectures take place in the conference room of the rectorate of the University of Málaga (Av. de Cervantes, 2, 29016 Málaga).



Poster sessions take place in the “Cofradía de Estudiantes” (Calle Alcazabilla, 3) on Wednesday 16<sup>th</sup> and Thursday 17<sup>th</sup> from 18:20 to 19:15 h.



There is Wi-Fi connection available: Access to Internet connecting to the Education Roaming (eduroam) wireless network.

## MEALS



Three coffee breaks, two lunches and a gala dinner are included in the registration fee. Please check the Conference program for time frames.

- The coffee breaks are served in the “Cofradía de Estudiantes” (Calle Alcazabilla, 3).
- The midday meals are served in “Bodega Bar El Pimpi” (Calle Postigo San Agustín, 8) on Wednesday 16<sup>th</sup> and in “Los Patios de Beatas” (Calle Beatas, 43) on Thursday 17<sup>th</sup>.
- The gala dinner is served in “La Reserva del Olivo” (Plaza del Carbón, 2) on Thursday 17<sup>th</sup> afternoon.



# COMMITTEE

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- **Rocío Ponce** Universidad de Málaga (UMA)
- **Samuel Valdivia** Universidad de Málaga (UMA)

# CONFERENCE PROGRAM

## WEDNESDAY 16<sup>TH</sup>

11:00	CHECK-IN
12:00	OPENING
	<b>SESSION 1-F. MARTÍN</b>
12:10	PL 1-A. L'HUILLIER
12:55	IL 1-R. MONTERO
13:20	LUNCH
	<b>SESSION 2-R. DE NALDA</b>
15:00	KL 1-A. PALACIOS
15:35	IL 2-L. PLAJA
16:00	OC 1-L. BAÑARES
16:15	OC 2-E. PALACINO
16:30	COFFEE BREAK
	<b>SESSION 3-L. PLAJA</b>
17:00	IL 3- W. GAWELDA
17:25	IL 4-P. PUYUELO
17:50	OC 3-A. ORDÓÑEZ
18:05	OC 4-S. GANGULY
18:20	POSTER SESSION I



## THURSDAY 17<sup>TH</sup>

	SESSION 4-J. SIEGEL
9:00	PL 2-K. SOKOLOWSKI-TINTEN
9:45	IL 5-J. TRULL
10:10	IL 6-A. JOHNSON
10:35	IL 7-I. SOLÁ
10:50	COFFEE BREAK
	SESSION 5-C. COJOCARU
11:35	IL 8-G. GONZÁLEZ
12:00	IL 9-B. COHEN
12:25	IL 10-J. CABANILLAS
12:50	OC 5-M. BALLABIO
13:05	OC 6-J. M. MARÍN-BELOQUI
13:20	LUNCH
	SESSION 6-W. GAWELDA
15:00	KL 2-R. BORREGO
15:35	IL 11-A. J. ALEJO
16:00	OC 7-I. SOLA
16:15	OC 8- A. MARTÍN-JIMÉNEZ
16:30	COFFEE BREAK
	SESSION 7-A. PALACIOS
17:00	IL 12-M. GARCÍA-LECHUGA
17:25	IL 13-G. MÍNGUEZ
17:50	OC 9-A. BURGOS
18:05	OC 10-O. PEÑA
18:20	POSTER SESSION II
20:30	CONFERENCE DINNER



## FRIDAY 18<sup>TH</sup>

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	SESSION 8-L. BAÑARES
9:00	PL 3-S. PASCARELLI
9:45	KL 3-K. ATTENKOFER
10:20	IL 14-G. FIGUEIRA
10:45	COFFEE BREAK
	SESSION 9-A. LONGARTE
11:15	IL 15-M. IZQUIERDO
11:40	IL 16-A. CURZIO
12:05	OC 11-R. DE NALDA
12:20	OC 12-M. RICO
12:35	CL-L. ROSO
13:20	CLOSURE

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# PLENARY LECTURES

# PL 1-Attosecond pulses for studying ultrafast electron dynamics

A. L'Huillier

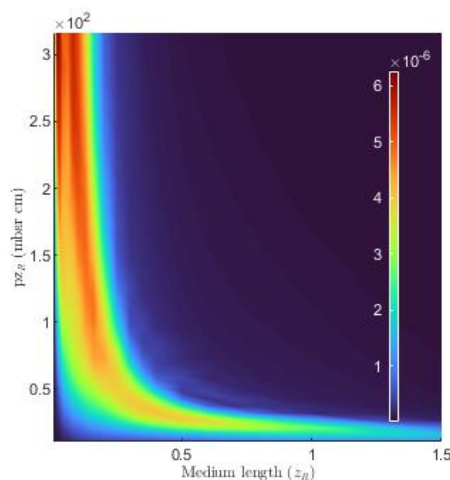
Physics Department, Lund University, SE-221 00 Lund, Sweden.

E-mail presenting author: [anne.lhuillier@fysik.lth.se](mailto:anne.lhuillier@fysik.lth.se)

Extreme ultraviolet light sources based on high-order harmonic generation (HHG) in gases are now used in many areas of science. The radiation consists of a train of extremely short light bursts, in the attosecond range, allowing for outstanding temporal resolution. HHG has opened the field of attosecond science, capturing ultrafast electron dynamics in matter. This presentation will give a short historical perspective on this field of research.

The HHG efficiency depends both on the response of a single atom to a strong laser field and, as in any coherent nonlinear optical process, on the phase matching between the waves emitted by individual atoms in a macroscopic medium. An introduction to the physics of HHG will be given, covering both aspects of the process. The importance of phase matching is illustrated in the Figure below, showing the conversion efficiency of the 23<sup>rd</sup> harmonic in Ar.<sup>1</sup> A good conversion efficiency can be obtained both with a long medium, low pressure and a short-medium, high pressure, allowing for versatile setups.

The short pulse duration and broad bandwidth of attosecond pulses allow the measurement of the phase and amplitude of an ionizing electronic wave packet using the RABBIT interferometric technique.<sup>2,3</sup> We will describe recent applications of this technique, towards high spectral resolution.



**Figure 1.** The conversion efficiency of the 23<sup>rd</sup> harmonic of 800 nm radiation generated in argon at an intensity of  $2.5 \times 10^{14}$  W/cm<sup>2</sup>.  $z_R$  denotes the Rayleigh length. Adapted from ref [1].

## REFERENCES

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- [2] Paul PM et al., Science 292, 1689 (2001)
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## PL 2-European XFEL: New science opportunities and first results

S. Pascarelli

European XFEL, Holzkoppel 4, Schenefeld, 22869 Germany.

E-mail presenting author: [sakura.pascarelli@xfel.eu](mailto:sakura.pascarelli@xfel.eu)

In the past decade, we have seen very important developments in the field of accelerator-based X-ray user facilities, with the advent of 4th generation synchrotron sources and MHz rate hard X-ray free electron lasers (FEL). Among the latter, the EuXFEL is the first hard XFEL powered by a superconducting linear accelerator, which enables MHz rate pulse generation, leading to an increase of more than 2 orders of magnitude in the number of delivered photon pulses per second to the sample.

A unique feature of X-ray FELs is the combination of the extremely short (fs) photon pulses with the short (Å) wavelengths of hard x-rays produced. These new user facilities are opening novel avenues in the investigation of fundamental processes in many areas of science, from physics to chemistry, biology and materials science, because they allow investigation of matter at the time scales of electron and nuclear dynamics (down to fs), with chemical selectivity and bulk sensitivity.

After many years of construction, user operation at the EuXFEL has finally begun. All the accelerator design parameters have been achieved, and between the fall of 2017 and the summer of 2019, six instruments started user operation. Applications address fundamental processes of light-matter interaction, ultra-fast dynamic processes in liquids and solids, investigation of electronic and structural properties and their dynamics in biomolecules, nanostructures, disordered materials and high energy density states of matter.

In this talk, I will first briefly introduce the European XFEL facility and report on the first results from early user experiments. Finally, I will comment on some important challenges ahead.



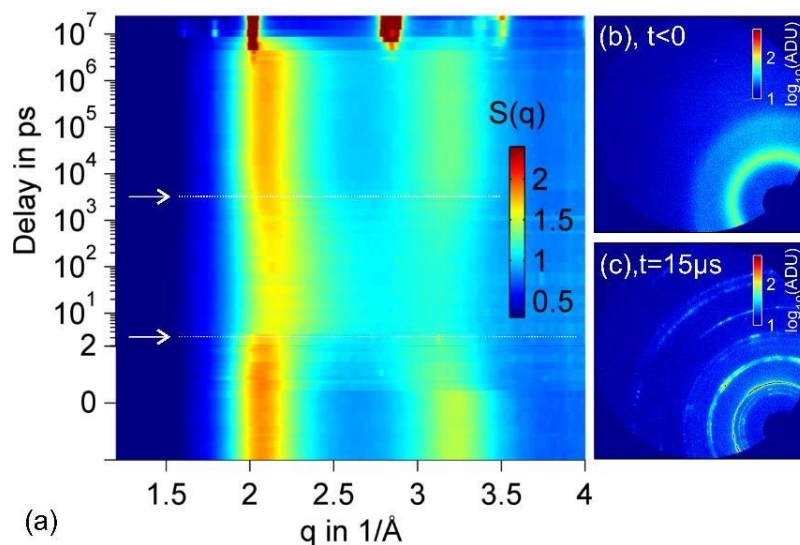
# PL 3-Irreversible material dynamics studied with time-resolved X-ray and electron scattering

K. Sokolowski-Tinten

Faculty of Physics and Center for Nanointegration Duisburg-Essen, University of Duisburg-Essen, Lotharstrasse 1, 47057 Duisburg, Germany.

E-mail presenting author: [klaus.sokolowski-tinten@uni-due.de](mailto:klaus.sokolowski-tinten@uni-due.de)

Excitation of solid materials with ultrashort pulses represents an efficient way to generate states of high energy density, which are characterized by an initially extreme electronic excitation. The subsequent relaxation can lead to phase transitions on very short time scales under conditions far away from thermal equilibrium. In this contribution, I will present a few examples from our recent work on the non-equilibrium structural response of laser-excited materials, which we study by ultrafast electron- and X-ray diffraction.



The Figure shows as an example the evolution of the atomic structure of the phase change material AIST after fs laser excitation, as measured by ultrafast X-ray scattering at LCLS X-ray free electron laser. (a) Structure factor  $S(q)$  of AIST throughout the entire laser-induced melt-quench cycle from the initial as-deposited amorphous state (b) to the final crystalline state (c). Analysis of the data reveals a liquid-liquid phase transition in the deeply supercooled state.<sup>1</sup>

## REFERENCES

[1] Zalden, P et al. *Science* **2019**, *364*, 1062–1067

# CLOSING LECTURE

# CL 1-Ultrafast ultraintense lasers: The new frontier

L. Roso<sup>a,b</sup>

<sup>a</sup> Centro de Láseres Pulsados, CLPU, E-37185 Villamayor, Spain.

<sup>b</sup> Universidad de Salamanca, Departamento de Física Aplicada, E 37008 Salamanca, Spain.

E-mail presenting author: [roso@usal.es](mailto:roso@usal.es)

Ultraintense ultrafast lasers (tens of femtoseconds) are evolving quite dramatically. A petawatt laser is operative at Salamanca and now multipetawatt is the frontier, with several lasers close to ten petawatt operative or almost operative around the world. The world intensity record is 1023 W/cm<sup>2</sup> achieved in the Korean multi-petawatt laser.

As normal in science, as power increases, more questions appear. In this talk, we are going to give a general view of this field and we will focus our discussion in two points: what are those multi petawatts good for; and how can we measure such extreme intensities. All that is related to beam quality, and the two points end in common questions, what is the optical quality of such lasers, how to measure it, and how laser parameters can be adjusted for a given application.

It is clear that petawatt lasers have relevant applicability for particle acceleration. With a petawatt, it is possible to accelerate electrons to several GeV and protons to tens of MeV. In this direction the frontier land is not to increase the peak power but to improve the beam quality and, particularly relevant, the repetition rate. Without a high repetition rate is not possible to develop a technology suitable for industrial applications.

For example, new beam profiles such as OAM intense beams are now feasible at extreme powers, and they open a new portfolio of possibilities.

However, besides particle acceleration and plasma physics, there are other possibilities. Have you considered that a multipetawatt laser can be used to analyze new concepts such as radiation reaction or the quantum vacuum and certain kinds of dark matter? This is going to be briefly discussed in this talk.

Another problem of general interest is metrology. At such extreme fields, it is impossible to introduce any meter because its atoms would be instantaneously ionized, and the meter will be destroyed. The only thing that survives is the electrons. In this talk, we will comment on new metrology tools based on relativistic electrons. We suggest the name of relativistic metrology for that.

To end this review talk we will briefly mention that some of those extreme lasers are open to users (though collaborations or through competitive access mechanisms) in Spain (CLPU), in Europe (Laserlab Europe), and in the US (US Lasernet).

As a conclusion, ultrafast ultra-intense lasers are a new and rapidly evolving frontier of our knowledge, and this is going to be briefly reviewed here to expand the scope of future editions of USTS.

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# KEYNOTE LECTURES

# KL 1-XUV and X-ray induced ultrafast electron dynamics: state-of-the-art theoretical methods for attosecond science

A. Palacios

Department of Chemistry, Universidad Autónoma de Madrid, Cantoblanco 28049, Madrid, Spain.

E-mail presenting author: [alicia.palacios@uam.es](mailto:alicia.palacios@uam.es)

Imaging electron and ultrafast nuclear dynamics in atoms and molecules with attosecond time resolution is the fundamental step in order to design realistic strategies to achieve control of relevant processes that occur on longer time scales.<sup>1-5</sup> Currently available femtosecond and attosecond technologies have demonstrated, by means of different protocols, that it is possible to control electron dynamics in atoms,<sup>4,5</sup> small molecules,<sup>6-8</sup> and even in biologically relevant complex systems.<sup>2,3,6,9</sup> The most extended technique is the attosecond pump-probe spectroscopy where an attosecond or few-fs XUV pulse ionizes the molecule and a time-delayed fs IR pulse probes the pumped dynamics.<sup>2,6,7</sup> Novel approaches are introducing few-fs IR, XUV and even X-ray pulses to probe the dynamics generated by the XUV pulse. Significant experimental efforts are also focused on generating few-fs pulses in a wide range of frequencies, from VIS (using high-order harmonic generation techniques- HHG) to hard X-ray (in free electron lasers - FEL), increasing the experimental toolbox that allows us to ignite chemical reactions that eventually follow quite distinct pathways. In the case of molecules, the use of ultrashort pulses leads to the sudden ionization or excitation of the target, creating a charge hole that subsequently migrates on a time scale ranging from few-femtoseconds down to hundreds of attoseconds. Depending on the initial wave packet that is created, which can be tailored by changing the pulse characteristics, the interplay between the electronic and nuclear dynamics can lead to the final localization of the charge on one specific molecular site or another, thus leading to different molecular fragments or different electron/charge transfer processes. In order to interpret, guide and predict the outcome of this plethora of XUV-induced ultrafast processes triggered in atoms and molecules, solid theoretical support is required. This talk will review the state-of-the-art theoretical methods, as well as their most significant applications in the last few years, which are capable of accurately describing these ultrafast phenomena in atoms and molecules when employing the most advanced coherent laser sources. The talk will focus on gas-phase experiments realized in table-top HHG experimental set-ups, which are able to produce attosecond pulses as short as a few tens of attoseconds, and in FEL and X-FEL facilities, which enables the study of a manifold of non-linear phenomena experimentally inaccessible until now, thanks to their high brilliance and intensity.

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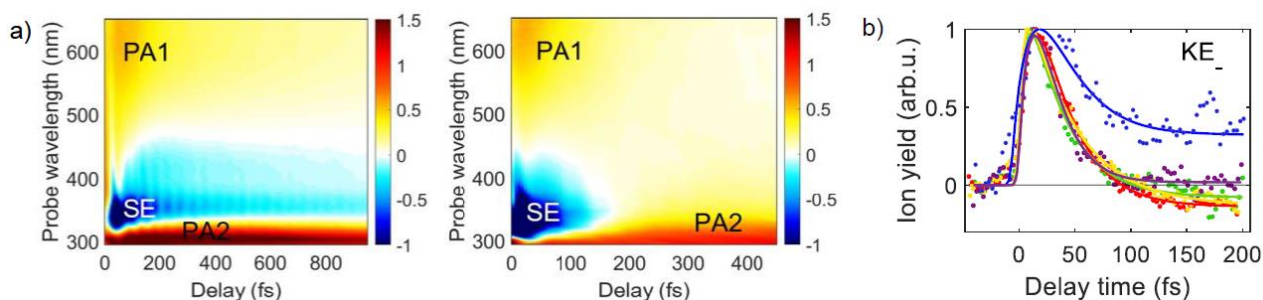
## KL 2-Ultrafast nuclear and electron dynamics in aromatic molecules

R. Borrego-Varillas

IFN-CNR, Piazza Leonardo da Vinci 32, 20133 Milan, Italy).

E-mail presenting author: [rocio.borregovarillas@cnr.it](mailto:rocio.borregovarillas@cnr.it)

Many fundamental processes in nature occur on time scales ranging from femtosecond (nuclear motion) to attosecond (electron motion) time scales.<sup>1</sup> Some prominent examples are photoinduced charge transfer (a process at the basis of core technologies aiming at effective conversion of solar energy into electrical energy) or the photo-protection mechanism of DNA. The deep understanding of these processes is strongly linked to technological advancements in the field of ultrafast spectroscopy. In this talk I will present our recent developments on the fields of femtosecond and attosecond spectroscopy and their application to the investigation of two fundamental physical processes. In a first example, we demonstrate a transient absorption spectroscopy in the ultraviolet (UV) range with sub-20 fs temporal resolution<sup>2-4</sup> and applied it to the investigation of the DNA photoprotection mechanism. DNA bases absorb strongly in the UV range but present remarkable photo-stability. These chromophores indeed relax in ultrafast time scales (tens of femtoseconds) after UV photo-excitation, preventing photochemical reactions that could end up on a photo-damage of the molecule.<sup>5</sup> In those ultrafast processes, conical intersections (CI) play a crucial role but tracing them is extremely challenging as it calls for ultrashort pulses in the UV range.<sup>6</sup> Thanks to the high temporal resolution of our setup, we have tracked for the first time the wave-packet motion through a CI in DNA nucleosides.<sup>7</sup>



**Figure 1.** a) Experimental transient absorption maps for the nucleosides thymidine (left) and uridine (right). b) Ion yield of the NO<sup>+</sup> photofragment versus pump-probe delay. Solid lines are the fitting curves with an exponential relaxation time of 36 fs for p-nitroaniline (green), m-nitroaniline (purple), N,N-dimethyl-4-nitroaniline (red) and 4-amino-4'-nitrobiphenyl (yellow) 30 fs plus an offset for 4-dimethylamino-4'-nitrobiphenyl (blue).

In a second scientific case, we aim at real-time imaging of the electronic motion in push-pull molecules. To this end we combine time-resolved photo-ion spectroscopy and synchrotron spectroscopies for investigating the electronic structure and ultrafast dynamics initiated by XUV radiation in nitroanilines, the simplest prototype donor and acceptor systems. Ultrafast dynamics are initiated by a short train of attosecond pulses (<2 fs) and probed with a 5-fs near-infrared (NIR) pulse. We observe two different dissociation processes. A first fragmentation channel, responsible for the central peak, involves the generation of NO<sup>+</sup> and the radical C<sub>6</sub>H<sub>4</sub>NH<sub>2</sub>.<sup>8</sup> The side peaks, instead, are assigned to Coulomb explosion of the NO fragment coming from a double ionization process. Photoelectron photoion coincidence experiments confirm this interpretation. Our results open the way to the investigation and control of electron transfer and charge transfer in push-pull molecules at the electronic time scale.

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# KL 3-Structure determination along the reaction pathway: Key to design functional chemical compounds

K. Attenkofer

ALBA synchrotron light source, Carrer de la Llum 2-26, 08290 Cerdanyola del Vallès, Barcelona, Spain.

E-mail presenting author: [KAttenkofer@cells.es](mailto:KAttenkofer@cells.es)

Undergoing a chemical reaction is often a chain of structural and electronic changes of the involved molecules, surfaces and interfaces requiring the breaking and reforming of chemical bonds and the transport of individual fragments and ions. Visualizing the structure and the correlated electronic state of these metastable states is at the center of understanding the reaction and a first step toward optimizing the conditions and tailoring the components for specific reaction products with the hope that individual components like catalysts, facilitating specific reactions, can be rationally designed.

Besides other local probes, X-ray spectroscopy in combination with a laser pump has been proven as a useful tool to characterize these intermediates for materials in solution<sup>1</sup> which go through a photochemical reaction or excitation.<sup>2</sup> In the case of heterogeneous catalysis, a key technology for producing most of our commodity chemicals and the basis for carbon neutral circular economies, the reaction is often thermally or electrically driven making the trigger significantly more difficult. Moreover, the complex interactions of the different gas or liquid components with the surface of the catalyst makes the structural interpretation of the spectral results significantly more difficult.

This talk will discuss two approaches to overcome these two challenges: 1) modulation spectroscopy, a method which modulates periodically the reaction conditions and detects the spectroscopic features in fixed phase in respect to the modulation, as a way to overcome the trigger challenges of classical pump-probe experiments.<sup>3</sup> 2) Data analytics to solve the structure of the active site based on spectroscopic data and massive theoretical data.<sup>4</sup>

Besides an explanation of the methods, the talk will also discuss the limits and the future developments necessary to develop this method to a standard technique applicable by the public user.

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# INVITED LECTURES

# IL 1-Real-time observation of electron photodetachment in solution

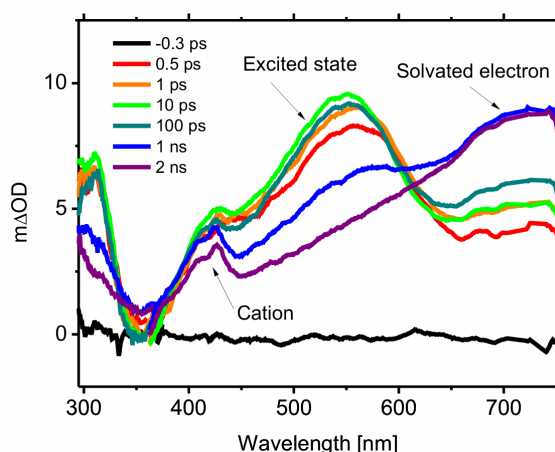
R. Montero,<sup>\*a</sup> I. Lamas<sup>b</sup> and A. Longarte<sup>b</sup>

<sup>a</sup>SGIKER Laser Facility (UPV/EHU), Sarriena s/n, 48940 Leioa, Spain.

<sup>b</sup>Departamento de Química Física, Universidad del País Vasco (UPV/EHU), Sarriena s/n, 48940 Leioa, Spain.

E-mail presenting author: [raul.montero@ehu.eus](mailto:raul.montero@ehu.eus)

Since its discovery by Hart et al.<sup>1</sup> the nature of solvated electron has attracted a great deal of scientific interest due to its relevance in Chemistry and Biology.<sup>2,3</sup> In neat solvents, and particularly in water, hydrated electrons can be formed by VUV radiation or by UV two photon absorption,<sup>4</sup> and its appearance can be easily detected owing to its well known optical absorption.<sup>5</sup> Additionally, the use of anion solutions is a very efficient method for the production of hydrated electrons, being I<sup>-</sup> the most paradigmatic case.<sup>6</sup> Although the details of the electron detachment mechanism are less known, the ability of some simple organic molecules solvated in water to ionize has been observed after excitation along the nearest absorption of their UV spectra, well below the measured gas phase ionization potential. In this contribution, we describe the case of aniline, whose electron emission dynamics after the UV excitation has been tracked by pump-probe broadband transient absorption (TA) methods. The complex TA spectrum recorded at the earliest delay times is analyzed by using a pump-repump-probe scheme that permits to disentangle the nature of the contributing species. The collected data indicate that in parallel to the long-living  $\pi\pi^*$  state responsible of the fluorescence, a charge transfer to solvent state (CTTS) is formed shortly after the excitation. The photoionization of the molecule will be discussed in terms of the electronic nature of this charge-transfer character state and the influence of the water-solute interactions.



**Figure 1.** Transient absorbance of aqueous aniline after 267 nm excitation.

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# IL 2-Symmetry matters! High-order harmonic generation from low-dimensional crystals driven by structured light

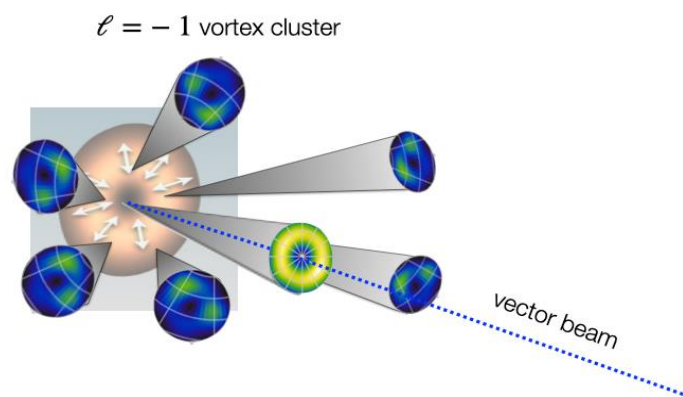
L. Plaja

Grupo de Investigación en Aplicaciones del Láser y Fotónica, Departamento de Física Aplicada, Universidad de Salamanca, E-37008, Salamanca, Spain.

E-mail presenting author: [lpaja@usal.es](mailto:lpaja@usal.es)

One of the most striking consequences of the interaction of matter with intense fields is the generation of high-order harmonics with efficiencies orders of magnitude above the perturbative expectation. Until recently, HHG has been mainly studied in atomic, molecular gases and plasmas. The experimental demonstration a decade ago of HHG in solids<sup>1</sup> sparked the interest in the study of the non-perturbative optical phenomena in crystalline solids. Due to their higher electron density, an obvious advantage of solids is the increased efficiency of the process. On the other hand, also during this last decade there has been also a considerable interest in exploiting HHG as a new route for the production of structured high-frequency harmonic radiation. Structured light has a non-trivial, and sometimes interleaved, distribution of spin and angular momentum. In this sense, HHG offers an extraordinary playground, as is capable to map some of the structural parameters of the driving field to the harmonic field. In particular, gas targets irradiated by vector beams have been demonstrated to map the vectorial driver's symmetry to high-order harmonics.<sup>2</sup>

One interesting aspect in HHG from crystalline targets is to study the interplay of the target symmetries with the symmetries of the structured light. In this sense, low dimensional crystals offer an extraordinary scenario, as their narrow widths (typically of atomic size) exclude propagation phenomena that may obscure relevant phenomena. In this talk we shall analyze the characteristics of HHG from graphene irradiated by radial vector beams. We shall demonstrate that graphene's non-linear anisotropy<sup>3,4</sup> arising from its crystalline structure, couples with the azimuthally variant polarization of the driver beam to generate a complex structured harmonic field. We interpret this structure as the superposition of a central vector beam with a topological background structured as a cluster of vortices (see figure).



**Figure 1.** Scheme of the multibeam geometry of the harmonic field driven by a radial vector beam in graphene.

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# IL 3-Unravelling excited states dynamics in base metal dyads for photocatalytic proton reduction using ultrafast X-ray spectroscopy

W. Gawelda,<sup>a,b,c</sup> M. Nowakowski,<sup>d</sup> T. Kyu Choi,<sup>e</sup> H. Elgabarty,<sup>d</sup> M. Huber-Gedert,<sup>d</sup> J. Steube,<sup>d</sup> D. Khakhulin,<sup>e</sup> M. Alaraby Salem,<sup>d</sup> M. Biednov,<sup>e</sup> F. A. Lima,<sup>e</sup> A. Kertmen,<sup>c</sup> N. Pierunek,<sup>c</sup> J. Kubicki,<sup>c</sup> T. Kühne,<sup>d</sup> and M. Bauer.<sup>d</sup>

<sup>a</sup> Departamento de Química, Universidad Autónoma de Madrid, 28049 Madrid, Spain.

<sup>b</sup> IMDEA Nanociencia, 28049 Madrid, Spain.

<sup>c</sup> Faculty of Physics, Adam Mickiewicz University, 61-614 Poznań, Poland.

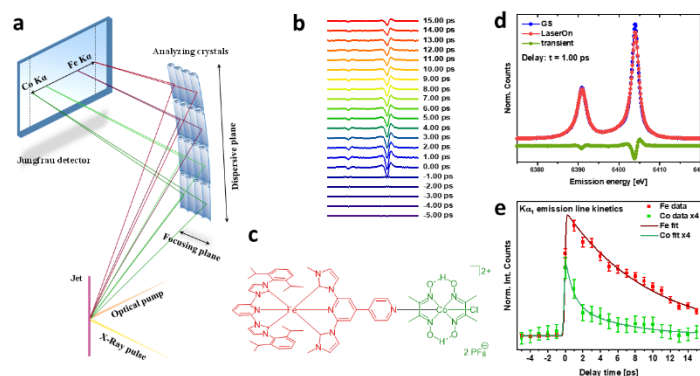
<sup>d</sup> Department Chemie und Center for Sustainable Systems Design, Universität Paderborn, 33098 Paderborn, Germany.

<sup>e</sup> European XFEL, 22869 Schenefeld, Germany.

E-mail presenting author: [wojciech.gawelda@uam.es](mailto:wojciech.gawelda@uam.es)

Base metal photoactive compounds are sustainable alternatives to their noble metal counterparts. Bimetallic assemblies of the later are often used for direct conversion of sun light into chemical energy carriers like H<sub>2</sub>. The transition to base metal dyads as active compounds is partially hindered by the so far missing understanding of the working principle of such hetero-bimetallic complexes, which is required to improve their photocatalytic performance. Therefore, more detailed investigation of the ultrafast photoinduced electron transfer (ET) is of utmost importance on the way to active base metal dyads for photocatalytic ET reactions.

In this study, we focused our attention on FeCo molecular dyads. We used femtosecond X-ray emission spectroscopy (XES) in combined optical pump/X-ray probe experiments at X-ray free electron lasers (XFELs) to track the electronic and structural dynamics in a bimetallic assembly, connecting an Fe photosensitizer (PS) to a Co catalyst. Due to the very fast processes, partially unknown excited states at the iron PS and the optically dark states at the cobaloxime catalyst, an ultrafast two-color emission experiment is applied for the first time, detecting simultaneously the Fe and Co X-ray emission on femtosecond timescales to avoid ambiguities about time zero. In this manner it is possible to unequivocally correlate the ultrafast excited state dynamics to ET processes within the dyad. The X-ray results are complemented with ultrafast UV-VIS transient absorption data to correlate the observed changes in the electronic and geometric structures of the dyad with the population dynamics derived from optical data.



**Figure 1.** (a) Schematic setup of the experiment showing the von Hamos geometry of the dispersive XES spectrometer; (b) A time series of Fe Kα<sub>1</sub> ΔXES spectra as a function of pump-probe time delay; (c) Structure of the investigated Fe-Co dyad; (d) Fe Kα<sub>1</sub> XES ground and excited state spectra and a corresponding transient spectrum (bottom) at Δt = 1 ps; (e) Long timescale kinetics of Fe and Co Kα<sub>1</sub> lines with fitted multiexponential decay functions.

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# IL 4-Thin, flat water target capable of high-repetition-rate MeV-range proton acceleration in a high-power laser

P. Puyuelo-Valdes,<sup>\*a</sup> D. de Luis,<sup>a</sup> J. Hernandez,<sup>a</sup> J. I. Apiñaniz,<sup>a</sup> A. Curcio,<sup>a</sup> J. L. Henares,<sup>a</sup> M. Huault,<sup>a</sup> J. A. Pérez-Hernández,<sup>a</sup> L. Roso,<sup>a</sup> G. Gatti<sup>a</sup> and L. Volpe<sup>a</sup>

<sup>a</sup> Centro de Láseres Pulsados, Building M5, Science Park, Calle Adaja 8, 37185 Villamayor, Salamanca, Spain.

E-mail presenting author: [ppuyuelo@clpu.es](mailto:ppuyuelo@clpu.es)

Laser-driven ion acceleration has been studied in depth in the last decade due to a large potential range of applications, from fundamental science to medicine. Traditionally, high-power laser facilities were only available at low repetition rate (LRR). Thin metallic solid targets were usually used due to their simple fabrication and ability to produce high-quality ion beams using Target Normal Sheath Acceleration (TNSA) mechanism.<sup>1</sup> However, after the interaction the target is destroyed and realignment of a new one is mandatory. Moreover, the debris from the interaction could damage the surrounding optics and detectors.

A large effort has been made to develop high repetition rate (HRR) targets: cryogenic ribbons,<sup>2,3</sup> water droplets,<sup>4,5,6</sup> liquid crystal films,<sup>7,8</sup> bulk glass targets<sup>9</sup> or even gas-jet targets.<sup>10,11</sup> These targets can regenerate *in situ* and could be a clean source of protons, free of contaminants and could operate at HRR. However, an effort of research and development is needed to understand, test and optimized such HRR targets.

In this work, we use a versatile quasi-planar water target from the collision of two equal diameter and velocity liquid microjets. This target is easy to align by shadowgraphy techniques, the thickness can be controlled (submicron range) and the ablated interaction region refresh in tens of  $\mu\text{m}$ .<sup>12</sup>

Up to now, 2 MeV proton energy with an ethylene glycol sheet target was reported<sup>13</sup> in *low vacuum* conditions (3 mbar). Here, we report proton acceleration up to 4 MeV from the interaction of the HRR VEGA-2 laser with the HRR water sheet target in *high vacuum* conditions ( $10^{-4}$  mbar). These results are published in ref [14].

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# IL 5-Harmonic generation in nanolayers with femtosecond pulses

J. Trull,<sup>a</sup> L. Rodríguez-Suné,<sup>a</sup> M. Scalora<sup>b</sup> and C. Cojocaru<sup>a</sup>

<sup>a</sup> Department of Physics, Universitat Politècnica de Catalunya, Rambla Sant Nebridi 22, 08222 Terrassa, Spain.

<sup>b</sup> Aviation and Missile Center, US Army CCDC, Redstone Arsenal, AL 35898-5000 USA.

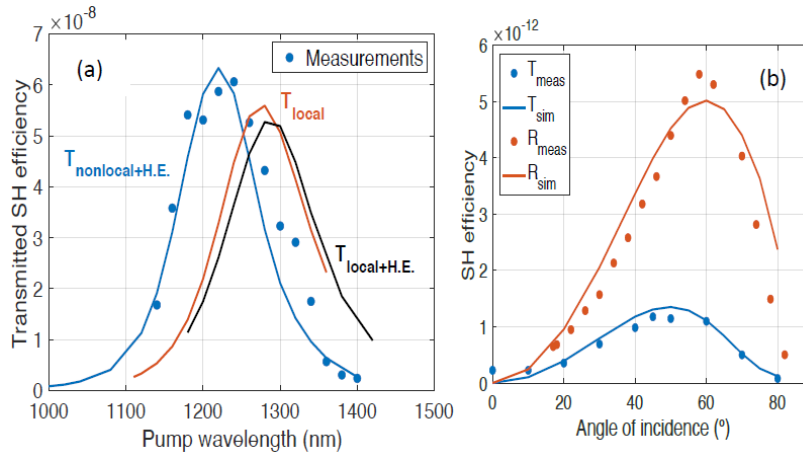
E-mail presenting author: [jose.francisco.trull@upc.edu](mailto:jose.francisco.trull@upc.edu)

At the nanoscale, light-matter interactions display physical phenomena that may not be explained by conventional approximations. This is the case of typical nonlinear processes like second harmonic generation (SHG) and third harmonic generation (THG) that have been traditionally studied under phase-matched (PM) conditions, primarily to improve efficiencies in bulk, optically thick materials with large nonlinearities and low material absorption. Under these circumstances, the leading nonlinear polarization term corresponds to the bulk contribution described by dipolar second and third order nonlinear susceptibility tensors ( $\chi^{(2)}$  and  $\chi^{(3)}$ ).

However, at the nanoscale and under pulsed illumination, PM may play no role, light can propagate and resonate even in spectral ranges that are usually opaque, and surface nonlinear effects may be as efficient as the bulk components. The effective  $\chi^{(2)}$  and  $\chi^{(3)}$  may not coincide with their bulk, local counterparts. In addition, contributions to the nonlinear polarizations arising from electric quadrupole-like and magnetic sources should also be considered. A detailed analysis and understanding of SHG and THG from experimental and theoretical points of view would be very useful to further integrate these materials in the design of nanometric devices.

We report a collection of experimental results of nonlinear harmonic generation at nanoscale. The comparison of these experimental results with numerical predictions of our theoretical model identifies, distinguishes and explains the different nonlinear contributions to the harmonics generated by these materials. Our model accounts for surface, magnetic and bulk nonlinearities arising from free and bound charges, nonlocal effects due to pressure and viscosity, convective terms or temperature dependent effective masses.

We have studied different materials under this common approach: (i) Harmonic generation by nm-thick Au layers. (ii) harmonic generation on conductive oxides (Indium Tin Oxide (ITO) at epsilon-near-zero condition (iii) Harmonic generation in semiconductors, GaAs and Si, in the opaque transmission regime of the harmonic radiation. Some examples of the measured efficiencies compared to the simulations are shown in Figure 1.



**Figure 1.** (a) Experimental measurements of transmitted SH efficiency as a function of the incident wavelength from an ITO nanolayer and numerical simulations for three different scenarios: taking nonlocal effects and hot electrons into account (blue), considering only the hot electrons contributions (black), and neglecting both nonlocal effects and hot electrons (red). (b) transmitted and reflected SH efficiency in 20 nm Au as a function of the angle of incidence

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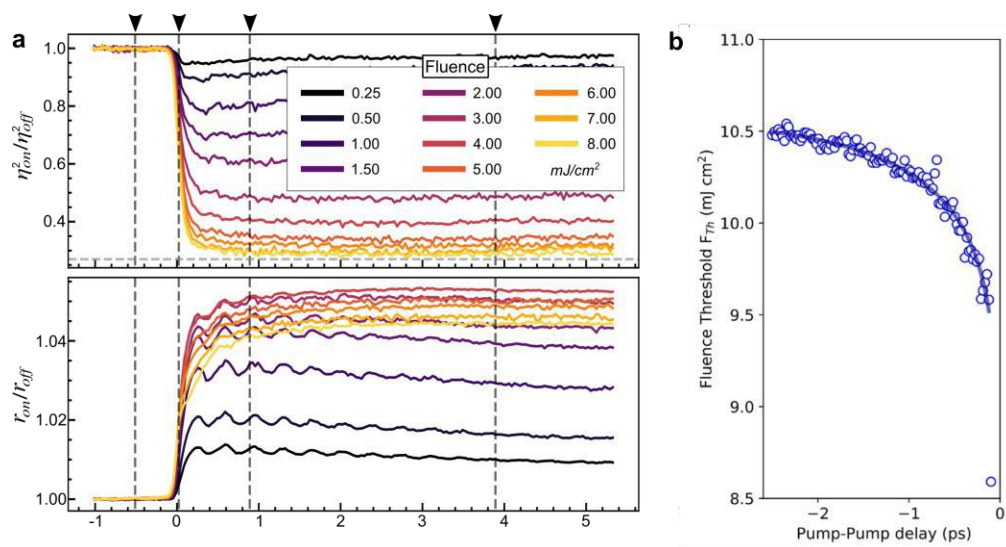
# IL 6-Light-induced phase transitions: disorder, inhomogeneity and control

A. S. Johnson

ICFO-Institut de Ciències Fotòniques, The Barcelona Institute of Science and Technology, Castelldefels, Barcelona, Spain.

E-mail presenting author: [allan.johnson@icfo.eu](mailto:allan.johnson@icfo.eu)

In the last decade ultrafast laser excitation has emerged as a powerful method for controlling the properties of quantum materials, driving phase transitions, and creating new out-of-equilibrium phases that have no equilibrium analogues. These systems are often understood in terms of a displacive or coherent type mechanism,<sup>1,2</sup> where the ultrafast excitation induces correlated motion in selected degrees of freedom quite unlike in the thermal transition. However, there is a growing body of evidence that even on the femtosecond timescale, disordering can dominate light-induced phase transitions in quantum materials. Furthermore, dynamic inhomogeneity at the nanoscale is also believed to place a key role in light-induced phase transitions,<sup>2</sup> further complicating the situation. In this talk I will describe some recent work characterizing disorder and inhomogeneity during light-induced phase transitions in a variety of quantum materials.<sup>3-5</sup> We find that that disorder can dominate the transition in wide variety of systems (LSMO,  $\text{V}_2\text{O}_3$  and  $\text{VO}_2$ ) which do not share any of crystal structure, charge state, or spin state, pointing towards the ubiquity of the process. This naturally challenges how applicable light-based control methods will be for selectively guiding the phase of quantum materials. However, I will also present results showing how disorder can actually be used to control the light-induced phase transition in  $\text{VO}_2$ . A double-pump scheme generating optical polarons suppresses the total energy barrier for the phase transition by over 10% and suggests that while coherent control methods may have to be re-imagined in quantum materials,<sup>6</sup> light-based control of such systems is still feasible.



**Figure 1.** (a) Simultaneous measurement of order parameter and reflectivity dynamics in LSMO. The order parameter shows an incoherent response reflective of disordering, while coherent optical phonons can still be observed in the reflectivity. (b) Total energy threshold for inducing the insulator to metal phase transition in  $\text{VO}_2$  as a function of pump-pump time delay. The normal single pump threshold is slightly above 10.5 mJ/cm<sup>2</sup>, showing that at short time delays there is a significant suppression of the barrier energy.

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# IL 7-Entangling gates with atoms: spatio-temporal control of Rydberg interaction

I. Sola<sup>a</sup> and B. Chang<sup>\*b</sup>

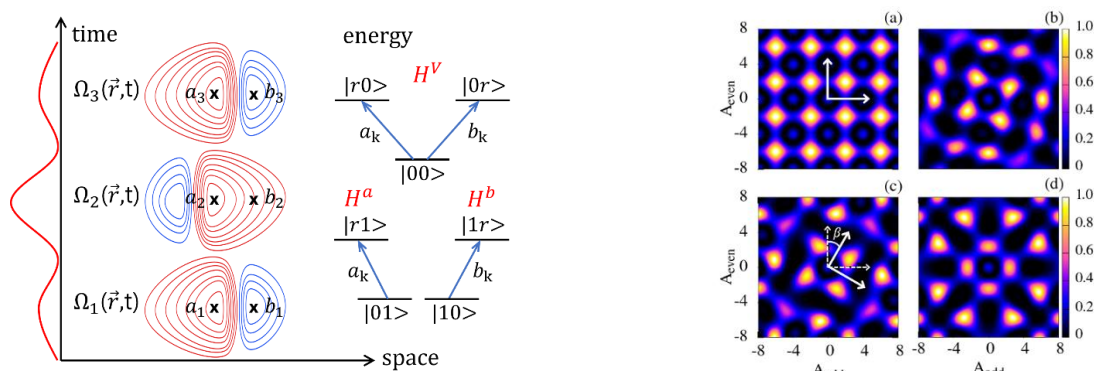
<sup>a</sup> Departamento de Química Física, Universidad Complutense, 28040 Madrid, Spain.

<sup>b</sup> School of Chemistry, Seoul National University, Seoul 08826, Republic of Korea.

E-mail presenting author: [isolarei@ucm.es](mailto:isolarei@ucm.es)

Atoms trapped by optical tweezers, interacting through Rydberg blockade, can be used to generate few multi-particle entanglement and simple quantum circuits.<sup>1,2</sup> To further the quest of the quantum computer, we need to improve the system addressability and controllability. One of the problems in the usual designs with ordered arrays of atoms largely separated in optical traps, is the long duration of the two-qubit gates, which must operate near the microsecond regime to avoid exciting more than one atom in a Rydberg state using the dipole blockade mechanism.<sup>3</sup> Optimal control theory can be used to find efficient and robust pulse sequences,<sup>4</sup> but it remains very challenging to accelerate the processes without changing the set-up. In this work we speed up the gates by using denser arrays of atoms that allow to boost the dipole-blockade. The price to pay is that the qubits are no longer independent, as the laser beams overlap significantly with more than one qubit site. This problem is addressed by controlling the phase and amplitudes of the light at different locations, using structured light.

As a first application we show how to prepare a C-PHASE gate in two-and three-atomic systems, defining the Symmetric Orthogonal protocol (SOP), explained in the figure (left). The SOP extends the well-known Jaksch protocol<sup>3</sup> to non-independent qubits, and can operate in the nanosecond regime. Using hybrid modes of light (superpositions of TEM<sub>00</sub> and TEM<sub>01</sub>) with orthogonal amplitudes in the qubits, the population is driven through a dark state. Protocols that maximize the gate fidelity form a lattice in the space of solutions. The rotation of the lattice depends on the proximity of the qubits, observed in frames (a) to (d), with  $b=0$  for separated qubits and  $b=p/4$  for close qubits (or equal amplitudes of the field). Our results suggest that full spatio-temporal control of light can generate robust and efficient protocols for entangling gates in two-and three-dimensional arrays of closely spaced atoms.



**Figure 1.** The gate mechanism relies on a 3-pulse symmetric sequence using orthogonal hybrid modes of light that act on the computational basis of the qubits  $|00\rangle$ ,  $|01\rangle$ ,  $|10\rangle$ ,  $|11\rangle$  by exciting Rydberg states, as shown in the energy diagrams. Maximal fidelities show up at certain pulse areas ( $A_{\text{odd}} = A_1 + A_3$ ,  $A_{\text{even}} = A_2$ ) forming a lattice of solutions, that is rotated depending on the proximity of the atoms, measured as a function of parameter  $b$ . With  $b = 0$  the atoms are separated beyond the beam waist, for  $b^2 = 0.5$  they have maximum overlap. From (a) to (d) we use  $b^2 = 0, 0.1, 0.2, 0.5$  respectively.

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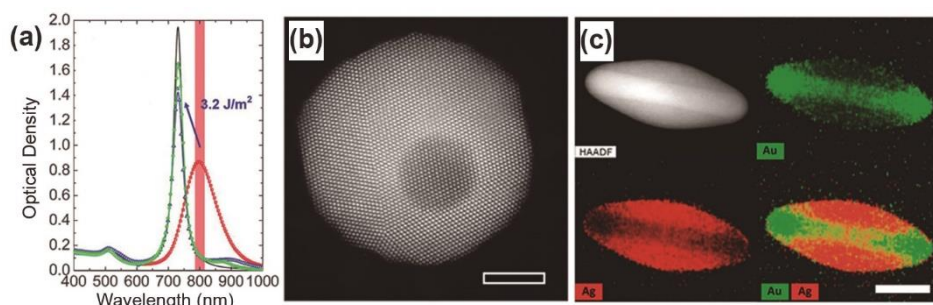
# IL 8-Tailoring gold nanocrystals with ultrafast laser irradiation

G. González-Rubio

Physical Chemistry, Department of Chemistry, University of Konstanz, Universitätsstraße 10, D-78465 Konstanz, Germany.

E-mail presenting author: [guillermo.gonzalez-rubio@uni-konstanz.de](mailto:guillermo.gonzalez-rubio@uni-konstanz.de)

Gold nanocrystals (NCs) are among the materials that have drawn the most attention in nanoscience due to their strong plasmonic properties, high chemical stability, and low toxicity. Moreover, the advancement of wet-chemistry methods has paved the way for the synthesis of gold NC with distinct morphologies and dimensions, critical aspects determining their optical performance.<sup>1</sup> In this regard, ultrafast pulsed laser irradiation has emerged as a valuable tool to prepare gold NC with distinctive morphological and plasmonic features. The absorption of laser pulse energy by gold NCs can lead to high lattice temperatures, resulting in melting, evaporation, and even fragmentation of the irradiated NCs.<sup>2</sup> Notably, the rate of heat release to the surrounding critically determines the fate of gold NC after ultrafast laser excitation. In practice, it implies that it is possible to govern the melting and reshaping of the irradiated NCs by controlling the interplay between the heating and cooling rates. We have illustrated this idea for the case of colloidal gold nanorods (NRs), where femtosecond pulsed laser irradiation was utilized to induce a controlled reshaping that resulted in ultranarrow plasmonic bands, near the theoretical limit (Figure 1a).<sup>3</sup> The required delicate balance between heating and cooling processes was achieved via the irradiation fluence ( $3.2 \text{ J}\cdot\text{m}^{-2}$ ) and the amount of surfactant molecules located on the NR surface, where they act as thermal insulators, providing control over the time the NR remains hot. In this sense, because the lattice cools down very fast, between a few tens of ps to a few ns, gold NCs excited with ultrafast pulse laser can evolve into metastable structures. A prominent case is that of hollow gold nanostructures obtained via nanosecond pulsed laser irradiation ( $200 \text{ J}\cdot\text{m}^{-2}$ ) of spherical gold NCs (Figure 1b).<sup>4</sup> In this work, we found that cavity creation involved trapping organic matter, probably surface ligands, from the surrounding. This fact pointed to the formation of intermediate structures with a porous nature where surrounding matter is entrapped during the fast cooling and recrystallization processes. Complex metal NCs can also be obtained through irradiation of plasmonic core@shell gold@silver NRs.<sup>5</sup> In this case, we demonstrate that the extreme temperatures reached by plasmonic gold@silver NRs upon ultrafast laser excitation can be utilized to fabricate NCs with different degrees of alloying and shape modifications. For instance, NRs with hot-dog-and rice-like morphology were obtained at low ( $3.2 \text{ J}\cdot\text{m}^{-2}$ ) and intermediate ( $6.4 \text{ J}\cdot\text{m}^{-2}$ ) pulse fluences, respectively (Figure 1c). Moreover, partial alloying was noticed at the intermediate pulse fluences, while completely alloyed spherical NCs formed at higher pulse energies ( $92 \text{ J}\cdot\text{m}^{-2}$ ). All these results will be presented in detail, aiming to reveal the potential of ultrafast laser irradiation as a unique tool for fabricating unique plasmonic nanomaterials.



**Figure 1.** (a) Femtosecond pulsed laser irradiation (red vertical band,  $3.2 \text{ J}\cdot\text{m}^{-2}$ ) of gold NRs (red) yields ultranarrow plasmonic bands (blue and green) that nearly match the calculated for a single NR (black). The symbols and lines represent the experimental and fitted optical density spectra, respectively. (b) HAADF-STEM image of a hollow gold NC obtained via nanosecond laser excitation of gold nanospheres ( $200 \text{ J}\cdot\text{m}^{-2}$ ). (c) HAADF-STEM images (top left) and quantified EDX maps (top right and bottom) of rice-like partially alloyed gold@silver NRs obtained via irradiation of gold@silver NRs with  $6.4 \text{ J}\cdot\text{m}^{-2}$  femtosecond-laser pulses. Scale bars: 4 nm (b) and 50 nm (c). Modified from Ref [3] (a), [4] (b), and [5] (c).

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## IL 9-The effect of bulky substituents on the relaxation dynamics of tetraphenyl ethylene derivative

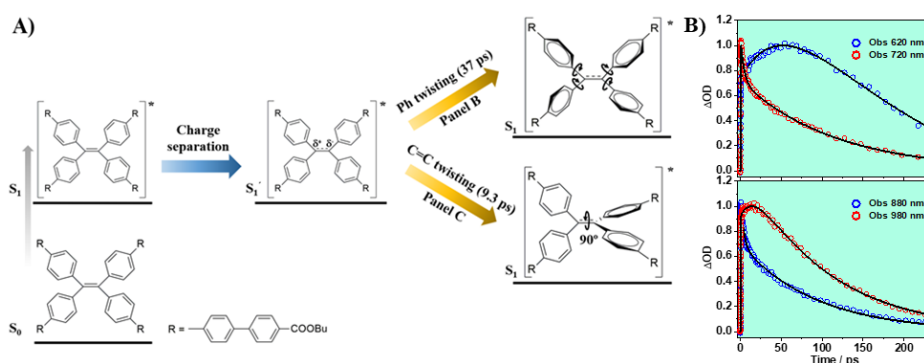
B. Cohen,<sup>a</sup> M. de la Hoz Tomás,<sup>a</sup> M. Yamaguchi,<sup>b</sup> I. Hisaki,<sup>b</sup> A. Douhal<sup>\*a</sup>

<sup>a</sup> Departamento de Química Física, Facultad de Ciencias Ambientales y Bioquímica, and INAMOL, Universidad de Castilla-La Mancha, Avenida Carlos III, S/N, 45071 Toledo, Spain.

<sup>b</sup> Graduate School of Engineering Science, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan.

E-mail presenting author: [boyko.koen@uclm.es](mailto:boyko.koen@uclm.es)

The photophysics and photochemistry of tetraphenylethylene (TPE) and its derivatives, including the role of rotation of the phenyl groups, ethylene bond twisting and photocyclization reaction, have been extensively studied since the end of the last century.<sup>1-5</sup> Like other molecules with ethylene bond the isomerization process takes place on an ultrafast timescale in addition to an efficient photochemical cyclization.<sup>2</sup> However, the underlying relaxation mechanisms of the excited TPE remain a topic of ongoing discussion, while the effect of bulky substituents is still under scrutiny.<sup>5</sup> Here, we report on the photophysical and photochemical properties of a new TPE derivative with two phenyl groups (TTECOOBu) in different solvents with different viscosity (Figure 1A). Fs-ps time-resolved measurements demonstrate that TTECOOBu undergoes an ultrafast charge redistribution along the ethylenic bond within 600 fs leading to a resonance structure with a lifetime of ~1 ns in DCM and DMF solutions. This structure may undergo a relaxation process via twisting motion of ethylene bond (~9 ps) and rotation of the phenyl rings (30-950 ps) leading to conformationally relaxed species with a largely Stokes-shifted emission. The latter motion clearly depends on the viscosity of the environment. A reversible twisting motion of the phenyl rings was also observed in DCM and DMF solutions but not in triacetin (Figure 1B). The fluorescence lifetimes of the conformationally twisted and relaxed species are ~70 ps and ~90 ps for DCM and DMF, respectively. These results provide new perspective into the relaxation mechanisms of TPE derivatives and will help in the development of new molecular rotors with aggregation-induced emission properties.



**Figure 1.** Proposed relaxation mechanism (A) and representative decays of the fs-transient absorption in the visible and near infrared spectral region (B) of the excited TTECOOBu.

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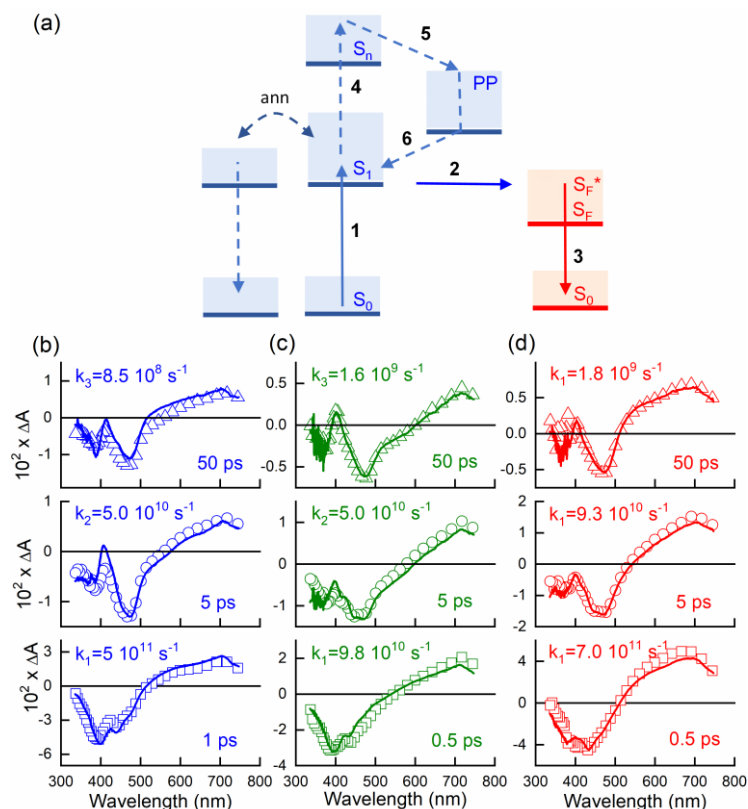
# IL 10-Boosting the stimulated emission properties of host:guest conjugated polymer blends by disruption of exciton transport

J. Cabanillas-Gonzalez

IMDEA Nanociencia, Calle Faraday 9, Ciudad Universitaria de Cantoblanco, Madrid 28049, Spain.

E-mail presenting author: [juan.cabanillas@imdea.org](mailto:juan.cabanillas@imdea.org)

Conjugated polymer blends coupled by Förster resonance energy transfer (FRET) have been widely exploited to achieve optically-pumped lasers operating at very low pumping thresholds. Among the plaid of conjugated polymers and molecules exploited for optical gain, fluorene-based polymers are considered front-runners, based on their high photoluminescence quantum yields, large optical gain coefficients and their processability assets in films of high optical quality. Two archetypes of polymers with these properties are poly(9,9-dioctyl-fluorene) (PFO) and its green-emitting relative Poly(9,9-dioctylfluorene-alt-benzothiadiazole) (F8BT) which work as excellent FRET-coupled blends but unexpectedly do not exhibit stimulated emission. In this talk we will shed light into the optical gain limiting factors of these blends. Upon investigating a series of polyfluorenes bearing different chain length<sup>1-3</sup> or chain twists,<sup>4</sup> we reveal improvements on the stimulated emission properties by harnessing exciton annihilation. We ascribe these effects to disruption of exciton transport, reducing the yield of charge-transfer states and associated photoinduced absorption losses.



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# IL 11-Measurements of the long-term evolution and stability of the 45TW-STELA laser operating at 10Hz

A. Alejo,<sup>a</sup> A. Bembibre,<sup>a</sup> J. Peñas,<sup>a</sup> J. Benlliure,<sup>a</sup> L. Martín<sup>b</sup> and M. T. Flores-Arias<sup>b,c</sup>

<sup>a</sup> Instituto Galego de Física de Altas Enerxías (IGFAE). Universidade de Santiago de Compostela, Spain.

<sup>b</sup> Laboratorio Láser de Aceleración y Aplicaciones (L2A2). Universidade de Santiago de Compostela, Spain.

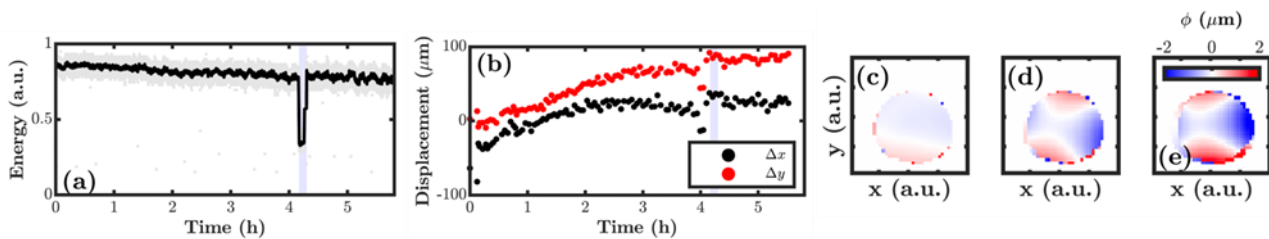
<sup>c</sup> Instituto de Materiales (iMATUS). Universidade de Santiago de Compostela, Spain.

E-mail presenting author: [aaron.alejo@usc.es](mailto:aaron.alejo@usc.es)

The continuous developments in laser technology over the last decades has led to a broad range of applications in science and technology. In particular, the advent of commercial laser systems capable of producing intensities in excess of  $10^{18}$  W/cm<sup>2</sup> have increased the interest in laser-driven particle accelerators. These accelerators, based on the interaction of the laser with a plasma, benefit from the huge acceleration gradients sustainable by the plasma, more than three orders of magnitude greater than the fields achieved in conventional accelerators. Recent results include the acceleration of electron beams to energies up to 7.8 GeV,<sup>1</sup> or the acceleration of proton beams to energies up to 100 MeV.<sup>2</sup> Following these impressive results highlight the maturity of laser-plasma accelerators, allowing for their widespread use as alternative to conventional accelerators, with ongoing efforts such as the EuPRAXIA project.<sup>3</sup>

In this context, the Laser Laboratory for Acceleration and other Applications facility (L2A2) was deployed at the Universidade de Santiago de Compostela, which hosts the STELA (Santiago TeraWatt Laser) system, a fs-class Ti:Sapphire laser providing a peak power of 45 TW and operating at a repetition rate of up to 10 Hz, to study applications of laser-driven radiation sources, such as X-rays or ion beams. In addition to work on the optimisation of the laser-plasma interaction and developments on targetry systems that can operate at multi-Hertz repetition rate, significant efforts are being directed to ensure a stable laser operation that can ensure the radiation sources can remain stable for extended periods of time. Several effects can affect the evolution of a laser system, including changes in humidity or temperature, the long term evolution of the laser pumps, or the heating of optical elements along the beamline, particularly the heating of the compressor gratings.<sup>4</sup>

Here, we present recent progress on the operation of the STELA laser for ion acceleration, including the analysis of the identification of new elements to be installed that allow for safe operation of the laser system. Furthermore, we present measurements of the evolution of the main laser parameters of the system operating at 10 Hz, showing that a significant change in the wavefront and direction of propagation, even when the laser energy remains sufficiently constant. These results highlight the importance for future laser systems to integrate closed-loop beam tracking diagnostics that can correct the temporal evolution.



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# IL 12-Controlling silicon phase-change transformations by ultrashort laser pulses

M. Garcia-Lechuga,<sup>a,b</sup> N. Casquero,<sup>a</sup> J. Solis,<sup>a</sup> I. Solana,<sup>b</sup> M. D. Ynsa,<sup>b,c</sup> A. Wang,<sup>d</sup> D. Grojo<sup>d</sup> and J. Siegel<sup>a</sup>

<sup>a</sup> Instituto de Óptica Daza de Valdés (IO), CSIC, 28006 Madrid, Spain.

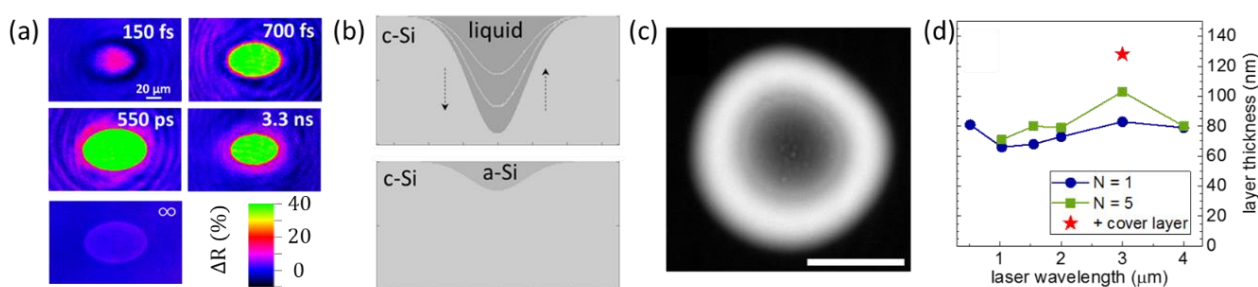
<sup>b</sup> Centro de Microanálisis de Materiales (CMAM), Universidad Autónoma de Madrid, 28049 Madrid, Spain.

<sup>c</sup> Departamento de Física Aplicada, Universidad Autónoma de Madrid, 28049 Madrid, Spain.

<sup>d</sup> Aix-Marseille Université, CNRS, LP3, UMR7341, 13009 Marseille, France.

E-mail presenting author: [mario.garcia.lechuga@csic.es](mailto:mario.garcia.lechuga@csic.es)

Direct laser writing with ultrashort pulses on crystalline silicon is a well-known methodology for creating thin amorphous surface layers. The appeal of this method for applications in the field of silicon photonics lies in the possibility of fabricating low-loss optical guiding structures, taking advantage of the high refractive index and low loss of the amorphous phase at telecommunication wavelengths. However, the reported maximum layer thickness to date (70 nm) is insufficient for supporting the propagation of guided modes. Therefore, our research is focused on understanding and expanding those limitations, exploring both the physics of laser-matter interaction and material engineering. Firstly, we report the characteristic times scales of the underlying mechanisms by using ultrafast time-resolved microscopy.<sup>1</sup> Visualizing the reflectivity evolution of the surface after irradiation reveals the formation of a dense free electron plasma (Fig. (a), delay 150 fs), followed by non-thermal melting (Fig. (a), delay 700 fs) and eventually fast quenching of the molten phase (Fig. (a), delays 550 ps and 3.3 ns). Melting and solidification processes lead to the formation of a permanent thin amorphous layer, as sketched in Fig. (b). Even though the mechanisms are revealed, their complex dynamics and depth dependence makes it difficult to theoretically predict the optimal conditions for amorphization of a thicker layer. Secondly, we present experimental strategies aimed at increasing the layer thickness beyond today's limit. Exploring novel excitation strategies, we present detailed experimental studies of the role of laser wavelength,<sup>2</sup> ranging from 258 nm to 4000 nm. Of particular interest is the underexplored mid-infrared region of the silicon transparency regime ( $\lambda > 1400$  nm), providing access to a drastic change of the nonlinear ionization response. Our results show that with optimized parameters (multipulse and infrared radiation) amorphous layers with a maximum thickness significantly exceeding the mentioned 70 nm-limit (c. f. Fig. (c, d)) can be fabricated. Finally, we have performed experiments on preconditioned silicon samples. We show that silicon samples with a SiO<sub>2</sub> cover layer are beneficial for thick amorphization due to a faster cooling of molten material. Moreover, we report the synergistic effect of MeV ion implantation prior to fs-laser irradiation, which enables a greater tunability of the obtained thickness as well as a good control of the induced phase-changes: from c-Si to a-Si but also from a-Si to c-Si.



**Figure 1.** (a) Time-resolved images (illumination 400 nm) of single-pulse excitation (800 nm, 120 fs) of silicon. The reflectivity change  $\Delta R$  is encoded in false-color, where green colors correspond to the reflectivity of molten silicon. (b) Sketch of the transformation dynamics of melting and resolidification after irradiation (top) and permanent formation of amorphous silicon (bottom). (c) Optical microscopy image of an amorphous spot in crystalline silicon, formed upon single pulse exposure (200 fs) at  $\lambda = 3000$  nm. Reflectivity changes (bright and dark regions) are a consequence of an interference pattern due to the formation of an amorphous silicon layer. The experimental reflectivity analysis indicates a thickness of 80 nm (lateral scale bar 10  $\mu\text{m}$ ). (d) Maximum thickness values of the laser induced amorphous layer as a function of laser wavelength, for single pulse (N = 1) and multiple pulse (N = 5) irradiation. The red symbol marks the maximum thickness achieved by using the presence of a SiO<sub>2</sub> cover layer.

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# IL 13-Nanomaterials fabricated by pulsed laser synthesis in liquids

C. Doñate-Buendía,<sup>a,b</sup> S. Molina-Prados,<sup>a</sup> F. R. Cortes,<sup>a</sup> J. Lancis<sup>a</sup> and G. Mínguez-Vega<sup>\*a</sup>

<sup>a</sup> GROC-UJI, Institut de Noves Tecnologies de la Imatge (INIT), Universitat Jaume I, Spain.

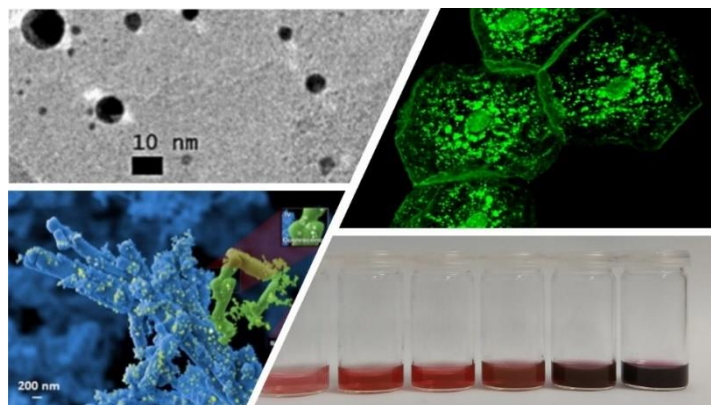
<sup>b</sup> Materials Science and Additive Manufacturing, School of Mechanical Engineering and Safety Engineering, University of Wuppertal, Germany.

E-mail presenting author: [gminguez@uji.es](mailto:gminguez@uji.es)

Conventional synthesis methods as wet chemical synthesis or mechanical milling are well suited for nanoparticle mass production. However, they are negatively affected by contamination, background noise in analytical chemistry, and time consuming purification methods. The bottleneck incorporation of nanotechnology in our lives and its link to industrial progress incites the development of novel nanoparticle synthesis techniques, that may provide different physicochemical properties as the characteristics of the nanoparticles depend on the production method. In this direction, fabrication of nanomaterials by pulsed laser synthesis (PLS) in liquids provides key advantages as simplicity of the experimental setup, broad library of available materials covering nearly the entire periodic table, and, overall, pureness of the nanomaterials, since only the solvent and desired material are required, avoiding the presence and generation of toxic or hazardous adducts and byproduct.

In this talk, we will review the fundamentals of PLS in liquids, focusing on the pioneering work applying a disruptive technique, Simultaneous Spatial and Temporal Focusing (SSTF) to femtosecond (fs) PLS.<sup>1</sup> In fs PLS the nanoparticle production is limited, in comparison with longer pulses, by the nonlinear effects in the liquid. However, SSTF-PLS is a unique way to maximize energy delivery to the target, thanks to the fact that the temporal pulse width becomes a function of the propagation distance, with the shortest pulse duration confined to the spatial focus, avoiding the nonlinear losses in the liquid. This fact increases nanoparticle production by a factor of 2.4 compared to the conventional fs PLS system.

Further applications of the nanomaterials developed in our group will be discussed during the talk. Specifically, fluorescent carbon dots for bioimaging,<sup>2</sup> decorated graphene oxide with metal nanoparticles,<sup>3</sup> chitosan/ $\alpha$ -Ag<sub>2</sub>WO<sub>4</sub> composites for the inactivation of SARS-CoV-2,<sup>4</sup> or solar nanofluids.<sup>5</sup>



**Figure 1.** Example of several nanomaterials generated by PLS in liquids in our group: a) gold nanoparticles anchored in graphene oxide b) fluorescence carbon dots internalized in oral epithelial cells c) silver (in green) nanoparticles formed by fs irradiation of Ag<sub>2</sub>WO<sub>4</sub> and d) gold colloids synthesized by fs SSTF-PLS system with different fluences.

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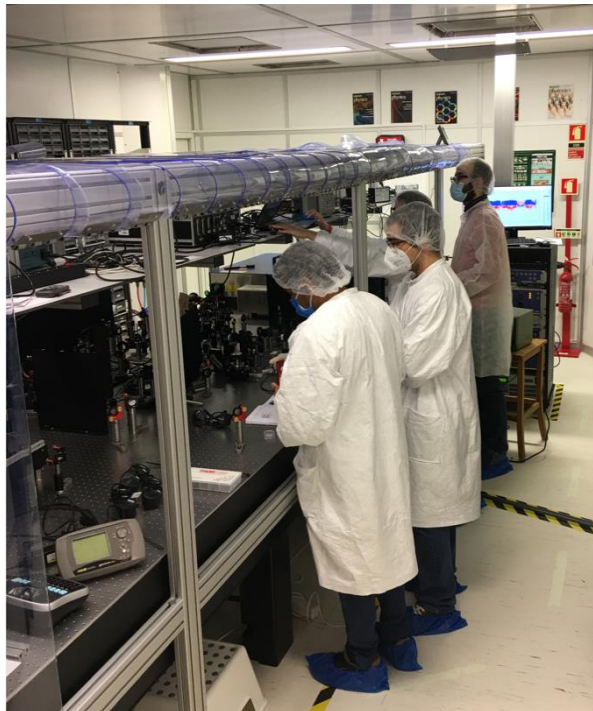
# IL 14-Ultrafast laser research in Portugal: overview

G. Figueira

GoLP/IPFN, Instituto Superior Técnico, Av. Rovisco Pais, Lisbon 1049-001, Portugal.

E-mail presenting author: [goncalo.figueira@tecnico.ulisboa.pt](mailto:goncalo.figueira@tecnico.ulisboa.pt)

In this talk I will provide an overview of the panorama of ultrafast laser research in Portuguese research institutions, including laser development and applications. In the last 25 years the country has developed a small but active community working in a range of topics, concentrated at the main universities. In the last decade, the main distributed laser infrastructure-Laserlab-Portugal, bringing together facilities at the universities of Lisbon and Coimbra-was selected for the national roadmap of top research infrastructures, providing access to national researchers. The teams are also active in international networks such as Laserlab-Europe and have strong links to large-scale facilities such as ELI.



**Figure 1.** Laboratory for Intense Lasers (L2I), IST, Lisbon.

# IL 15-Soft X-ray femtosecond time-resolved photoelectron spectroscopy opportunities at the European XFEL

M. Izquierdo

European XFEL, Holzkoppel 4, 22869 Hamburg, Germany.

E-mail Presenting Author: [manuel.izquierdo@xfel.eu](mailto:manuel.izquierdo@xfel.eu)

The successful implementation of the baseline instruments at the European XFEL has motivated the development of a new program to extend the portfolio of experimental techniques. In the soft X-ray undulator system, SASE3 a third instrument, the soft X-ray Port (SXP), is currently under construction. The first experimental technique to be implemented is femtosecond time-resolved photoelectron spectroscopy (TR-XPES) on solids. This technique, successfully implemented using laser sources, has proven to be a very powerful tool for the understanding and coherent control of materials. However, the experimental requirements limit the accessible photon energy range to around 100 eV. The extension of the technique to higher photon energies is only possible at MHz free electron lasers (FELs). In this contribution, the SXP instrument at the European XFEL and the implementation of TR-XPES using a momentum microscope will be presented (1). The photon energy range available at the SASE3 undulator system, 0.25 to 3 keV, and the variable polarization will allow a simultaneous characterization of the electronic, magnetic, and structural properties of materials with femtosecond resolution. To this end, a wide range of laser wavelengths, from the XUV to the THz region, will be available.

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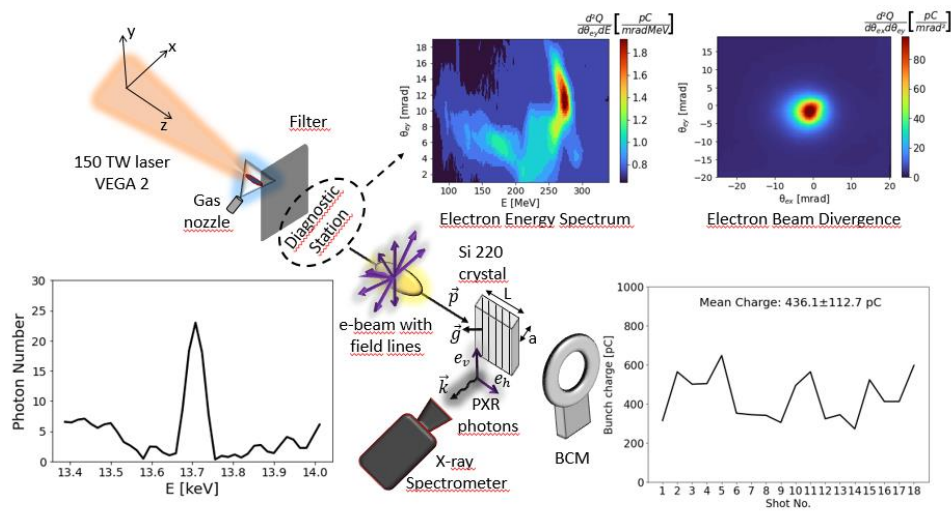
# IL 16-Observation of tunable parametric X-ray radiation emitted by laser-plasma electron beams interacting with crystalline structures

A. Curcio,<sup>a</sup> M. Ehret,<sup>a</sup> J. A. Perez<sup>a</sup> and G. Gatti<sup>a</sup>

<sup>a</sup> Centro de Láseres Pulsados (CLPU), Edificio M5. Parque Científico. C/ Adaja, 8. 37185 Villamayor, Salamanca, Spain.

E-mail presenting author: [acurcio@clpu.es](mailto:acurcio@clpu.es)

Parametric X-ray Radiation (PXR) is the quantum mechanism analogue to Laue diffraction where a pseudophoton carried by an electron is scattered out of a crystalline structure as a radiation photon. In this work PXR emitted by electron beams generated with a compact laser-plasma accelerator and interacting afterwards with a Si 220 crystal is observed in single-shot operations. The combination of laser wakefield acceleration and PXR is an efficient and table-top way to obtain monochromatic, pulsed, ultrashort, and stable emission of high energy photons. Unlike other relevant radiation mechanisms, such as betatron radiation or incoherent bremsstrahlung, PXR is insensitive to the beam energy spread. With a long crystal, we demonstrate the stability of the PXR bandwidth toward shot-to-shot variations of the beam energy and divergence.



**Figure 1.** Experimental setup for PXR emission by laser-produced electron beams at VEGA 2.

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# ORAL COMMUNICATIONS

# OC 1-Wavelength dependence of the multiphoton ionization of CH<sub>3</sub>I by intense femtosecond laser pulses through Freeman resonances

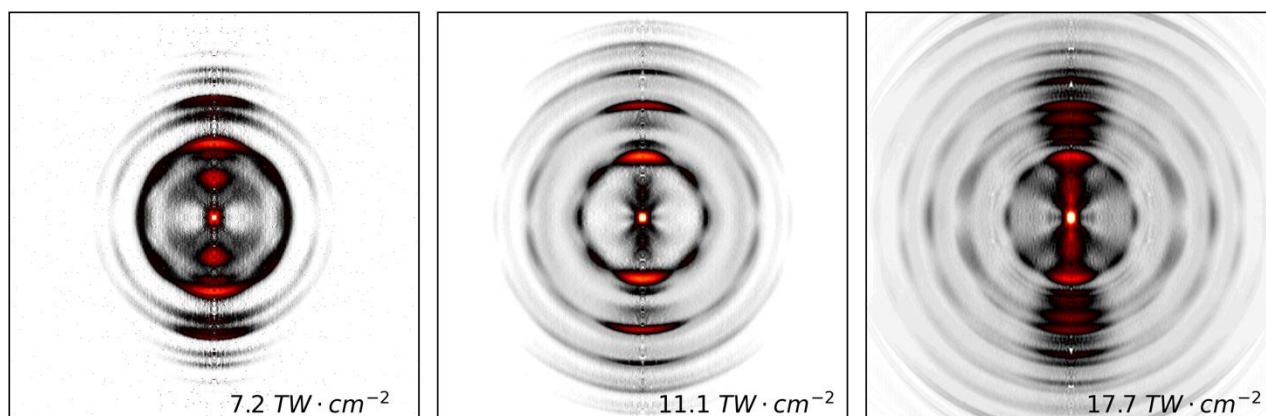
L. Bañares,<sup>\*a,b</sup> I. M. Casasús<sup>a</sup> and M. E. Corrales<sup>a</sup>

<sup>a</sup> Departamento de Química Física, Facultad de Ciencias Químicas, Universidad Complutense de Madrid, 28040 Madrid, Spain.

<sup>b</sup> Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA Nanoscience). Campus de Cantoblanco, 28049 Madrid, Spain.

E-mail presenting author: [lbanares@ucm.es](mailto:lbanares@ucm.es)

Multiphoton ionization (MPI) of methyl iodide, CH<sub>3</sub>I, has been investigated with the photoelectron imaging (PEI) technique, using high intensity femtosecond laser pulses at different central wavelengths. The use of high laser field strengths alters the way in which matter-radiation interaction takes place. This generates highly nonlinear phenomena, among which we can highlight the Stark shift effect. It can distort the potential energy surfaces of atoms and molecules, varying both the energy of electronic and rovibrational states of these systems and their ionization potentials. In this way, the Stark shift can generate resonances between intermediate states and an integer number of laser photons of a given wavelength, which would be absent in the low intensity regime. The main purpose of this work is the generation, detection and characterization of resonances produced by the Stark shift, commonly known as Freeman resonances, induced by multiphoton ionization of gas-phase CH<sub>3</sub>I at different laser wavelengths. The results obtained reveal that a multitude of resonances are induced in the ionization of CH<sub>3</sub>I in the range of intensities employed, involving several Rydberg states. Ionization pathways associated with different degrees of vibrational excitation in both the intermediate states and the molecular cation generated in each of the experiments are proposed.



**Figure 1.** Selected Abel-inverted PE images obtained by photoionization of CH<sub>3</sub>I with femtosecond pulses centered at 800 nm. Laser intensities at the intercept with the molecular beam are indicated in the right-bottom corner of each image. The linear polarization of the laser beam is aligned vertically with respect to the image plane.

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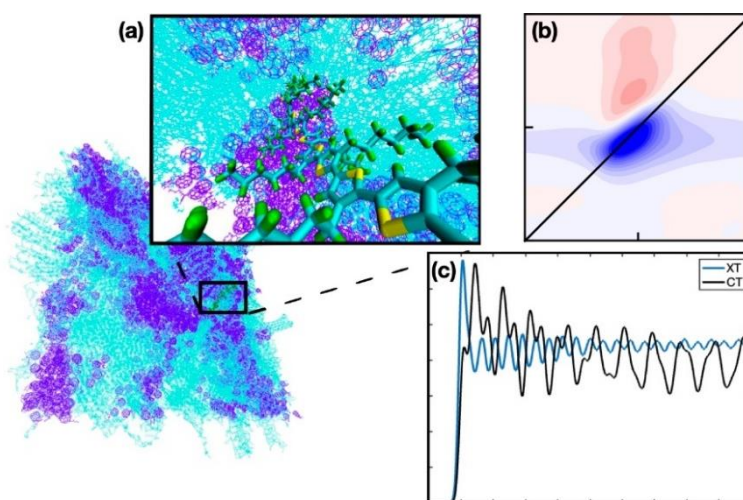
# OC 2-Modelling ultrafast charge-transfer in organic photovoltaic materials: the role of quantum vibrations, environment disorder and light pulse shape

E. Palacino-González<sup>\*a</sup> and T. la Cour Jansen<sup>a</sup>

<sup>a</sup> Zernike Institute for Advanced Materials, University of Groningen, Groningen, The Netherlands.

E-mail presenting author: [e.palacino.gonzalez@rug.nl](mailto:e.palacino.gonzalez@rug.nl)

We introduce a first-principles modelling of the ultrafast charge separation dynamics and spectroscopy at the donor-acceptor heterojunction of the P3HT:PCBM organic photovoltaic (OPV) blend.<sup>1</sup> Our approach combines Molecular Dynamics (MD) simulations to describe the blend environment<sup>2</sup> with TDDFT calculations to construct a Merrifield-type Hamiltonian describing the heterojunction after photoexcitation. The time-resolved spectroscopic observables are then evaluated using the response function-based NISE package<sup>3</sup> and compared with available experimental measurements to further validate the model and understand the underlying dynamics. The predicted spectra nicely reproduce the available experimental data.<sup>1</sup> The origin of the line broadening is analysed in terms of the conformational disorder. This reveals for instance the importance of the torsional flexibility for the different regions of the donor P3HT polymer chains, allowing an identification of the homogeneous and inhomogeneous spectral broadening.<sup>4</sup> Subsequently, the light-induced dynamics of the charge separation in the photovoltaic blend are simulated quantum mechanically with an explicit treatment of the photoexcitation pulses and vibronic effects from specific intramolecular modes. Dissipation rates are computed from the spectral densities derived from the MD trajectory, which allows a realistic description interaction of the bath blend environment with the system. Our study delivers a complete first-principles picture of the electronic and vibrational coherence dynamics characterizing ultrafast charge-transfer in prototypical OPVs, emphasizing the role of the dominant vibrational modes controlling the process, the effect of disorder and the explicit description of different excitation conditions in the laser fields.



**Figure 1.** (a) Thin film of a P3HT:PCBM blend with close up at the heterojunction (b) Simulated 2D spectra of the P3HT system in the blend (c) Population quantum dynamics of the bright excitonic state (blue) and the dark charge-transfer state (black) after exciting the heterojunction with a short pulse.

**Acknowledgments:** This work is supported by the Marie Skłodowska-Curie actions MSCA-IF-2020-Individual Fellowships (Horizon 2020 framework): <https://cordis.europa.eu/project/id/101027783>

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# OC 3-Symmetry-protected chiral sensitivity in photoionization with bichromatic fields

A. Ordóñez,<sup>\*a,b</sup> D. Ayuso<sup>b,c</sup> P. Decleva,<sup>d</sup> and O. Smirnova<sup>b,e</sup>

<sup>a</sup> ICFO-Institut de Ciències Fòniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels (Barcelona), Spain.

<sup>b</sup> Max-Born-Institut, 12489 Berlin, Germany.

<sup>c</sup> Imperial College London, SW7 2AZ London, United Kingdom.

<sup>d</sup> Università degli Studi di Trieste, 134127 Trieste, Italy.

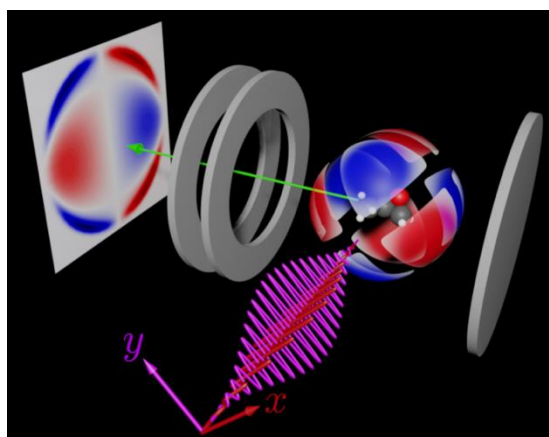
<sup>e</sup> Technische Universität Berlin, Berlin, 10623, Germany.

E-mail presenting author: [andres.ordonez@icfo.eu](mailto:andres.ordonez@icfo.eu)

The fact that molecular chirality can be very efficiently recorded using table-top femtosecond lasers has spurred a flurry of activity in the ultrafast science community, partly driven by the immense potential of ultrafast chiral techniques for industrial applications. A property common to all ultrafast chiral techniques is that reversing the ellipticity of the driving field results in a reversal of the enantio-sensitive signal. Indeed, recent simulations and experiments<sup>1,2</sup> have shown that even in bichromatic fields of the form  $E = E_x \cos(\omega t)x + E_y \cos(2\omega t + \varphi)y$  tracing 8-like Lissajous figures, the enantio-sensitive part of the photoelectron angular distribution (PAD) is reversed upon reversal of their instantaneous ellipticity, i. e. upon a change of  $\pi$  in the relative phase  $\varphi$  between the  $x$  and  $y$  components.

Here we have found that, contrary to intuition, bichromatic fields also lead to an enantio-sensitive contribution to the PAD which does not change sign upon reversal of the instantaneous ellipticity. Moreover, this Enantio-sensitive and Non-Elliptically-Dichroic (ENED) signal can be entirely independent of the relative phase  $\varphi$  and thus provides an enantio-sensitive signal robust with respect to variations of  $\varphi$ , a very desirable feature for industrial applications.

This robustness stems from the symmetry properties of the Hamiltonian of the interacting system. Our analysis<sup>3</sup> reveals that these symmetries, constraining which tensors contribute to the PAD, allow for the emergence of a subset of ENED tensors, in particular for direct ionization pathways. The simplest scenario to obtain an ENED signal requires consecutive absorption of one photon of each color and a 3D detection setup such as COLTRIMS or a VMI setup with the measurement geometry shown below (previous works have used a different geometry for which the ENED signal is averaged out). We derived analytical expressions for this case<sup>3</sup> and performed numerical computations in methyloxirane that reveal an ENED signal corresponding to 1% of the total cross section, i. e. within current experimental capabilities.<sup>4</sup>



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# OC 4-Giant photoactuated effects on freestanding ferroelectric oxide membranes

S. Ganguly,<sup>a,\*</sup> D. Pesquera,<sup>a,\*</sup> D. Moreno Garcia,<sup>c</sup> U. Saeed,<sup>a</sup> N. Mirzamohammadi,<sup>a</sup> J. M. Caicedo Roque,<sup>a</sup> J. Santiso,<sup>a</sup> C. Laulhe,<sup>d,e</sup> L. Guillermo Villanueva<sup>c</sup> and G. Catalan.<sup>a,b,\*</sup>

<sup>a</sup> Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, Barcelona, 08193 Spain.

<sup>b</sup> ICREA-Institució Catalana de Recerca i Estudis Avançats, Catalonia, Barcelona, 08010 Spain.

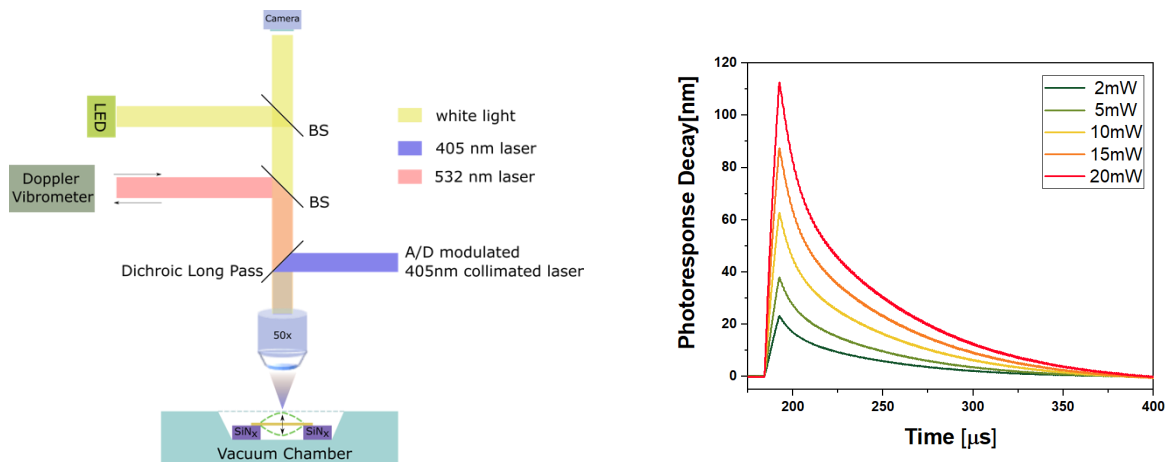
<sup>c</sup> Advanced NEMS Laboratory, Institute of Mechanical Engineering, École Polytechnique Fédérale de Lausanne (EPFL), 1015, Lausanne, Switzerland.

<sup>d</sup> Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin BP 48, 91192 Gif-sur-Yvette Cedex, France.

<sup>e</sup> Université Paris-Saclay (Université Paris-Sud), F-91405 Orsay Cedex, France.

E-mail presenting author: [saptam.ganguly@icn2.cat](mailto:saptam.ganguly@icn2.cat)

Advent of an epitaxy compatible water-soluble oxide [ $\text{Sr}_3\text{Al}_2\text{O}_6$  (SAO)] has created a niche in Oxide physics research, which was primarily based on studying single crystals and epitaxial thin films.<sup>1,2</sup> Using SAO as a sacrificial layer, it is thus possible to fabricate free-standing complex oxide thin films. This enables us to study mechanical, electrical and magnetic properties of single-crystal thin films free from any mechanical boundary conditions such as substrate clamping. In this work, we have fabricated large area free-standing membranes of ferroelectric Barium Titanate [ $\text{BaTiO}_3$ (BTO)] and paraelectric Strontium Titanate [ $\text{SrTiO}_3$ (STO)], of various thicknesses (10-80 nm) and suspended on holes of different diameters (10-40  $\mu\text{m}$ ), all integrated on a Silicon based architecture. Using Doppler Vibrometry (as shown in Figure 1a), we observe giant photoinduced effects on the ferroelectric membranes upon illuminating with near-bandgap light. The temporal decay profile of such light induced deflection is depicted in Figure 1b. We investigate the origin of this phenomena using time resolved x-ray microdiffraction using synchrotron radiation. Finally, we characterize these membranes as resonators, where they stand out in terms of performance when compared to previously reported oxide membrane resonators of similar architecture. This work emphasises that the photoinduced strains in ferroelectrics can be exploited on freestanding membranes to create photoactuation systems with largely enhanced performances.



**Figure 1.** a. A schematic micrograph of the optical actuation and detection system. b. Giant photoinduced deflection and its decay profile as a function of laser peak power.

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# OC 5-Band-like charge transport in phytic acid-doped polyaniline thin films

M. Ballabio,<sup>a,g</sup> T. Zhang,<sup>b,c,d\*</sup> C. Chen,<sup>e</sup> P. Zhang,<sup>b</sup> Z. Liao,<sup>f</sup> M. Hamsch,<sup>g</sup> S. C. B. Mannsfeld,<sup>g</sup> E. Zschech,<sup>f</sup> H. Sirringhaus,<sup>e</sup> X. Feng,<sup>b\*</sup> M. Bonn,<sup>a\*</sup> R. Dong<sup>b\*</sup> and E. Cánovas<sup>a,h\*</sup>

<sup>a</sup> Max Planck Institute for Polymer Research, Mainz, Germany.

<sup>b</sup> Center for Advancing Electronics Dresden (CFAED) and Faculty of Chemistry and Food Chemistry Chair of Molecular Functional Materials, Technische Universität Dresden, Dresden, Germany.

<sup>c</sup> Key Laboratory of Marine Materials and Related Technologies Zhejiang Key Laboratory of Marine Materials and Protective Technologies, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo, China.

<sup>d</sup> School of Chemical Sciences, University of Chinese Academy of Science Beijing, China.

<sup>e</sup> Cavendish Laboratory, Cambridge, United Kingdom.

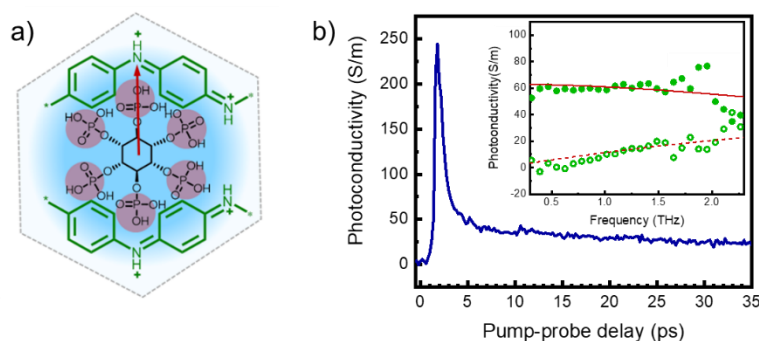
<sup>f</sup> Fraunhofer Institute for Ceramic Technologies and Systems (IKTS), Dresden, Germany.

<sup>g</sup> Center for Advancing Electronics Dresden (CFAED) and Faculty of Electrical and Computer Engineering, Technische Universität Dresden, Dresden, Germany.

<sup>h</sup> Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA Nanociencia), Madrid, Spain.

E-mail presenting author: [marco.ballabio@imdea.org](mailto:marco.ballabio@imdea.org)

We explore the charge transport properties of phytic acid (PA) doped polyaniline thin films prepared by the surfactant monolayer-assisted interfacial synthesis (SMAIS). Charge transport properties are interrogated by Time-Resolved Terahertz Spectroscopy (TRTS). The THz spectral region is primarily sensitive to free carrier motion and as such, it allows the conductivity to be probed in a contactless, fully optical manner. Furthermore, by using a pump-probe scheme (Optical Pump-Terahertz Probe, OPTP), THz spectroscopy can time-resolve the (photo)conductivity of a semiconducting system with sub-picosecond resolution. While other conventional pump-probe schemes record the time-integrated intensity of the probe pulse, in THz spectroscopy time-dependent electric field of the THz pulses is measured. This allows characterizing the complex-valued, frequency dependent conductivity of a system. By the means of OPTP, we observe that in PA-doped polyaniline, the frequency-resolved complex conductivity spectra in the THz region can be properly described by the Drude model, independently of doping content and crystallinity of the material. This demonstrates band-like charge transport in the samples, reaching state-of-the-art charge carrier mobilities of approximately  $1 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ . A temperature-dependent analysis for the conductivity further supports band-like charge transport and suggest that charge carrier mobility is primarily limited by impurity scattering.



**Figure 1.** a) Sketch of the molecular structure of PA-doped polyaniline. b) Time-resolved THz photoconductivity of PA-doped polyaniline excited with a 800nm, 50fs laser pulse. In the inset frequency-resolved photoconductivity at 10ps after photoexcitation showing Drude-like conduction.

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# OC 6-Transient absorption for organic electronics characterisation: Diradicals, solar cells and symmetry breaking

J. M. Marin-Beloqui<sup>\*a</sup> and J. Casado<sup>a</sup>

<sup>a</sup> Department of Physical Chemistry, University of Málaga, Blvd Louis Pasteur 31, 29010, Málaga, Spain.

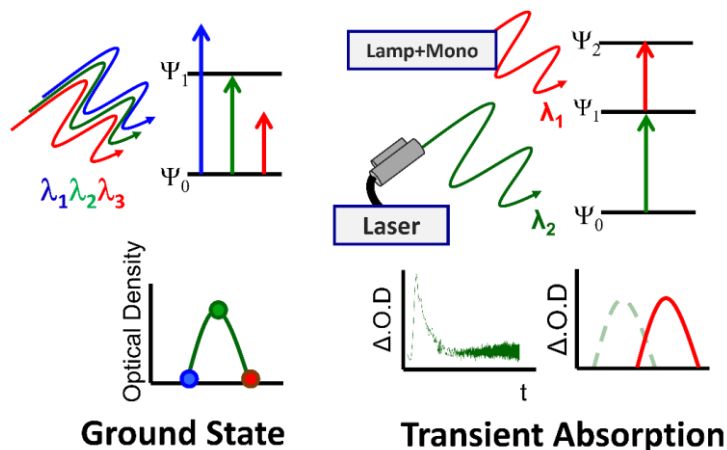
E-mail presenting author: [jm.marinbeloqui@uma.es](mailto:jm.marinbeloqui@uma.es)

Organic electronics have the capability to overcome conventional inorganic materials due to several advantages such as easier processability, potential for cheaper manufacture and a large tunability. However, their efficiency in terms charge formation, charge transport and lifespan are not at the same level than their inorganic counterparts. One of the main reasons is the large number of phenomena that take place in organic materials upon photon absorption that hinders that efficiency.

Transient absorption spectroscopy (TAS) represents the perfect technique to study this kind of processes. TAS is a two beam technique where one beam excites the sample while the other ones probes the excited species (like excitons, triplets and charges) and their evolution with time. With TAS it is possible to analyse the different reactions that take place upon excitation, and therefore, elucidate the origin of this lack of efficiency.

In this work, TAS has been used in the elucidation of the behaviour of several materials for organic electronics. The application of these materials go from their use in solar cells to their use as detectors in bioapplications. For instance, this work shows that TAS is able to make a relationship between the diradical character of Difluorenoheteroles and the exciton relaxation lifetime as well as the relaxation process. Moreover, it is possible to see the charge generation and recombination processes in different organic materials for OPV (like ZZ115). Finally, with the results obtained by TAS we were able to identify that the origin for the 2-Photon absorption phenomenon was a breaking symmetry process in the excited state.

This works demonstrate the capabilities and advantages of TAS for the study of photoexcited species in organic electronics.



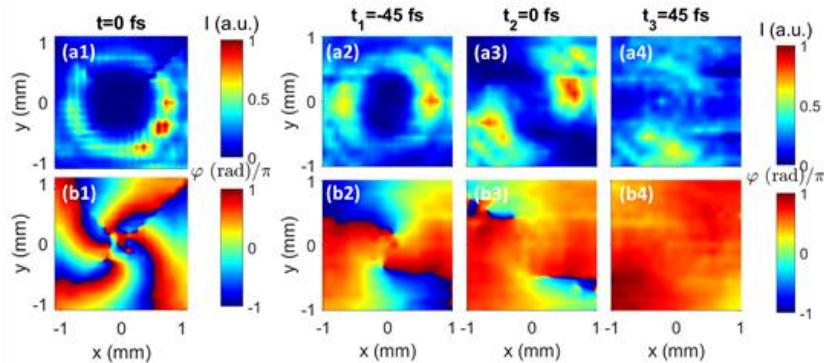
# OC 7-New ultrafast imaging technique applied to the retrieval of beams with time evolving optical angular momentum

Í. Sola,<sup>\*a</sup> M. López-Ripa<sup>a</sup> and B. Alonso<sup>a</sup>

<sup>a</sup> Grupo de Aplicaciones del Láser y Fotónica (ALF), Departamento de Física Aplicada, Universidad de Salamanca, Plaza de la Merced s/n, Salamanca, Spain.

E-mail presenting author: [ijisola@usal.es](mailto:ijisola@usal.es)

The use of ultrafast pulse beams is increasing along the years, rising their complexity. In particular, ultrafast pulsed beams carrying optical angular momentum (OAM), or optical vortices, are of great interest and a hot topic on several research fields (e. g., attoscience, microscopy, microprocessing, optical tweezers, etc. ). However, such kind of complex light structures involve a challenge for their proper characterisation. In this contribution, we present firstly a new technique for spatio-temporal reconstruction of ultrafast pulsed beams, consisting on a bulk in-line architecture using spectral and lateral interferometry.<sup>1</sup> In addition, the time reference measurement was performed using the amplitude swing technique,<sup>2</sup> also in a bulk set-up. Therefore, the set-up is very insensitive to perturbations and is able not only to measure the pulse fronts, but also to resolve temporally and spectrally the beam wavefront. With this tool, ultrafast pulsed beams carrying OAM exhibiting diverse complexity can be studied. The pulsed light source used in the experiments was a CPA Ti:Sapphire laser (central wavelength: 800 nm). The OAM was imprinted in the beam by means of the so called s-waveplates (i. e., nanostructured retarded waveplates exhibiting space dependent orientation) and it is possible to generate different kind of OAM structures. Thus, diverse cases have been generated at different wavelengths. Fig 1. a1 presents a snapshot at the pulse peak ( $t=0$  fs) of the intensity spatial profile of an optical vortex at a central wavelength of 400 nm (second harmonic of the laser beam) carrying a topological charge  $l=4$  constant on time, while its spatial phase is depicted at Fig1. b1. On the other hand, it is possible to generate and measure beams exhibiting time evolving OAM. As an example, two ultrafast pulsed beams with topological charge  $l_1=2$  and  $l_2=0$  were generated presenting a time delay of the order of the pulses time duration, around 60 fs. Therefore, in the front part of the pulse, at  $t_1=-45$  fs (Fig 1. a2 and Fig. b2 for intensity and phase respectively), the beam presents characteristics of a  $l_2=2$  vortex, while in the rear part, at  $t_3=45$  fs (Fig 1. a4, intensity, and Fig. b4, spatial phase), it is a beam without OAM ( $l_2=0$ ). Around the pulse peak, at  $t_2=0$  fs, both natures are present (Fig 1. a3, intensity, and Fig. b3, spatial phase).



**Figure 1.** Snapshots of spatiotemporal reconstruction of (a1, b1) a pulsed beam with topological charge  $l=4$  constant on time ( $t=0$ , central wavelength 400 nm) and a time evolving vortex at (a2, b2)  $t_1=-45$  fs, (a3, b3)  $t_2=0$  fs, and (a4, b4)  $t_3=45$  fs (central wavelength: 800 nm). a: intensity spatial distributions; b: corresponding spatial phases.

To conclude, complex ultrafast spatiotemporal beams, even those exhibiting time evolving OAM, can be retrieved and characterised by means of a new bulk lateral interferometry technique allowing to measure wavefronts resolved on time and spectrum, being specially insensitive to perturbations. This technique has potential to work even at industrial environment and to be applied to imaging applications (e. g., microscopy). On the other hand, time varying vortices are singular light structures with very interesting potential in ultrashort science.

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# OC 8-Simultaneous attosecond-temporally and Angstrom-spatially resolved imaging of electron dynamics in molecules

M. Garg,<sup>a</sup> A. Martín-Jimenez,<sup>a</sup> M. Pizarra,<sup>b,c,†</sup> Y. Luo,<sup>a</sup> F. Martín,<sup>b\*</sup> and K. Kern<sup>a,d</sup>

<sup>a</sup> Max Planck Institute for Solid State Research, Stuttgart, 70569, Germany

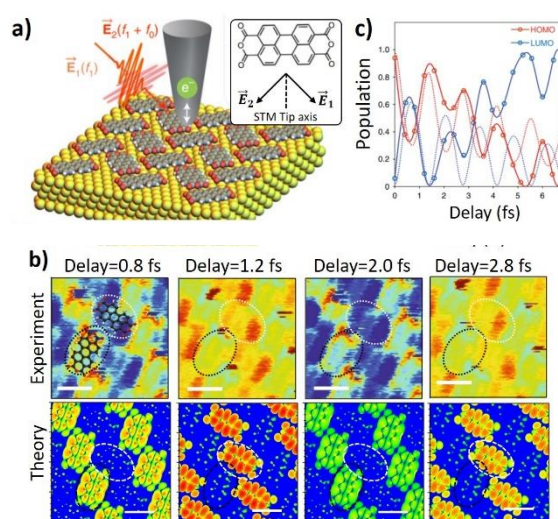
<sup>b</sup> Departamento de Química, Universidad Autónoma de Madrid, and Imdea Nano, Madrid, 28049, Spain.

<sup>c</sup> INFN-LNF Gruppo Collegato di Cosenza, Rende (CS), 87036, Italy.

<sup>d</sup> Institut de Physique, Ecole Polytechnique Fédérale de Lausanne, Lausanne, CH-1015, Switzerland.

E-mail presenting author: [alberto.martinj@imdea.org](mailto:alberto.martinj@imdea.org)

‘Seeing’ electron motion at both its natural length and time scales is a long-awaited dream of natural sciences. Reaching such spatial or temporal resolutions is nowadays possible, but not simultaneously. Current attosecond science techniques allows generating and tracking electronic motion in real time with incredible time resolution (200 attoseconds to few femtoseconds) in ever larger systems, but not in real space. Scanning tunneling microscopy (STM), on the other hand, can locally probe the electron density in molecules and solids with the necessary (picometer) resolution, but cannot provide by itself the dynamical information at the ultrafast time scale that is inherent to electronic motion. In this work,<sup>1</sup> we break this space-time limitation by combining STM and attosecond technologies (see Fig. 1a). We show that the coherent electronic motion generated by two temporally delayed CEP-stable near-infrared pulses of < 6 fs duration can be locally and non-invasively probed with both picometer and 300 attosecond resolution, thus allowing for a direct visualization of electron dynamics without the need for any additional reconstruction. We apply our technique to self-assemblies of PCTDA molecules deposited on a Au(111) surface. Thanks to state of the art DFT calculations we are able to correctly identify the specific electronic states that participate in the coherent oscillations and the corresponding dipole transition matrix elements. By combining this information with a two-state solution of the time-dependent Schrödinger equation, we are able to correctly predict the population of each state and the STM images recorded in the experiment as a function of the delay between the two pulses (see Fig. 1b and c).



**Figure 1.** a) Sketch of the experiment: Two ultrashort laser pulses impinge on a self-assembly of PCTDA molecules deposited on a gold surface, in tunnel contact with an STM. b) Comparison between the experimental and calculated STM images for different laser pulse delays for a 4-Layer PCTDA self assembly on Au(111): depending on the delay between the laser pulses either the HOMO or the LUMO in different molecules are visualized.<sup>1</sup> c) Experimental (solid lines and dots) and computed (dashed lines) populations for the HOMO and LUMO as function of the laser pulse delay.

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## OC 9-Ultrafast X-ray and optical time-resolved investigation of CuInS<sub>2</sub> quantum dots

A. Burgos-Caminal,<sup>a</sup> B. R. C. Vale,<sup>b</sup> A. F. V. Fonseca,<sup>c</sup> L. García,<sup>d</sup> J. F. Hidalgo,<sup>a</sup> O. Borrell-Grueiro,<sup>e</sup> T.K. Choi,<sup>f</sup> T. Katayama,<sup>g</sup> D. Fan,<sup>h</sup> S. Nozawa,<sup>h</sup> L. Bañares,<sup>e</sup> M. A. Schiavon<sup>c</sup> and W. Gawelda<sup>\*a,d</sup>

<sup>a</sup>IMDEA Nanoscience, Ciudad Universitaria de Cantoblanco, Calle Faraday 9, Madrid, 28049 Spain.

<sup>b</sup>Ultrafast laboratory spectroscopy, "Gleb Wataghin" Institute of Physics, University of Campinas, Unicamp, 13083-859, Campinas, Brazil.

<sup>c</sup>Grupo de Pesquisa Química de Materiais, Departamento de Ciências Naturais, Universidade Federal de São João Del-Rei, Campus Dom Bosco, 36301-160 São João Del-Rei, Minas Gerais, Brazil.

<sup>d</sup>Departamento de Química, Facultad de Ciencias, Universidad Autónoma de Madrid, 28049 Madrid, Spain.

<sup>e</sup>Departamento de Química Física and Center for Ultrafast Lasers, Facultad de Ciencias Químicas, Universidad Complutense de Madrid, 28040 Madrid, Spain.

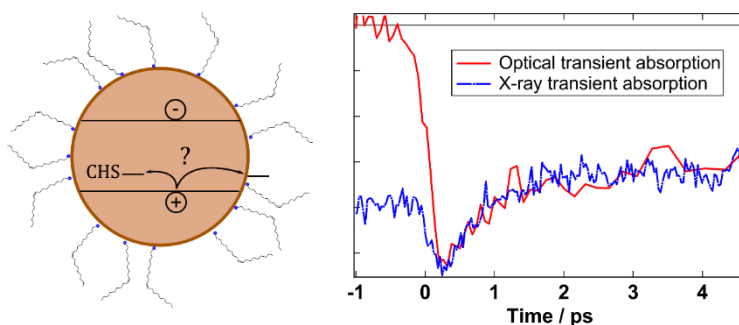
<sup>f</sup>Beamline Division, PAL-XFEL, 37673 Pohang, Korea.

<sup>g</sup>Japan Synchrotron Radiation Research Institute, Kouto 1-1-1, Sayo, Hyogo, 679-5198, Japan.

<sup>h</sup>Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), 1-1 Oho, Tsukuba, Ibaraki, 305-0801, Japan.

E-mail presenting author: [andres.burgos@imdea.org](mailto:andres.burgos@imdea.org)

The interest in copper-indium-sulfide (CIS) quantum dots (QDs), as heavy metal-free nanomaterials, has increased significantly in the past few years. CIS quantum dots (QDs) have been studied for many applications like photodynamic therapy, solar cells, LEDs, and bioimaging.<sup>1</sup> They show exciting optoelectronic properties, such as broad photoluminescence (PL) with a large Stokes shift and long charge carrier lifetimes. Several mechanisms for the radiative recombination in CIS QDs have been proposed over past years. In this work we aim to understand and confirm the possibility of radiative recombination resulting from an electron in the conduction band and a hole in a so-called confined hole state (CHS) or localized hole,<sup>2-3</sup> which is a mid-gap state arising from a defect of Cu<sup>+</sup> that can get oxidized to Cu<sup>2+</sup> by the hole. The range of possible defects would explain the broad PL and large Stokes shift. In addition, we aim to understand the effects of stoichiometry and Zn doping on the formation process of the CHS.<sup>4</sup> To these ends, the element and oxidation state specificity of X-ray techniques can be a great tool to carry out a more direct observation of such processes, allowing us to confirm the proposed mechanisms. We approached these questions through a combination of laser, synchrotron and XFEL techniques, including our recent results from SACLA XFEL and ALBA. We focused on tracking in real-time the oxidation state changes of Cu via femtosecond-resolved Cu K-edge XANES and comparing the structure of the different samples through steady-state EXAFS. We have complemented our investigation in the X-ray range with time-resolved optical studies monitoring ultrafast transient absorption and photoluminescence (with fluorescence up-conversion spectroscopy).



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# OC 10-Drilling holes in gold nanoparticles with ultrashort laser pulses

O. Peña-Rodríguez,<sup>\*,a</sup> J. C. Castro-Palacio,<sup>a</sup> G. González-Rubio,<sup>b</sup> P. Díaz-Núñez,<sup>a</sup> A. Prada,<sup>c</sup> R. I. Gonzalez,<sup>c</sup> T. Milagres de Oliveira,<sup>d</sup> W. Albrecht,<sup>d</sup> S. Bals,<sup>d</sup> L. Bañares,<sup>b</sup> L. M. Liz-Marzán,<sup>e</sup> A. Guerrero-Martínez,<sup>b</sup> J. M. Perlado,<sup>a</sup> J. Kohanoff<sup>a</sup> and A. Rivera<sup>a</sup>

<sup>a</sup> Instituto de Fusión Nuclear “Guillermo Velarde”, Universidad Politécnica Madrid, Spain.

<sup>b</sup> Departamento de Química Física I, Universidad Complutense de Madrid, Spain.

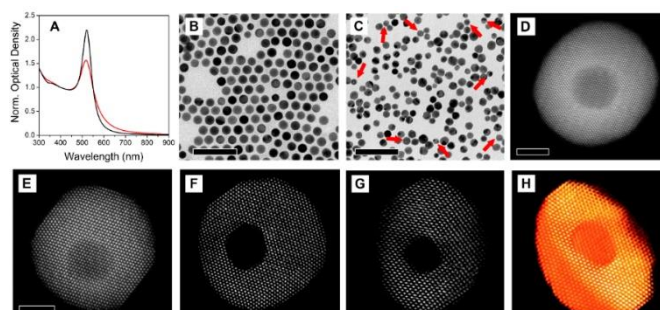
<sup>c</sup> Universidad Mayor, Santiago, Chile.

<sup>d</sup> EMAT-University of Antwerp, Antwerp, Belgium.

<sup>e</sup> BioNanoPlasmonics Laboratory, CIC biomaGUNE, Donostia-San Sebastián, Spain.

E-mail presenting author: [ovidio.pena@upm.es](mailto:ovidio.pena@upm.es)

Metallic hollow nanoparticles exhibit interesting optical properties that can be controlled by geometrical parameters.<sup>1</sup> Moreover, irradiation with laser pulses has emerged recently as a valuable tool for reshaping and size modification of plasmonic metal nanoparticles, thereby enabling the synthesis of nanostructures with unique morphologies. In this work, we demonstrate how we can use the irradiation with laser pulses to fabricate hollow nanoparticles. First, we use classical molecular dynamics simulations to investigate the solid-to-hollow conversion of gold nanoparticles upon femtosecond laser irradiation. Based on these results, we suggest an efficient method to produce hollow nanoparticles under certain specific conditions.<sup>2</sup> Next, we demonstrated that the irradiation of spherical nanoparticles with nanosecond laser pulses induces shape transformations yielding nanocrystals with an inner cavity (see Figure 1).<sup>3</sup> The concentration of the stabilizing surfactant, the use of moderate pulse fluences, and the size of the irradiated particles determine the efficiency of the process and the nature of the void. Hollow nanocrystals are obtained when molecules from the surrounding medium are trapped during laser pulse irradiation. These observations suggest the existence of a subtle balance between the heating and cooling processes, which induce the expansion and subsequent recrystallization of the nanocrystals, keeping exogenous matter inside. Therefore, we advance the experimental conditions to efficiently produce hollow nanoparticles, opening a broad range of possibilities for applications in key areas, such as gas and liquid storage and catalysis.



**Figure 1.** Irradiation of 20 nm Au NPs for 3 min with 8 ns laser pulses at a wavelength of 532 nm, a fluence of 0.2 J/cm<sup>2</sup>, and a repetition rate of 10 Hz. (A) Normalized optical density before (black) and after (red) irradiation. Low-magnification images (B) before and (C) after irradiation. (D, E) High-resolution images displaying the atomic structure of particles. (F, G) Orthoslices along different orientations. (H) Volume rendering, revealing the presence of a cavity inside a Au NP. Scale bars are 50 nm (B and C) and 4 nm (D and E).

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# OC 11-Laser-induced periodic surface structures in polymers with tailored laser fields

R. de Nalda,<sup>a</sup> M. E. Corrales,<sup>b</sup> P. Recio,<sup>b</sup> J. G. Izquierdo,<sup>b</sup> L. Bañares,<sup>b</sup> T. Ezquerra,<sup>c</sup> and E. Rebollar<sup>a</sup>

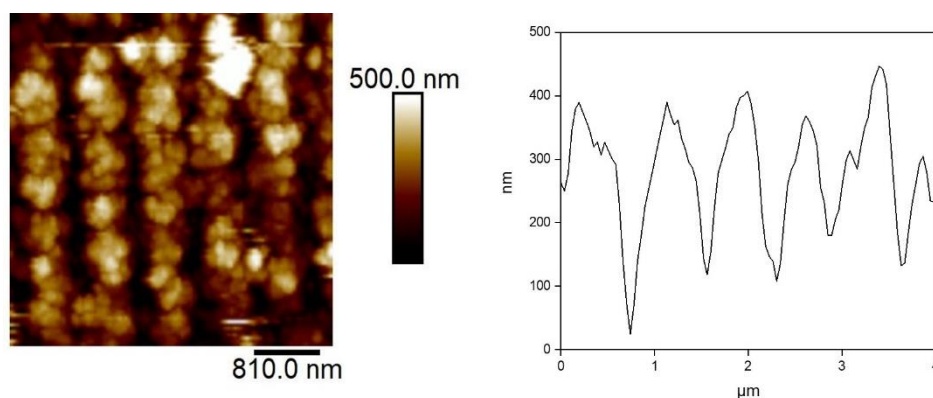
<sup>a</sup> Instituto de Química Física Rocasolano, CSIC, C/ Serrano 119, 28006 Madrid, Spain.

<sup>b</sup> Departamento de Química Física (Unidad Asociada I+D+i al CSIC), Facultad de Ciencias Químicas, Universidad Complutense de Madrid, 28040 Madrid, Spain.

<sup>c</sup> Instituto de Estructura de la Materia, CSIC, C/ Serrano 121, 28006 Madrid, Spain.

E-mail presenting author: [r.nalda@iqfr.csic.es](mailto:r.nalda@iqfr.csic.es)

The generation of periodic structures in material surfaces caused by intense, linearly polarized laser fields was observed shortly after the advent of the laser itself.<sup>1</sup> The observation of periodic patterns with periods of the order of the wavelength of the light, which are sensitive to the incidence angle and the laser polarization, was soon recognized as a universal phenomenon capable of inducing permanent ripples in metals, semiconductors and insulators. From its initial consideration as an unexpected laser phenomenon through a collateral effect of laser processing, it is now broadly considered an appealing process that can be used as a nanostructuring technique, and one that allows to address challenges in areas as varied as fluidics, optics, electronics or medicine, through its capacity to tune the properties of the laser-processed surfaces. In particular, in the case of polymers, surface micro- and nanostructures have been shown to allow for control of the optical and mechanical properties, as well as their hydrophobic/hydrophilic behaviour. Despite intense theoretical and experimental work over the last decades, the field of LIPSS is still a fruitful area, with important challenges to be addressed and indeed numerous open questions, many of which have been reviewed recently by Bonse and Gräf.<sup>2</sup> In the present work, periodic surface structures have been generated on the surface of thin films of aromatic polyesters fabricated by spin coating on Si. Previous studies had shown the appearance of LIPSS in this type of materials with femtosecond laser pulses in the near-infrared and the near-ultraviolet regions of the spectrum.<sup>3-4</sup> The present work consists of an exploration of the effect of adding complexity to the femtosecond laser fields employed for irradiation. The effect of bichromatic irradiation, as well as chirp and repetition rate, on surface topography and properties will be described and discussed.



**Figure 1.** AFM topography image of an 804-nm femtosecond laser-treated surface of poly(trimethylene terephthalate).

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# OC 12-Laser industrial applications and transfer tecnology from CLPU

M. Rico,\*<sup>a</sup> R. Lera<sup>a</sup> and L. Roso<sup>a</sup>

<sup>a</sup> Centro de Láseres Pulsados, Parque Científico, c/ Adaja 8, E-37185 Villamayor, Salamanca, Spain.

E-mail presenting author: [mrco@clpu.es](mailto:mrco@clpu.es)

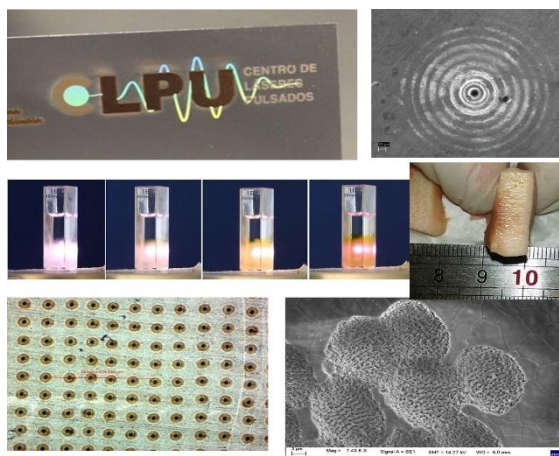
The scope of this summary is to describe the R&D work carried out by CLPU to respond to the needs of our scientific and industrial users over the last few years.

From the CLPU we have focused on providing support and solutions to our users in all possible applications where lasers (mainly with femtosecond pulsed lasers). To this end, we have applied the scientific knowledge acquired in the field of lasers in the search for practical solutions in different fields of work. In the last years we have been able to give a varied offer in biomedical applications, chemical process improvements, electrical switching systems, surface modification, laser drilling and cutting, laser microprocessing by ablation and filamentation, etc.

Figure 1 show some examples of pulsed laser applications: LIPSS manufacturing (witj logo of CLPU), laser microprocessing with axicon (Bessel beams), catalytic enhancement in chemical reactions (reduction time), biological material cutting (whitout damage) and structured surface modification for multiples applications.

In addition, different technology transfer projects have been developed in different fields, where the development of new pulsed light sources or the development of new lasers dedicated to specific applications have been the core of our work. To this end, we have collaborated with the automotive and aeronautical industries and in the defense and security sector.

The final objective of this abstract is to present some of our most relevant work within the framework of lasers and their applications, relating scientific knowledge with their final applications.



**Figure 1.** Six examples of pulsed laser applications are shown: LIPSS manufacturing, laser microprocessing with axicon, catalytic enhancement in chemical reactions, biological material cutting and structured surface modification.



# POSTERS

# P1- $\pi$ -Quinoidal molecules with excited state symmetry breaking: Towards efficient two photon absorption.

F. Gordillo Gámez,<sup>a</sup> J. Manuel Marín Beloqui<sup>a</sup> and J. Casado<sup>a</sup>

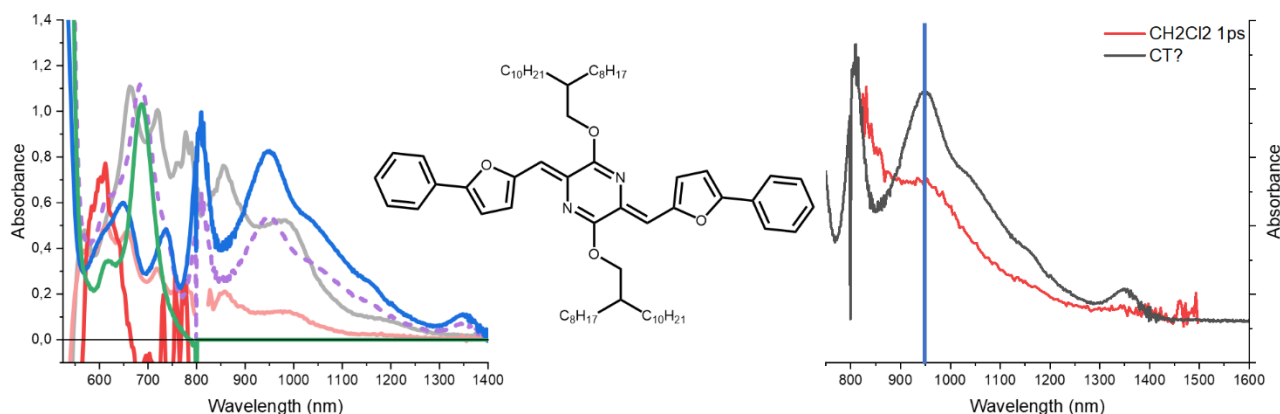
<sup>a</sup> Departamento de Química Física, Universidad de Málaga, Campus de Teatinos s/n, 29071 Málaga, Spain.

E-mail presenting author: [fgordillo@uma.es](mailto:fgordillo@uma.es)

In recent years, quinodimethanes have attracted the interest of the scientific community due to their particular optical and electronic properties, both of which are related to conjugated structures in open and closed shells.<sup>1</sup> These characteristics, derived from their intrinsic diradical character, allow their use in organic electronics and polymer science, since they are able to reduce the band gap of the resulting polymer, in a donor-acceptor approach, with electron-rich and electron-deficient units alternating in the polymer backbone. The addition of heteroatoms to the quinoid structure also improves the stability of the resulting molecule. This has recently been verified with derivatives of para-azaquinodimethane (p-AQM), where thiophene groups have been introduced,<sup>2</sup> and they have shown suitable properties, such as high hole transporting mobilities or low band gap via bond length alternation (BLA) when used as building blocks in polymers.<sup>3</sup> These properties make p-AQM an ideal building block in organic electronic materials.

One of the main characteristics of this molecule is coplanarity due to the interaction between N-S through space. In order to improve our knowledge of this kind of systems, we change thiophene groups (well known) by furane (new approach), to find new features while keeping the coplanarity of the organic backbone, due to a new N-O interaction.

In this work, we present a new molecule derived from p-AQM, named AQM-FPh<sub>2</sub>, with furan groups attached to phenyl. Its optical properties have been studied, through spectroelectrochemical and transient absorption spectra (TAS). With the spectroelectrochemistry characterization showed a band at a high wavelength, associated with a charge-transfer (CT) state, and TAS showed a band at the same region and similar to the former. The results obtained indicate the appearance of symmetry-breaking states, suggesting this molecule as a potential candidate for two-photon absorption.



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# P2-1D/3D modelling of cavity-free Nitrogen lasing

E. Oliva<sup>\*a,b</sup> and S. López<sup>a,b</sup>

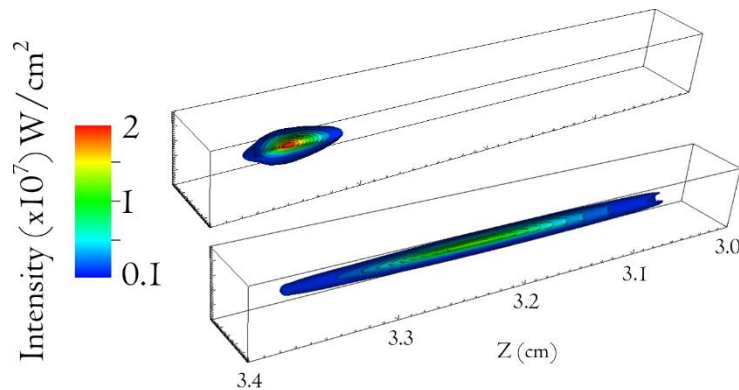
<sup>a</sup> Instituto de Fusión Nuclear “Guillermo Velarde”, Universidad Politécnica de Madrid, Madrid, Spain.

<sup>b</sup> Departamento de Ingeniería Energética, E. T. S. I. Industriales, Universidad Politécnica de Madrid, Madrid, Spain.

E-mail presenting author: [eduardo.oliva@upm.es](mailto:eduardo.oliva@upm.es)

The so-called air lasing, cavity-free coherent emission of UV light originating from the components of the atmosphere (mainly Oxygen and Nitrogen) has attracted a lot of attention the past years due to its potential as a remote source in the atmosphere for ultrasensitive LIDAR applications.<sup>1</sup> While lasing from molecular Nitrogen (neutral and ionized) has been demonstrated, there is still a lack of fundamental knowledge that hinder the development of these sources for applications.

In order to understand more thoroughly the physics and dynamics of these amplifiers, new experimental and modelling data is required. This is a multiscale, from femtoseconds (the duration of the intense IR pulse that creates the filament) to picoseconds (amplification dynamics) and nanoseconds (plasma hydrodynamics), multiphysics (plasma physics, atomic processes, laser-plasma interaction) problem. For this task we have used our Maxwell-Bloch, time-dependent codes, 1D-DeepOne<sup>2</sup> and 3D Dagon<sup>3</sup> to model cavity-free lasing in N<sub>2</sub> and N<sub>2</sub><sup>+</sup> plasma filaments. The combined experimental and modelling results allowed us to unveil the lasing mechanism in neutral Nitrogen,<sup>4,5</sup> electron collisional excitation. Recent modelling results obtained with our codes, related to N<sub>2</sub><sup>+</sup> lasing, provide hints about the mechanism responsible of the lasing effect. Moreover, our 3D simulations demonstrate that the amplified beam carries information in its wavefront about the spatio-temporal dynamics of the plasma amplifier. Thus, measuring the wavefront of the amplified beam is foreseen as a promising diagnosis technique for plasma filaments.



**Figure 1.** Intensity of an amplified UV seed beam after passing through more than 3 cm plasma amplifier of neutral Nitrogen. The upper panel shows the result when using the adiabatic approximation while the lower computes the full temporal dynamics of the plasma polarization.

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# P3-Amplification of HOH carrying orbital angular momentum in plasma-based amplifiers

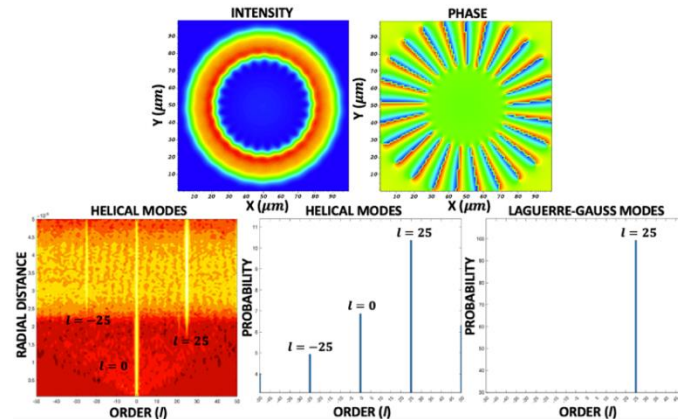
S. López,<sup>a,b</sup> E. de la Fuente,<sup>a,b</sup> A. Alonso<sup>a,b</sup> and E. Oliva<sup>\*a,b</sup>

<sup>a</sup> Instituto de Fusión Nuclear “Guillermo Velarde”-E. T. S. I. Industriales, Universidad Politécnica de Madrid, Madrid, Spain.

<sup>b</sup> Departamento de Ingeniería Energética-E. T. S. I. Industriales, Universidad Politécnica de Madrid, Madrid, Spain

E-mail presenting author: [s.lopezg@upm.es](mailto:s.lopezg@upm.es)

In this work, two concepts are brought together that by themselves generate a lot of interest today. On the one hand, for 30 years we have been aware of a hitherto unnoticed property of light, called orbital angular momentum (OAM).<sup>1</sup> The intrinsic display of this property by certain beams has allowed their application in fields such as telecommunications<sup>2</sup> or biomedicine.<sup>3</sup> On the other hand, free electron lasers (FEL) are currently the X-ray laser sources that achieve the best performance, essential for certain applications. However, the uniqueness and cost of these installations have led to the search for more compact sources. One of the most promising alternatives arises from the combination of other available sources, that is, the injection of high-order HOH harmonics, with excellent optical properties, into plasma amplifiers.<sup>4</sup> Now, what happens when you inject an HOH with OAM into a plasma? Is an amplified beam obtained with the same OAM or has this property been affected? This is what we have tried to explain here. Knowing the response of the OAM to this process will lay the groundwork for new applications, according to what was mentioned in the previous paragraph.



**Figure 1.** Left to Right. Up: Intensity and phase after propagating 4 mm. Down: radial distribution and decomposition in helical modes and Laguerre-Gauss decomposition

The results, which have been obtained from the 3D, time-dependent, Maxwell-Bloch code, Dagon, show that OAM is conserved in low-density, nickel-like, krypton plasmas (Figure 1). As this density increases, which is interesting for solid target plasmas such as nickel-like silver, the OAM is also preserved, but now there is curvature in the phase jump lines that should be taken into account in further studies. Additionally, a first study has been carried out for a plasma waveguide,<sup>5</sup> which makes it possible to reduce harmful phenomena such as refraction. The imprinted footprints, both in intensity and phase, resulting from the interaction with the spontaneous emission (ASE) and electronic density gradients, could find application in plasma diagnosis methods for applications like Inertial Confinement Fusion or Laboratory Astrophysics.

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# P4-Angle-resolved phases in Argon photoemission

R. Y. Bello,<sup>a</sup> V. J. Borràs,<sup>a</sup> J. González-Vázquez<sup>a</sup> and F. Martín<sup>a,b,c</sup>

<sup>a</sup> Departamento de Química, Universidad Autónoma de Madrid, 28049 Madrid, Spain

<sup>b</sup> Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA Nano), Cantoblanco, 28049 Madrid, Spain

<sup>c</sup> Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, 28049 Madrid, Spain

E-mail presenting author: [fernando.martin@uam.es](mailto:fernando.martin@uam.es)

The Attosecond science has paved the way for studying electron dynamics in real time, from atomic to molecular systems. In particular, attosecond interferometric techniques have been used to access not only the ionization amplitudes but also the phases associated with the scattering states describing the ejected photoelectron [1], thus opening up the prospect of steering molecular dynamics, extracting photoionization time delays and more importantly reconstruct electronic wave-packets. Currently, two methods are mainly used to retrieve the phases and amplitudes: the streaking technique, which combines a single attosecond pump pulse with an infrared probe pulse, and the RABBITT technique, which combines a train of attosecond pulses with the infrared pulse used to generate them. Recently an alternative method for measuring the phases in photoemission has been put forward [2, 3]. This method combines two phase-locked extreme ultraviolet pulses of frequency  $\omega$  and  $2\omega$ , from a free electron laser. By varying the relative optical phase between the two pulses, the phase difference between the two ionization channels can be measured. In the present work, we apply this technique to theoretically obtain the phase difference, as a function of the electron emission angle, between the two ionization channels in the Ar atom. We pay special attention to the energy region where one or both of the two colors ( $\omega$  or  $2\omega$ ) is in resonance with a bound electronic state or with an autoionizing Rydberg state.

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# P5-Developments for laser-driven ion acceleration at high repetition rate

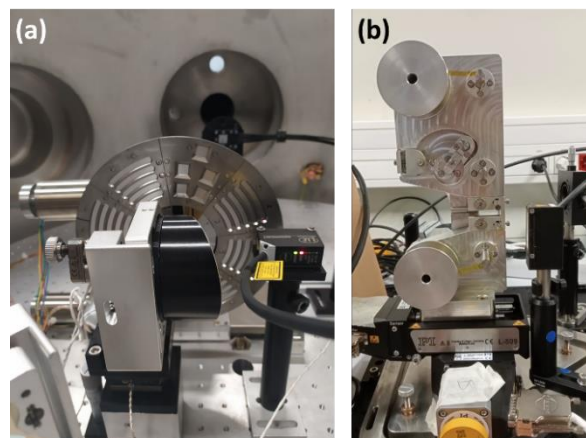
A. Bembibre,<sup>a</sup> J. Peñas,<sup>a</sup> J. Benlliure<sup>a</sup> and A. Alejo<sup>\*a</sup>

<sup>a</sup> Instituto Galego de Física de Altas Enerxías (IGFAE). Universidade de Santiago de Compostela (Spain).

E-mail presenting author: [adrian.bembibre.fernandez@usc.es](mailto:adrian.bembibre.fernandez@usc.es)

Interest in laser-driven ion acceleration has increased dramatically over last decades since it could lead to a competitive alternative to conventional ion accelerators, thanks to a comparatively reduced size, shielding and operation costs.<sup>1</sup> Laser-driven accelerators produce ultrashort bunches of ions and electrons, as well as gamma rays, with large flux and broad spectrum. These properties are beneficial for a number of techniques, including FLASH cancer treatment,<sup>2</sup> proton radiography of micro and nano structures, neutron source via a secondary target or the electron-driven fast ignition (FI) scheme in inertial confinement fusion (ICF).<sup>1,3</sup>

With the advent of high-power lasers working at multi-Hertz repetition rates, there has been a growing demand for novel target and diagnostics capable of operating at such rates. Here, we present some recent developments on laser-driven ion acceleration at the Laser Laboratory for Acceleration and Applications (L2A2, Universidade de Santiago de Compostela), which hosts the STELA laser, a 45 TW system that can deliver pulses with energies of up to 1.2 J and pulses of 25 fs at 10 Hz. Significant progress has been made on target and diagnostics development. As the target is destroyed following the laser-solid interaction, the target needs to be continuously replaced for a fresh one, while keeping the target at the focal plane with micron precision. In this context, two target systems have been built at L2A2. Firstly, a multi-target rotating wheel that allows continuous irradiation at 10 Hz up to hundreds of shots, thanks to the combined use of a positioning sensor (Fig. 1a). Secondly, in order to further extend the number of irradiations, a tape-drive system has been manufactured which enables the operation for thousands of shots while ensuring the tension and position of the tape target to remain stable within a few microns (Fig. 1b). In addition to targetry, recent work on diagnostics will be presented. Ion characterisation tools based on plastic scintillators have been tested to measure ion beam properties, particularly its spatial profile by means of stacks of plastic scintillating plates, as well as the energy spectrum taking advantage of the time-of-flight (ToF) technique. Furthermore, the use of scintillating fibres is currently being studied as a hybrid solution that would allow to simultaneously measure both the spatial and the spectral properties of the ion beams. Preliminary results resulting of ions accelerated at L2A2 from the aforementioned developments will be presented.



**Figure 1.** (a) Wheel system. (b) Tape-drive system.

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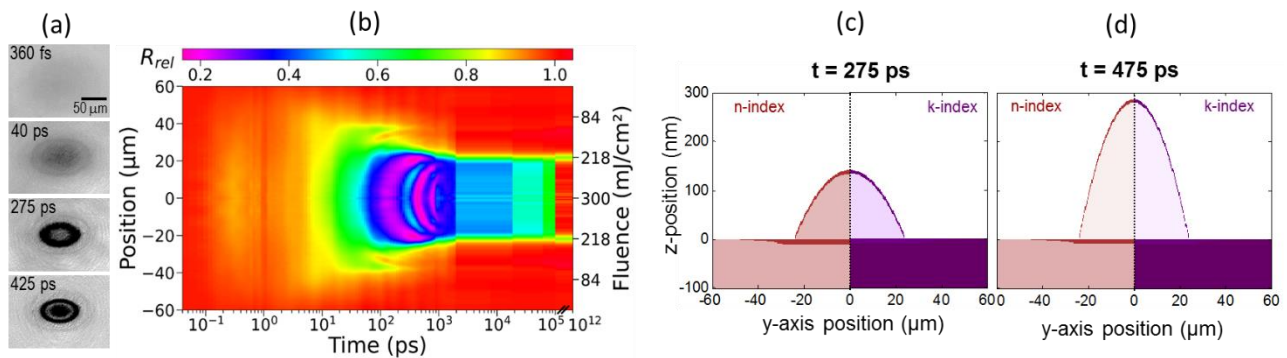
# P6-Electron dynamics and optical interference tomography of femtosecond laser excited steel

Y. Fuentes-Edfuf,<sup>a</sup> M. Garcia-Lechuga,<sup>a</sup> J. Solis<sup>a</sup> and J. Siegel<sup>a,\*</sup>

<sup>a</sup> Laser Processing Group, Instituto de Óptica, IO-CSIC, Serrano 121, 28006 Madrid, Spain.

E-mail presenting author: [j.siegel@csic.es](mailto:j.siegel@csic.es)

While femtosecond laser machining has already become a mature, high-precision technology, the complexity of the underlying laser-matter interaction processes, still imposes limits to reaching the exact surface morphology, topography and/or functionality intended. Even when powerful theoretical methods are available to simulate complex laser-matter interaction scenarios, it is desirable to verify their results with experimental studies based on techniques with high temporal resolution in order to follow the ultrafast material transformation dynamics triggered by ultrashort laser excitation. We report an optical pump-probe microscopy study of the electron dynamics and material transformation in steel upon single laser pulse irradiation (800 nm, 120 fs) incident at an angle of 52° and with an Gaussian intensity distribution at the sample plane.<sup>1</sup> Time-resolved reflectivity images of the irradiated regions were obtained by means of a home-built optical microscope and illuminating the sample with single, time-delayed 400 nm wavelength, 100 fs pulses at normal incidence. Selected time-resolved images are shown in Figure 1(a). A powerful strategy to visualize the complex excitation, relaxation, melting, ablation and solidification dynamics consists in generating a time-, position-and fluence-resolved map by extracting radial reflectivity profiles of the single set of images<sup>1</sup> recorded in Figure 1(a). The result is displayed in Figure 1 (b), in which the reflectivity is encoded in false colour and the fluence scale has been obtained by employing the direct relation of the radial position with respect to the spot center and the corresponding local fluence of the Gaussian intensity distribution. The richness in information of this map is evident, as it allows perceiving quickly the temporal evolution at different local fluences by drawing horizontal (temporal) reflectivity cross sections. This representation allows to observe both ultrafast phenomena (electron excitation) and longer lasting phenomena (material expansion), both in the heating/melting regime as well as in the ablation regime. Moreover, combining the obtained time- and fluence-dependent reflectivity map with modelling of the reflectivity of a multilayer material, using the optical properties of the different phases involved,<sup>2</sup> it is possible to obtain a 3D tomographic reconstruction of the ultrafast-evolving layered structure. That way, it has been possible to unravel the dynamics of the several processes involved, including excitation, melting, ablation and solidification.



**Figure 1.** (a) Time-resolved images of the relative reflectivity changes of a steel surface upon single pulse fs laser excitation ( $F = 300 \text{ mJ}/\text{cm}^2$ ), recorded at different delay times  $t$  (see labels) after the arrival the pump pulse (the pump pulse is centred at  $t = 250$  fs). (b) Map of laser-induced relative reflectivity changes  $R_{rel}$  (false colour scale) as a function of delay time (horizontal scale), spatial position with respect to the spot center (left vertical scale) and local fluence (right vertical scale, related to the spatial position), extracted from the full set of time-gated images, including those shown in (a). (c) and (d): Tomography cross sections of the ablating layer at two different delays after arrival of the fs excitation pulse.

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# P7-Fundamental mechanisms of high-order harmonic generation in graphene: perfect and imperfect recollisions

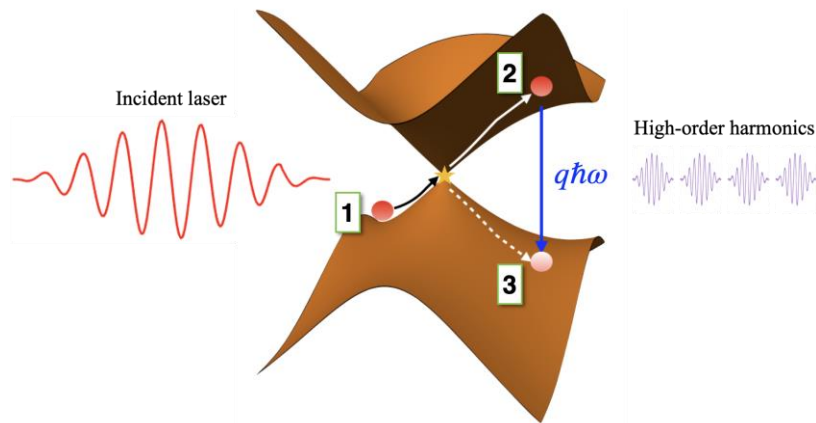
Ó. Zurrón Cifuentes,<sup>\*a</sup> R. Boyero-García,<sup>a</sup> A. García-Cabrera,<sup>a</sup> C. Hernández-García<sup>a</sup> and L. Plaja<sup>a</sup>

<sup>a</sup> Grupo de Investigación en Aplicaciones del Láser y Fotónica, Departamento de Física Aplicada, Universidad de Salamanca, E-37008, Salamanca, Spain.

E-mail presenting author: [ozurronci@usal.es](mailto:ozurronci@usal.es)

High-order harmonic generation (HHG) has become an essential tool to understand the interaction between intense laser fields and physical systems. In gases, the basics of such interaction are well known and can be readily described from a semiclassical point of view.<sup>1</sup> According to it, in a first step the intense laser radiation detaches an electronic wavepacket from the atom via tunnel ionization, which evolves in the continuum with the electric field; then, upon reversal of the field amplitude, the electronic wavepacket is redirected to the parent ion, where it recombines emitting a high-frequency photon. HHG in solid systems has attracted a considerable interest during the past decade, since their higher density should, in principle, increase the efficiency of the process. HHG in bulk solids follows a similar path than in gases, though the first step leads to the creation of an electron-hole pair which follows trajectories that depend on the band structure, and the emitted photon is resonant with the band gap at the recombination time.<sup>2</sup> Nevertheless, the remarkable electronic structure of Dirac-Weyl materials like single layer graphene (SLG) opens up new scenarios for the study of HHG.

More specifically, in this contribution we show that the interaction of SLG with a strong laser field exhibits a different behaviour than that of bulk solids. The gapless band structure of SLG plays a fundamental role in the formation of the electron-hole pair, so that tunnel excitation is replaced by the non-adiabatic crossing through the Dirac points (see figure).<sup>3</sup> Furthermore, it has been recently reported that a substantial part of the HHG emission in solids corresponds to situations where the electron and hole trajectories do not overlap in space.<sup>4</sup> According to the present knowledge, HHG from this *imperfect recollisions* reflects the quantum nature of the process, arising in systems with large Berry curvatures or for elliptically polarized driving fields. In this work, we demonstrate that imperfect recollisions are also relevant in the case of SLG –a system with null Berry curvature– irradiated by linearly polarized driving fields.<sup>5</sup>



**Figure 1.** Scheme of the mechanism for high-order harmonic generation in graphene

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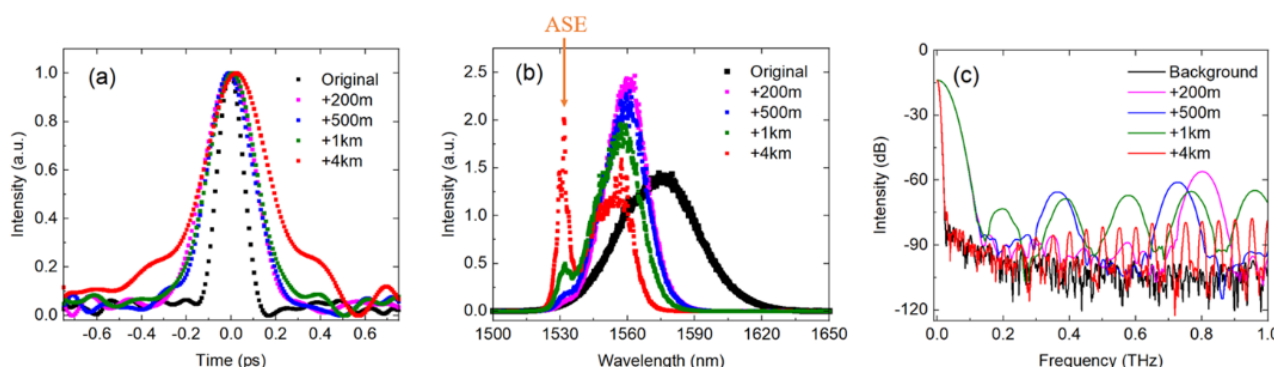
# P8-High-power ultrafast InN-based all-fiber laser

L. Monroy,<sup>\*a</sup> M. González-Herráez<sup>a</sup> and F. B. Naranjo<sup>a</sup>

<sup>a</sup> Grupo GRIFO, Dpto. Electrónica, Edificio Politécnico, Universidad de Alcalá, 28805, Alcalá de Henares, Spain.

E-mail presenting author: [laura.monroy@uah.es](mailto:laura.monroy@uah.es)

Ultrafast fiber lasers can be applied to a wide variety of applications, including laser micromachining, material processing, surgery, and electronics. Ultrashort pulses can be generated in an inexpensive and simple manner using mode-locked lasers, which have demonstrated greater characteristics, including efficiency, stability, and ease of implementation.<sup>1</sup> Furthermore, fiber lasers operating at 1550 nm are compatible with optical communications systems since they are located within a spectral range with a minimum loss region. In this work an all-fiber resonator cavity has been employed for high-power ultrafast mode-locking laser development, based on an InN semiconductor saturable absorber mirror (SESAM) using a graded-index (GRIN) lens as the fiber coupling device.<sup>2</sup> The rest of the fiber components are based on standard single-mode fiber (SMF) and erbium-doped fiber amplifier (EDFA) with a total length of 50 m and a dispersion coefficient of  $-0.21 \text{ ps}^2$ , thus working in the anomalous dispersion regime as a dispersion-managed cavity. This assembly permits to reduce the insertion losses generated by open-path configurations, as well as to increase the efficiency of the saturable absorber, leading to ultra-stable sub-100 fs optical pulses centered at 1560 nm with a maximum peak power of 65 kW. However, in order to increase the optical power within the fiber laser cavity, we here propose to change the length of the cavity, and thus the emission repetition rate, by adding additional fiber spans of SMF. The stability of the femtosecond pulse duration, spectral bandwidth and repetition rate has been analyzed for different lengths ranging from 50 m (original set-up) to up to 4km, as depicted in Fig. 1 (a), (b) and (c) respectively. The minimum temporal duration was measured for the fundamental mode for the original set-up with a value of 92 fs (42 nm in the spectral domain) and a repetition rate of 5.2 MHz, whereas a maximum of 180 fs (20 nm of spectral bandwidth) is obtained when a fiber span of 4 km is incorporated with a repetition rate of 50 kHz. This suggests that the dispersion coefficient is sufficiently high as to deteriorate the quasi-soliton behavior of the optical pulse propagation through the laser cavity. Furthermore, the central wavelength of the optical pulse is blue-shifted, due to the increase of the dispersion effects within the SMF. However, it can also be seen that as we increase the length of the laser cavity, a higher contribution of the continuous component is measured on the optical spectrum as we approach to the wavelength of the amplified spontaneous emission (ASE) of the EDFA. These effects are translated in a higher chirp estimated from the time-bandwidth product (TBP), ranging from 0.44 to 0.64 for the original and the 4 km fiber span configurations respectively. In relation to the peak power, it has been possible to increase the value from the original case, with a peak power of 65 kW to its maximum value of 1.08 MW for the 4km-long cavity at the output of the ring fiber resonator cavity. In summary, we here expose preliminary results of ultrafast pulse generation at high optical powers by the addition of SMF optical fibers to the ring fiber laser cavity based on the InN SESAM coupled to a GRIN-to-fiber device. The decrease of the insertion losses, along with the reduction of the repetition rate, leads to high peak powers of up to 1 MW.



**Figure 1.** Autocorrelation (a), spectrum (b) and repetition rate (c) results for each fiber span of the laser cavity.

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# P9-Microwave spectroscopy coupled with a laser ablation source: Chemical applications

S. Mata,<sup>a</sup> L. Kolesnikova,<sup>b</sup> S. Mato,<sup>a</sup> R. Aguado,<sup>a</sup> M. Sanz-Novo,<sup>a</sup> E. R. Alonso,<sup>a</sup> I. León<sup>a</sup> and J. L. Alonso<sup>\*a</sup>

<sup>a</sup> Grupo Espectroscopia Molecular (GEM), Edificio QUIFIMA, Parque Científico Uva, Valladolid, Spain.

<sup>b</sup> Department of Analytical Chemistry, University of Chemistry and Technology, Prague, Prague, Czech Republic.

E-mail presenting author: [jalonso@qf.uva.es](mailto:jalonso@qf.uva.es)

Microwave spectroscopy has long been considered to be a robust method for the precise determination of structures of any gaseous molecular system, being able to distinguish between different conformational structures of a molecule. Some compounds, specifically biomolecules, present low vapor pressures at room temperatures and tend to decompose by standard heating methods. Thus, the study of the whole series of relevant biomolecules escapes from high-resolution rotational spectroscopy due to its high melting points and associated low vapor pressures. Laser ablation offers an alternative approach in which the energy of a high-intensity pulsed laser is used for fast desorption from nonvolatile samples. At the University of Valladolid, laser ablation (LA) coupled with narrowband MB-FTMW and broadband CP-FTMW Fourier transform microwave techniques in supersonic jets have been developed in our laboratories<sup>1,2</sup> to overcome the problems of thermal decomposition associated with conventional heating methods. Detailed structural information has been reported on many building blocks using this experimental approach.<sup>3</sup> We are exploring the full scope and capabilities of rotational spectroscopy coupled to a laser-ablation vaporization source as a reliable characterization technique to unravel dynamic molecular systems.<sup>4</sup>

The high amount of reactive species participating in the chemistry in the Earth's upper atmosphere, interstellar medium, or combustion processes motivates laboratory experimentalists to develop efficient methods for the molecular *in situ* generations to characterize these species spectroscopically. We propose a new approach based on the laser ablation of solid organic compounds. Once formed in the throat of our laser ablation source, the products are stabilized in the cold environment of the supersonic expansion and are monitored by high-resolution Fourier transform microwave techniques. We experimentally demonstrate a simultaneous formation of an impressive number and variety of species.<sup>5</sup> With these achievements in our hands, we extend the boundaries for the molecular *in situ* generations beyond traditional techniques such as electric discharge and pyrolysis.

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# P10-Non-thermal radiation emission from an X-ray laser-produced plasma

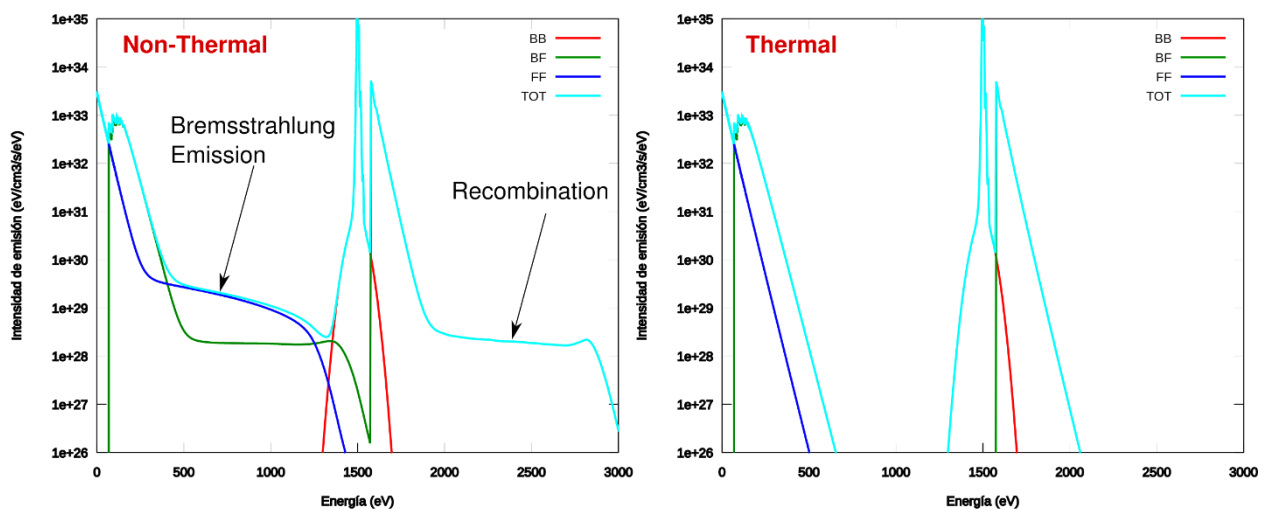
M. Coteló,<sup>a,b</sup> E. Vázquez,<sup>a,b</sup> E. Oliva<sup>a,b</sup> and P. Velarde<sup>a,b</sup>

<sup>a</sup> Instituto de Fusión Nuclear Guillermo Velarde, Universidad Politécnica de Madrid, calle Ramiro de Maeztu, Madrid, Spain.

<sup>b</sup> Departamento de Ingeniería Energética, Universidad Politécnica de Madrid, calle Ramiro de Maeztu, Madrid, Spain.

E-mail presenting author: [manuel.cotel@upm.es](mailto:manuel.cotel@upm.es)

With new ultra-intense lasers, such as the XFEL, both planned and in operation, the effects of non-thermal electronic distributions (EDF) are important. This deviation from the thermal distribution comes from the time dependence of the EDF and atomic populations. To study this problem, it is necessary to couple the time evolution of the atomic populations with the time evolution of the EDF and of the radiation energy distribution. We have studied the change in the radiation emission due to the time evolution of the EDF in dense plasmas generated by XFEL (see figure). To numerically simulate this process, we coupled the time evolution equations of the atomic populations with the Fokker-Planck time equation for electrons. This implies recalculating at each instant all atomic process rates, as well as the coefficients and independent terms of the FP equation. A signature of non-equilibrium processes is the emission of radiation differentiated from that of equilibrium. We present and explain the differences between such emissions with and without time dependence, pointing out the possibility of measuring the time effects of the EDF. To study this time-dependent phenomena, we used the atomic physics code BigBART that has been validated for the calculation of spectral opacities for plasmas in equilibrium. In addition, we have developed other computational tools that include an EM solver for time-dependent refractive index and calculations of X-Ray Thompson scattering for the diagnosis of plasmas.



**Figure 1.** Differences in emission intensity between the model including time evolution of the EDF (left panel) and the model using the equilibrium EDF (right panel). The main differences come from the recombination and the Bremsstrahlung emission. We detail the emission from different transitions: bound-bound (BB), bound-free (BF), free-free (FF) and the sum of these components, the total emission (TOT).

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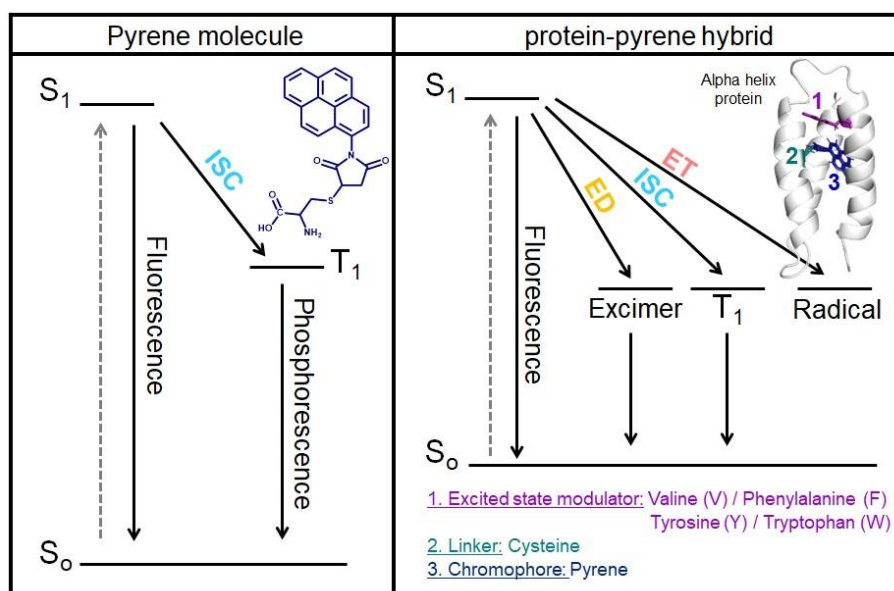
# P11-Protein-promoted chromophore excited-state decay modulation

S. H. Mejías

IMDEA Nanociencia. Faraday 9, Cantoblanco, Madrid. Spain.

E-mail presenting author: [sara.hernandez@imdea.org](mailto:sara.hernandez@imdea.org)

In natural photosynthesis, light energy transfers through a dynamic protein-pigment network with a remarkable near-unity quantum efficiency. Local fluctuations in the protein structures and conformational changes are essential for nature's efficient light conversion, but their design principles remain to be elucidated. We link pyrene chromophore to modified alpha helix proteins by specific linkers and develop bio-hybrid assemblies where chromophore's molecular environment is tunable at the nanoscale. We study pyrene excited state dynamics using time-resolved spectroscopy and correlate excited decay kinetics with protein's structural changes using NMR and molecular dynamics. In the protein, pyrene show multiple decay channels, which are not observed when pyrene is in solution (Figure 1). Interestingly, changes in the protein's backbone induced by specific amino acids make pyrene excitation follow a preferential decay pathway. Our study aims to develop protein-based bio-hybrids which use controlled protein dynamics to improve light energy conversion in biomimetic systems.



**Figure 1.** Left, pyrene S1 excited-state decay scheme via fluorescence and intersystem crossing (ISC) promoted phosphorescence. Right, pyrene S1 excited state decay scheme embedded in the protein via fluorescence, excimer emission, phosphorescence and radical recombination promoted by exciton dimerization (ED), intersystem crossing and electron transfer reactions inside the protein.

# P12-Role of bonding in metalloporphyrin-nanographene conjugates

V. Vega-Mayoral,<sup>a</sup> S. Garcia-Orrit,<sup>a</sup> A. Götz,<sup>b</sup> A. Narita,<sup>b,c</sup> K. Müllen<sup>b,d</sup> J. Cabanillas-González<sup>a\*</sup>

<sup>a</sup> Madrid Institute for Advanced Studies, IMDEA Nanociencia, c/Faraday 9, Campus de Cantoblanco, Madrid 28049, Spain.

<sup>b</sup> Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany.

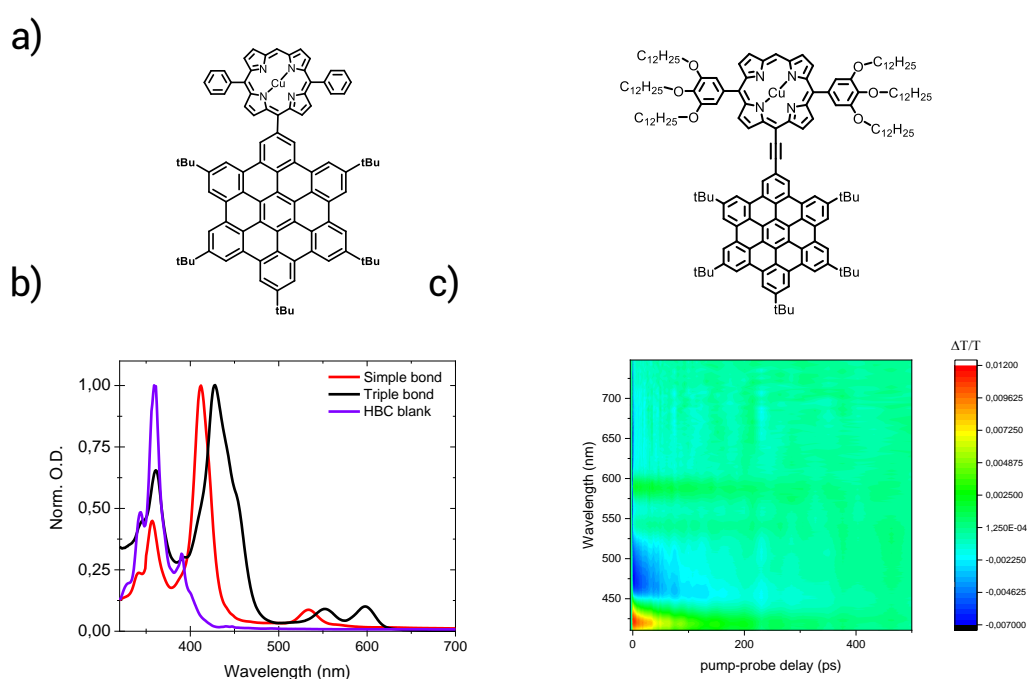
<sup>c</sup> Organic and Carbon Nanomaterials Unit, Okinawa Institute of Science and Technology Graduate University, Okinawa 904-0495, Japan.

<sup>d</sup> Institute for Physical Chemistry, Johannes Gutenberg University Mainz, Duesbergweg 10-14, 55128 Mainz, Germany.

E-mail presenting author: [victor.vega@imdea.org](mailto:victor.vega@imdea.org)

We present here, for the first time in our knowledge, a detailed study of photoexcited states dynamics in metalloporphyrin-nanographene conjugates. The potential application of these materials in the photovoltaic industry has rocketed the interest towards these materials.

Our study focuses on the differences merging from a different bonding between nanographene and porphyrin (simple or triple bond). Femtosecond transient absorption measurements and time resolved photoluminescence has been used to study the of photoexcited states dynamics immediately after photoexcitation.



**Figure 1.** a) Structures of the two studied metallo porphyrin-nanographene conjugates b) absorption spectra of the two studied conjugates and its comparison with HBC c) Transient absorption measurement of the triple bond conjugate.

# P13-Suppressing dissociation by circularly polarized light-induced potentials

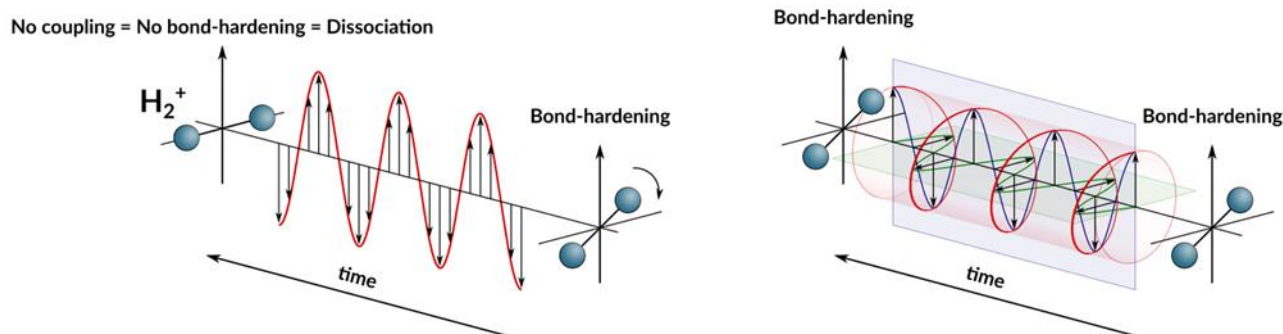
S. C. Carrasco,<sup>\*a</sup> V. S. Malinovsky<sup>a</sup> and I. R. Sola<sup>b</sup>

<sup>a</sup> DEVCOM Army Research Laboratory, Adelphi, Maryland 20783, USA.

<sup>b</sup> Departamento de Química Física, Universidad Complutense, 28040 Madrid, Spain.

E-mail presenting author: [seba.carrasco.m@gmail.com](mailto:seba.carrasco.m@gmail.com)

The excited states of single-electron molecules, such as the  $H_2^+$ , experience bond hardening under strong electric fields when the molecular axis is aligned with the field vector. However, the equilibrium ultimately probes unstable and the bond-hardening brief, as the molecular axis rotates to a direction perpendicular to the field vector at the slightest perturbation. This effect leads to dissociation for the  $H_2^+$  molecule. In this work, we show analytically through a simplified model that a strong circularly polarized field could dynamically stabilize the excited state of the  $H_2^+$  molecule. To corroborate if that holds in a full quantum description and to what degree, we numerically characterize the excited electronic state dressed by the field and analyze its dependence on the bond length and angle and the stability of its vibrorotational eigenstates. We conclude that dynamics is metastable; most of the population remains trapped in this excited state for hundreds of femtoseconds, allowing quantum control.



**Figure 1.** Schematic comparing the dynamics under a linearly polarized field and a circularly polarized field. In the first case, the molecule anti-aligns and dissociates; in the second, the molecule never is anti-align for too long and almost always experiences bond-hardening.

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# P14-The role of symmetry in the photophysics of fused nanographene-metalloporphyrin conjugates

S. Garcia-Orrit,<sup>a</sup> V. Vega-Mayoral,<sup>a</sup> Q. Chen,<sup>b</sup> G. Serra,<sup>c</sup> G. M. Paternò,<sup>d</sup> E. Cánovas,<sup>a</sup> A. Narita,<sup>b,e</sup> K. Müllen,<sup>b,f</sup> M. Tommasini<sup>c\*</sup> and J. Cabanillas-González<sup>a\*</sup>

<sup>a</sup> Madrid Institute for Advanced Studies, IMDEA Nanociencia, c/Faraday 9, Campus de Cantoblanco, Madrid 28049, Spain.

<sup>b</sup> Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany.

<sup>c</sup> Dipartimento di Chimica, Materiali e Ing. Chimica "G. Natta", Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy.

<sup>d</sup> Physics Department, Politecnico di Milano, Piazza Leonardo Da Vinci, 32, 20133 Milano, Italy.

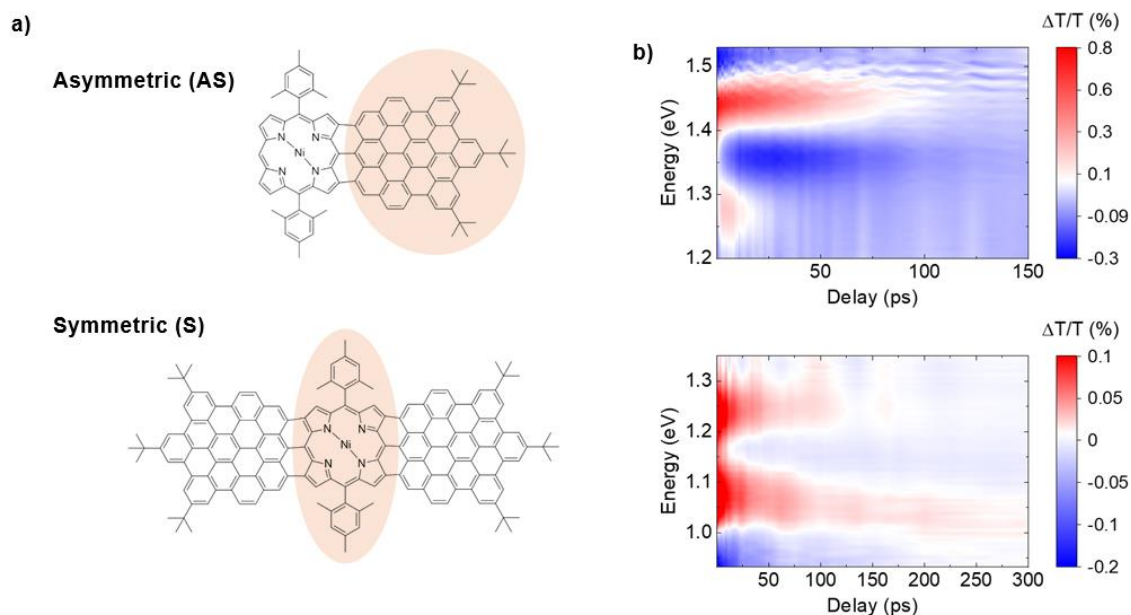
<sup>e</sup> Organic and Carbon Nanomaterials Unit, Okinawa Institute of Science and Technology Graduate University, Okinawa 904-0495, Japan.

<sup>f</sup> Institute for Physical Chemistry, Johannes Gutenberg University Mainz, Duesbergweg 10-14, 55128 Mainz, Germany.

E-mail presenting author: [saul.garcia@imdea.org](mailto:saul.garcia@imdea.org)

Femtosecond optical transient absorption spectroscopy is employed to study, for the first time, photoexcited carrier relaxation in  $\beta$ -meso-,  $\beta$ -triplyfused porphyrin-nanographene conjugates AS and S.<sup>1</sup> These compounds are panchromatic nanographenes (NGs) with an onset of absorption matching the solar spectrum, making these compounds suitable candidate for high efficiency solar cells.

Symmetry has a profound effect of the excited state pathways in these molecules. The symmetric structure, (S orange region Fig. 1a), depicts a strong picosecond excited state funnelling towards the Ni-Porphyrin centre, which preserves an electronic metal centred character, in contrast with the larger interplay of the nanographene on the absorption spectrum. The asymmetric structure instead (AS orange region Fig. 1a), displays excited-state among the fused conjugated ribbon, allowing the appearance of stimulated emission and photoluminescence at 955 nm (1.3 eV) as well as an overall faster decay kinetics than in S. Hence, the substitution symmetry can be used as a versatile tool for tuning the opto-electronic properties of these NGs.



**Figure 1.** a) Structures of triply fused porphyrin-nanographene conjugates AS and S [1], the orange regions indicate the deexcitation regions in each molecule. b) Differential transmission signal obtained for conjugates AS and S after pumping with a 778 nm laser and 605 nm respectively.

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# P15-Ti:Sapphire ultra-precise FoM measurement

V. Miller<sup>a,b</sup> and K. Židek<sup>c</sup>

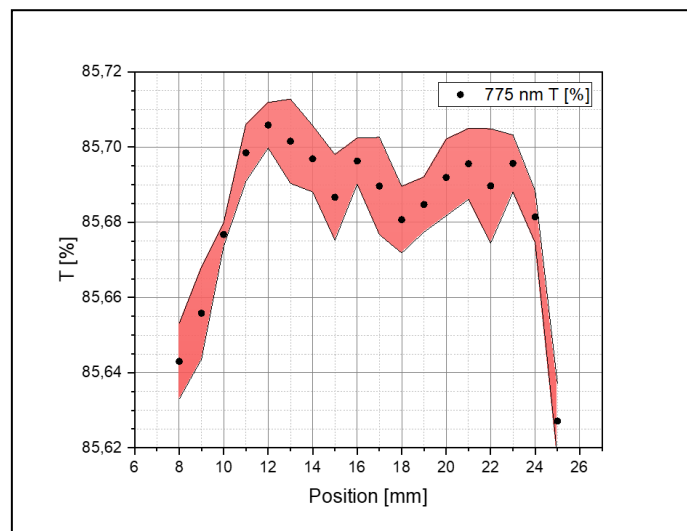
<sup>a</sup> Technical University of Liberec, Studentská 1402/2 461 17 Liberec 1, Liberec, Czech Republic.

<sup>b</sup> CRYTUR, spol. s r. o., Na Lukách 2283, 511 01 Turnov, Czech Republic.

<sup>c</sup> TOPTEC, Institute of Plasma Physics of the Czech Academy of Sciences, Za Slovankou 1782/3, 182 00 Prague 8, Czech Republic.

E-mail presenting author: [vojtech.miller@tul.cz](mailto:vojtech.miller@tul.cz)

Ti:Sapphire (Ti:Sa) is an exceptional material used as a tunable infrared (IR) lasing medium. The advantages of Ti:Sa are mainly in the tunability of the lasing elements in the exceptionally large range of 600–1100 nm. The large lasing range is a fundamental requirement for using Ti:Sa lasers to generate ultra-short pulses (femtosecond scale). The evaluation of the Ti:Sa material quality is done through a parameter denoted as the figure of merit (FoM). The FoM is defined as the ratio of absorption in the pumping spectral region (typically 514 or 532 nm) divided by a residual absorption in the lasing spectral region (typically 800±50 nm). FoM measurements have been reported in a number of articles from several groups over the world.<sup>1,2</sup> However, none of the reports has discussed the actual precision of FoM measurements. The FoM values depend on various factors, including crystal inhomogeneity, volume defects of any kind (bubbles, fog, impurities), and rotation of the crystal c-axis with respect to the electric field of the incident light. Precise determination of FoM is currently carried out on the laser element in a laser oscillator, which is costly and time-consuming. Therefore, it is highly interesting to evaluate FoM with a high precision on a bulk Ti:Sa crystal. In this contribution, we present a setup for a single-pass high-precision transmission measurement on three different wavelengths (532 nm, 775 nm, and 1550 nm) based on Nd:YAG and Er:YAG lasers. A synchronous detection via a double integrated sphere enabled us to gain high-precision measurement, reaching the level of 0,01 %. With the presented setup, we show that it is possible to determine FoM with the precision of ±5 % while the uncertainties introduced in the commonly used experimental devices limit the precision up to ±60 %. We will also demonstrate the experimental data for real-life samples of Ti:Sa crystals produced by a commercial manufacturer (Crytur). Our measurements demonstrate the effect of crystal homogeneity (see Figure 1) and other crucial parameters on the FoM precision. Our contribution will provide a new reliable method for the evaluation of laser material prior to manufacturing the elements.



**Figure 1.** Example of a local scan of transmittance through Ti:Sa sample on 775 nm wavelength (perpendicular propagation).

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# P16-Tracking photoinduced structural dynamics in metal nanoparticles with ultrashort X-rays

J. F. Hidalgo,<sup>a</sup> A. Burgos Caminal,<sup>a</sup> V. Manzaneda-Gonzalez,<sup>b</sup> A. Guerrero-Martínez<sup>b</sup> and W. Gawelda<sup>a,c</sup>

<sup>a</sup>IMDEA Nanociencia, 28049 Madrid, Spain

<sup>b</sup> Departamento de Química Física, Facultad de Ciencias Químicas, Universidad Complutense de Madrid, 28040 Madrid Spain.

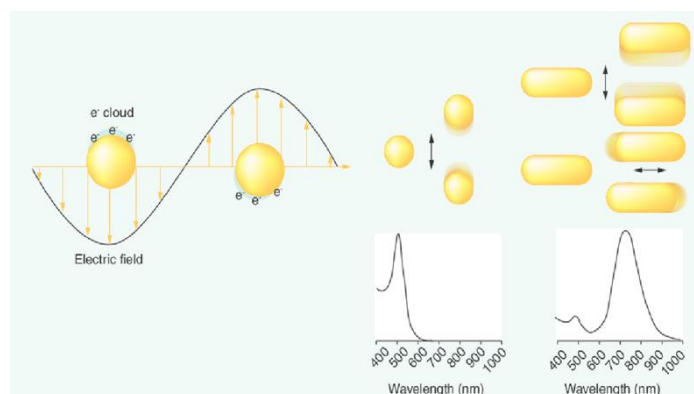
<sup>c</sup> Departamento de Química, Facultad de Ciencias, Universidad Autónoma de Madrid, 28049 Madrid, Spain.

E-mail presenting author: [juan.hidalgo@imdea.org](mailto:juan.hidalgo@imdea.org)

Plasmonic metal nanoparticles (NPs) have been studied for many years, due to their conduction electrons oscillation generated when an electromagnetic field interacts with them. This process, called plasmon resonance,<sup>1</sup> is surface dependent, so that NPs with different shapes do not exhibit this resonance at the same wavelength. Gold nanorods have two plasmon resonances, one of them generated across the longitudinal direction and the other one across the transversal direction. Upon interaction with ultrafast laser pulse, nanorods can undergo melting, which leads to subsequent re-crystallization with substantially different shapes, i.e., as gold nanospheres.<sup>2</sup>

The purpose of our research is to implement structure-sensitive techniques using ultrashort X-ray pulses to track the NP shape variation in time during the melting process. So far, ultrafast laser techniques have succeeded to capture the shapes of the NPs before (nanorod) and after (nanosphere) the laser irradiation, but at this moment there is no real-time structural information about the intermediate changes during that process, except for theoretical models employing Molecular Dynamics simulations.<sup>3</sup>

To order to elucidate all the details of the undergoing structural dynamics we need to employ time-resolved structure-sensitive methods. The time-resolved X-ray scattering<sup>3</sup> can give us information about the previously mentioned process with sufficient temporal resolution to track the fastest process. Hence, time-resolved beamlines at synchrotrons or XFELs are the best facilities to investigate the proposed changes in nanoparticle's shape in real time. The aim is to record the X-ray diffraction of the gold nanorods in a short time interval with the laser off, and at later times to register the photoinduced changes in the diffraction patterns for time delays up to many nanoseconds.



**Figure 1.** Localized surface plasmon resonance effect representation of a gold nanosphere and a gold nanorod.<sup>4</sup>

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# P17-Transient absorption facility at SCAI of the University of Málaga: fs/ps pump-probe spectroscopy

J.C. Otero,<sup>a</sup> M. Moreno,<sup>a</sup> R. Ponce,<sup>a</sup> S. Valdivia,<sup>a</sup> C. Ruano<sup>a,b</sup> and J. Román-Pérez<sup>a,b</sup>

<sup>a</sup> Department of Physical Chemistry, Faculty of Science University of Málaga, Campus de Teatinos S/N, E-29071, Málaga, Spain.

<sup>b</sup> Servicios Centrales de Apoyo a la Investigación (SCAI), Universidad de Málaga, Campus de Teatinos S/N, E-29071 Málaga, Spain.

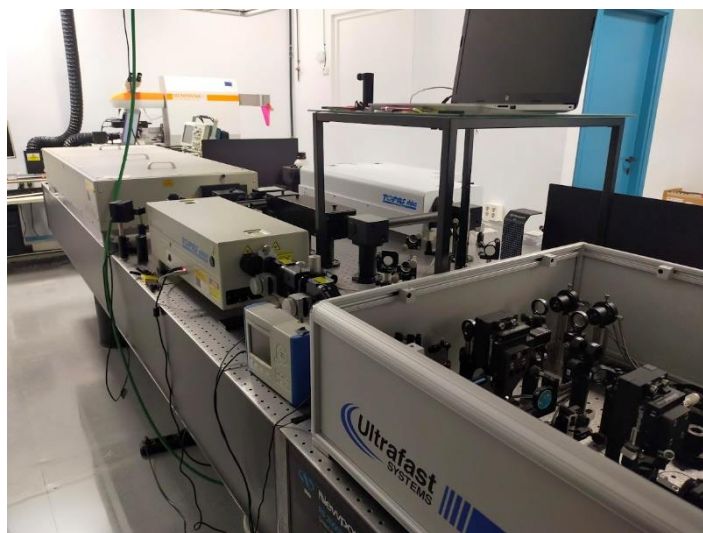
E-mail presenting author: [jcotero@uma.es](mailto:jcotero@uma.es)

Transient absorption spectroscopy (TAS) is a nonlinear spectroscopy for the study of electronically excited species or photogenerated products with a short lifetime. Excited states are probed by monitoring their specific absorption, a method that causes very limited damaged of the system.<sup>1</sup>

The Non-Linear Microscopy and Spectroscopy Unit is a public laser facility at the University of Málaga (SCAI) open to institutional or private users.<sup>2</sup> For TAS experiments a regenerative Ti: Sapphire amplifier (Spitfire Ace, Spectra Physics, 1kHz) that provides 100 fs high power pulses at 800 nm is required. This system is seeded by a fs laser system to provide the input pulses and a pump laser to energize the output. Figure 1 shows a scheme of the transient absorption system. The regenerative amplifier is seeded using a Ti: Sapphire femtosecond laser (MaiTai SP, Spectra Physics, 80 MHz) and pumped by a diode-pumped Q-switched laser of Nd: YLF (Empower 45, Spectra Physics, 1 kHz).

The amplified radiation (800nm, 100fs, 5W) is split for pumping a tunable optical parametric amplifier (TOPAS prime, Spectra Physics) in the range 290-1600 nm for the excitation of the sample and for generating white light for recording the absorption spectrum of the transient species in a HELIOS spectrometer (Ultrafast). The pump and probe beams are overlapped in the sample and the time delay between them is adjusted. The pump beam is chopped to allow for the subtraction of signals arising from the probe beam solely.

Moreover, the setup is also able to convert the fs regime of the amplifier to tunable picoseconds pulses by means of a compressor (Second Harmonic Bandwidth Compressor, SHBC, Light Conversion) coupled to the corresponding OPA (TOPASps-400WL, Light Conversion).



**Figure 1.** Transient Absorption System of the SCAI.

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# P18-Ultrafast chiral imaging with photoelectron vortices

A. Ordóñez,<sup>\*a</sup> X. Barcons-Planas,<sup>b</sup> M. Lewenstein<sup>a,b</sup> and A. Maxwell<sup>a,c</sup>

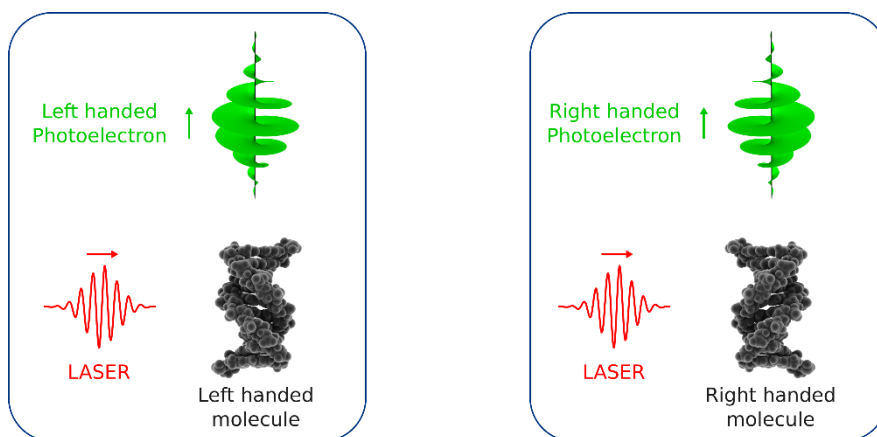
<sup>a</sup> ICFO-Institut de Ciències Fòniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels (Barcelona), Spain.

<sup>b</sup> ICREA, Pg. Lluís Companys 23, 08010 Barcelona, Spain.

<sup>c</sup> Department of Physics and Astronomy, Aarhus University, DK-8000 Aarhus C, Denmark.

E-mail presenting author: [andres.ordonez@icfo.eu](mailto:andres.ordonez@icfo.eu)

Molecular chirality plays a decisive role in determining the outcome of molecular reactions, plays a fundamental role in biological systems, and has immense importance for the chemical industry. The investigation of the interaction between intense short pulses of light and chiral matter has unveiled an array of highly enantio-sensitive phenomena,<sup>1</sup> among which photoelectron techniques have received much attention.<sup>2</sup> Simultaneously, there is an ongoing effort aimed at generating and characterizing electron vortices-electron waves with a helical phase front carrying orbital angular momentum.<sup>3</sup> Electron vortices are expected to provide fundamentally new ways to image chiral matter but their study has been mostly limited to electron transmission microscopy and their potential for ultrafast imaging remains largely unexplored.<sup>4</sup> Here we provide what is to our knowledge the first example of how to exploit photoelectron vortices for the purposes of ultrafast imaging of molecular chirality. Namely, we predict that an intense, linearly polarized, few-cycle, infrared pulse can project the chirality of the molecule onto the phase of the photoelectron wavepacket resulting from the strong-field ionization event, yielding a photoelectron vortex whose chirality is determined by the molecular chirality.<sup>5</sup> We support our prediction with accurate TDSE simulations in a model chiral system.



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# P19-Ultrafast laser facilities for non-linear microscopy/spectroscopy at SCAI of the University of Málaga

J. C. Otero,<sup>a</sup> F.J. Avila Ferrer,<sup>a</sup> F. García-González,<sup>a</sup> C. López Moreno,<sup>a</sup> C. Ruano<sup>a,b</sup> and J. Román-Pérez<sup>a,b</sup>

<sup>a</sup> Department of Physical Chemistry, Faculty of Science University of Málaga, Campus de Teatinos S/N, E-29071, Málaga, Spain.

<sup>b</sup> Servicios Centrales de Apoyo a la Investigación (SCAI), Universidad de Málaga, Campus de Teatinos S/N, E-29071 Málaga, Spain.

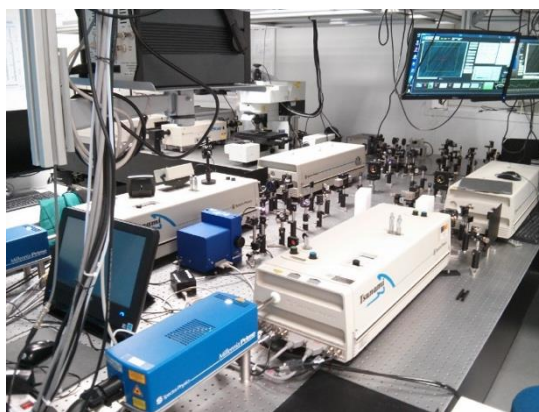
E-mail presenting author: [jcotero@uma.es](mailto:jcotero@uma.es)

The University of Málaga (SCAI) offers a Non-Linear Microscopy and Spectroscopy Unit as a laser-based open facility.<sup>1</sup> This installation began in 2013 and it is based on several femtosecond and picosecond lasers sources that allow for different experimental setups like pump-probe excitation and non-linear spectroscopy or microscopy.

Concerning microscopy applications, the Unit is able to register confocal and multiphoton fluorescence or Raman-based images up to four-photon processes involved in CARS (Coherent Antistokes Raman Spectroscopy).

Confocal microscopy is recorded by exciting with one of the five available lines of 405, 488, 514, 552 and 638 nm, while multiphoton non-linear fluorescence is excited using a dual-output laser source (InSight X3, Spectra Physics) and a Spectral Focusing Timing and Recombination Unit (SF-TRU, Newport) specifically designed for recombining and overlapping dual-output laser sources. This system provides two beams, one fixed at 1045 nm and a tunable one in the 680-1300 nm range. It is possible to switch between fs and chirped ps regime and between single and dual, collinear beam outputs for various research applications such as multiphoton microscopy and multimodal imaging including SHG or THG.

Besides these two conventional types of scanning microscopy, it is possible to record CARS images of unlabelled samples, without the need of prior treatments, by means of the non-linear Raman response corresponding to a selected vibrational wavenumber.<sup>2</sup> In order to have vibrational resolution, two independent laser lines of 3 to 10 picoseconds (tunable oscillators of Ti: sapphire in the 700–1080 nm range, Tsunami model, pumped by continuous Nd: YVO4 lasers at 532 nm (Millenia, Spectra Physics)) are spatially and temporally overlapped in order to induce the non-linear CARS signal when the energy difference between both excitations matches the selected vibrational wavenumber. Both beams can be doubled or tripled for other Raman or fluorescence processes and are optically coupled to an inverted optical microscope (Leica SP8) or to spectrometers.



**Figure 1.** Pulsed laser lines for CARS Microscopy

## REFERENCES

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# LASER MICROMACHINING

The development of femtosecond lasers has transformed the micromachining technology. It enables machining of thin, transparent and semi-transparent materials with high precision and quality at high speed. Femtosecond lasers offer a reliable method for producing cuts, holes, and scribes in brittle materials.



The ORIGAMI XP is the first all-in-one, single-box, microjoule femtosecond laser on the market. The laser head, controller and air-cooling system are all integrated in one small and robust package, with a footprint so small it even fits into a hand-luggage!

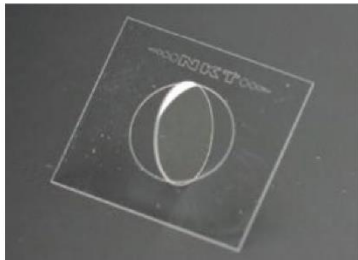


## OF THIN GLASS

Our results demonstrate that femtosecond pulses, delivered by NKT Photonics' ultrafast lasers, are well suited for micro-machining of thin glass.

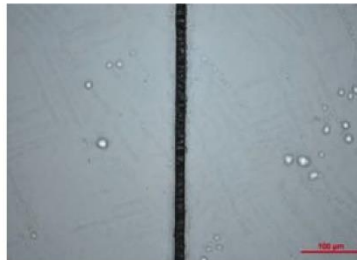
Ultra-short pulses and very high peak powers

Thin glass is widely used in photonics, microelectronics, displays, and biomedical chip manufacturing and has created a need for a reliable high-yield high-quality glass machining process.



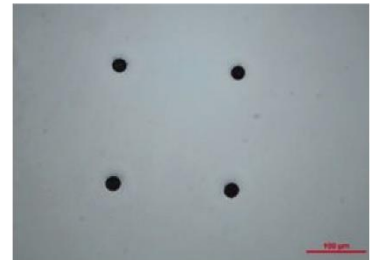
### Glass cutting

In contrast to machining achieved by internal modification of glass, known as "stealth" laser machining, the ablation process is overall faster and provides more flexibility for cutting various closed shapes, such as circles.



### Glass scribing

The high growth in the manufacturing of thin, flexible displays for mobile devices has driven investment into new thin glass cutting and dicing techniques. In one such technique, known as scribe and break, glass is scribed using lasers. Glass scribed using ultrafast lasers provides a more consistent and predictable breaking process with straighter-cut edges and higher yield.



### Glass drilling

Lasers provide a non-contact and clean hole drilling technology that allows drilling of small holes by percussion as well as trepanning technique.

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## OF THIN PMMA FILM

Our results show that our ORIGAMI XP femtosecond laser is well suited to machine PMMA films. We have seen that using infrared wavelengths to process PMMA films yields much better quality than a long-pulse laser CO2 laser. A green wavelength laser can improve quality even further.

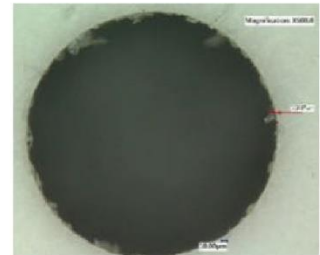
**Precise machining of thin materials calls for ultrafast, high-power lasers.** PMMA is also known as acrylic or acrylic glass. It is a transparent, ultraviolet (UV) light, and scratch-resistant, lightweight, rigid thermoplastic material with an excellent optical transmission. PMMA is a tough plastic that exhibits glass-like qualities at half the weight and with up to 10 times the impact resistance. It is a great alternative to the high cost glass and is widely used as a shatterproof replacement for glass.

High peak-powers remove material instantly

Femtosecond lasers provide a cold ablation micromachining process due to their high peak-power. The high peak-power breaks down the material almost instantly and removes it without affecting the surrounding material.

**Ultrafast pulses give a high precision.**

Ultrafast lasers have delivered high-quality solutions to similar challenges in the automotive, aerospace, and semiconductor industries. These lasers provide a precise, non-contact micromachining processing technique with a minimal need for post-processing.

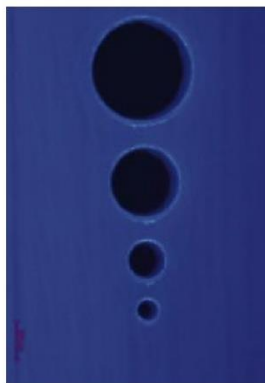


## LASER CUTTING OF POLYMER TUBES FOR BIOMEDICAL APPLICATIONS

It is straightforward to cut polymer tubes with ultrafast lasers. With high peak-power ultrafast lasers, you can make high-quality fine features without any thermal effects. Polyimides are particularly known for their mechanical performance and their chemical resistance.

### Cut and mark in one process

Ultrafast lasers can process a wide variety of polymers, regardless of transparency or thickness. You can use the same laser to cut fine features and to produce permanent, high-quality marks. This way, you simplify manufacturing since one laser can do more than one process. The high-quality cutting and marking eliminate the need for post-processing and reduce the overall manufacturing cost.



### Cold ablation ensures precise, debris-free features

The duration of laser pulses has a dramatic effect on laser cutting results. When the pulse-duration is below some tens of picoseconds, the laser-material interaction enters the "cold" or "athermal" ablation regime. The cutting quality significantly improves by breaking down the material instantly in a precise manner, while the surrounding material is unaffected.



The ultrashort pulses delivered by our ultrafast laser are well suited to precision cutting of polymer tubes.



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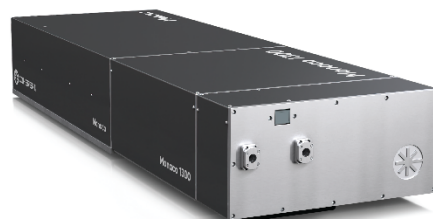


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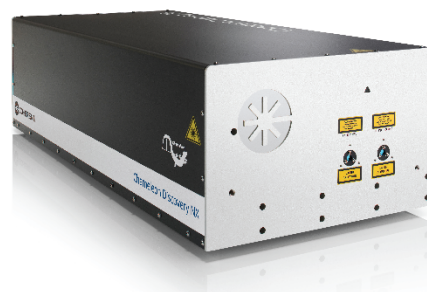


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## Wednesday 16<sup>th</sup>

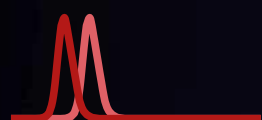
11:00	CHECK-IN
12:00	OPENING
<b>SESSION 1-F. MARTÍN</b>	
12:10	PL 1-A. L'HUILLIER
12:55	IL 1-R. MONTERO
13:20	LUNCH
<b>SESSION 2-R. DE NALDA</b>	
15:00	KL 1-A. PALACIOS
15:35	IL 2-L. PLAJA
16:00	OC 1-L. BAÑARES
16:15	OC 2-E. PALACINO
16:30	COFFEE BREAK
<b>SESSION 3-L. PLAJA</b>	
17:00	IL 3- W. GAWELDA
17:25	IL 4-P. PUYUELO
17:50	OC 3-A. ORDÓÑEZ
18:05	OC 4-S. GANGULY
18:20	POSTER SESSION I

## Thursday 17<sup>th</sup>

<b>SESSION 4-J. SIEGEL</b>	
9:00	PL 2-K. SOKOLOWSKI-TINTEN
9:45	IL 5-J. TRULL
10:10	IL 6-A. JOHNSON
10:35	IL 7-I. SOLÁ
10:50	COFFEE BREAK
<b>SESSION 5-C. COJOCARU</b>	
11:35	IL 8-G. GONZÁLEZ
12:00	IL 9-B. COHEN
12:25	IL 10-J. CABANILLAS
12:50	OC 5-M. BALLABIO
13:05	OC 6-J. M. MARÍN-BELOQUI
13:20	LUNCH
<b>SESSION 6-W. GAWELDA</b>	
15:00	KL 2-R. BORREGO
15:35	IL 11-A. J. ALEJO
16:00	OC 7-I. SOLA
16:15	OC 8- A. MARTÍN-JIMÉNEZ
16:30	COFFEE BREAK
<b>SESSION 7-A. PALACIOS</b>	
17:00	IL 12-M. GARCÍA-LECHUGA
17:25	IL 13-G. MÍNGUEZ
17:50	OC 9-A. BURGOS
18:05	OC 10-O. PEÑA
18:20	POSTER SESSION II
20:30	CONFERENCE DINNER

## Friday 18<sup>th</sup>

<b>SESSION 8-L. BAÑARES</b>	
9:00	PL 3-S. PASCARELLI
9:45	KL 3-K. ATTENKOFER
10:20	IL 14-G. FIGUEIRA
10:45	COFFEE BREAK
<b>SESSION 9-A. LONGARTE</b>	
11:15	IL 15-M. IZQUIERDO
11:40	IL 16-A. CURZIO
12:05	OC 11-R. DE NALDA
12:20	OC 12-M. RICO
12:35	CL-L. ROSO
13:20	CLOSURE



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