# ABSTRACTS

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# Broadband spectral characterization of the phase shift induced by population inversion in Ti:Sapphire

**R. S. NAGYMIHÁLY, H. CAO, P. JÓJÁRT, V. ZUBA, R. FLENDER, O. ANTIPOV, I. SERES, Á. BÖRZSÖNYI, V. CHVYKOV, K. OSVAY, and M. KALASHNIKOV**

**Optics Express**
**27 (2019) 1226-1235**

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The spectral phase shift of broadband amplified pulses, induced by population inversion, was measured in Ti:Sapphire at different pump fluence values. The measurement was performed for two orthogonal polarization directions and at two different crystal temperatures of 296 K and 30 K. Zero shifts and sign changes were observed in the spectral phase, which are connected to the gain spectrum of the crystal. The electronic refractive index changes were also numerically calculated by the Kramers-Kronig theory. The results are highly important for achieving sub-10 fs pulse duration and phase stability in the next generation of Ti:Sapphire-based laser systems.

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# Comparative study of an ultrafast, CEP-stable dual-channel mid-IR OPCPA system

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**JOSA B**
**36 (2019) 12, 3538-3546**

https://doi.org/10.1364/JOSAB.36.003538

In this theoretical work, the performances of ultrafast, carrier-envelope-phase-stable (CEP), mid-IR optical parametric chirped pulse amplification (OPCPA) systems were compared in two different amplification schemes. In the “idler scheme” the mid-IR pulse (idler) is produced in the first difference frequency generation stage of the OPCPA chain and then amplified in the following stages. In the “signal scheme,” the supercontinuum seed is amplified in the OPCPA chain and the mid-IR pulse is generated in the last amplifier stage. According to our results, the idler has higher energy and better energy stability in the idler scheme, while in the signal scheme the signal has the better characteristics in energy and stability. The CEP noise due to the pump intensity fluctuations was found to be lower in the signal scheme. Additionally, chirp optimization revealed that the peak power of the compressed idler and signal pulses at the output of the system is considerably higher if the chirp of the input signal pulse is positive.

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# Single-shot CEP drift measurement at arbitrary repetition rate based on dispersive Fourier transform

**M. KURUCZ, SZ. TÓTH, R. FLENDER, L. HAIZER, B. KISS, B. PERSIELLE, and E. CORMIER**

**Optics Express**
**27 (2019) 13387**

https://doi.org/10.1364/OE.27.013387

This paper presents a single-shot technique for measuring CEP. The Temporal dispersion based One-shot Ultrafast Carrier envelope phase Analysis method (TOUCAN) is an arbitrary repetition rate single-shot CEP drift measurement technique based on dispersive Fourier transformations and has been experimentally tested at 100 kHz. TOUCAN was validated by a direct comparison of decimated data with an independent traditional CEP drift measurement technique. The impact of a temporal jitter on the CEP drift measurement is investigated and a new mitigation technique is shown to produce high accuracy jitter-free CEP drift extraction.

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# Multipass-cell-based post-compression of radially and azimuthally polarized pulses to the sub-two-cycle regime

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**JOSA B**
**36 (2019) 9, 2517-2525**

https://doi.org/10.1364/JOSAB.36.002517

Nonlinear spectral broadening of radially and azimuthally polarized pulses by using the multipass cell technique is proposed and investigated with numerical simulations. The scheme features post-compression of optical pulses from 30 fs down to sub-5 fs centered around 800 nm wavelength with negligible beam distortion and high spectral broadening homogeneity. The performance of the multipass cell is studied with solid-state and gaseous media at μJ and mJ energy levels, respectively. The simulations show that dispersion management in the cell is crucial to obtain sub-two-cycle pulses, while dispersive coatings are considered. It is also shown that multipass cells with silver mirrors provide an alternative option that eases the requirement for dispersion control with an acceptable sacrifice on the net transmission. The sub-two-cycle radially/azimuthally polarized laser pulses are of great interest for generation of radially/azimuthally polarized isolated attosecond extreme ultraviolet pulses and for novel schemes of electron acceleration.

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# Cross thin slab kW-class Ti:Sapphire amplifiers

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Laser Phys.
29 (2019) 065802

https://doi.org/10.1088/1555-6611/ab174c

A scheme for cross thin slab (XTS) amplification is proposed, based on Ti:Sapphire (Ti:Sa), which reaches multi-mJ level ultrashort pulses with a kW average power. The laser crystal is pumped and cooled on two large surfaces to ensure uniform pump energy distribution and a high cooling efficiency. The extraction of energy is realized by multiple passes of seed pulses propagating in the plane perpendicular to the direction of pumping. The amplification and cooling performance of the XTS configuration was numerically simulated. Direct blue diode pumping was also considered to provide upscaling possibilities in the average power to the multiple kW regime with a high extraction efficiency.

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# Generation of few-cycle laser pulses with high temporal contrast via nonlinear elliptical polarisation rotation in a hollow fibre compressor

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**Laser Phys. Lett.**
**16 (2019) 095001**

https://doi.org/10.1088/1612-202X/ab3852

We report on the simultaneous temporal filtering and compression of 30 fs laser pulses down to 5 fs duration via nonlinear elliptical polarization rotation (NER) in a stretched flexible hollow fibre compressor. Both the achieved 2-cycle pulse duration and the internal conversion efficiency of ~47% are solely limited by the applied fibre geometry. Our investigations show that precise control of the NER process could help significantly improve the temporal fidelity of high-energy hollow fibre compression schemes.

eli attosecond

# Generation of spectrally unmodulated and broad pulses through non-linear propagation with elliptical polarization

Viktor PAJER

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Optics Communications 453 (2019) 124393

https://doi.org/10.1016/j.optcom.2019.124393

A fiber-based spectral broadening technique is reported which improves the temporal quality of ultrashort pulses by generating a wide, smooth spectrum from a sub-picosecond laser. The experimental results are confirmed by numerical simulations. It is demonstrated that the interplay between third order nonlinear effects such as cross-phase modulation and nonlinear elliptical polarization rotation have an important role in birefringent fibers and this is an appealing alternative to highly modulated spectrum caused by self-phase modulation. The theoretical model predicts the efficient generation of smooth, wide spectra with compensable phase and shows that proper polarization control is also crucial. An additional model is proposed which allows the determination of the proper set of parameters for the highest efficiency.

János CSONTOS

# Propagation effects in the characterization of 1.5-cycle pulses by XPW dispersion scan

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J. Sel. Top. Quant. Electr.
25 (2019 ) 4

https://
doi.org/10.1109/
JSTQE.2018.2867442

Few-cycle pulse characterization methods face a serious challenge in providing sufficient signal-to-noise ratios together with superior spectral fidelity, as imposed by phase-matching conditions and linear dispersion effects. Here we investigate the effect of linear dispersion inside the nonlinear medium inherently present in such arrangements. We demonstrate that pulse characterization using a cross-polarized wave generation dispersion scan is surprisingly insensitive to the group-velocity dispersion itself. We characterize sub-4 fs pulses at 780 nm center the wavelength utilizing crystals of different thicknesses, yielding nearly identical pulse shapes. Numerical simulations shed light on this behavior indicating practical limits of usable medium lengths.

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# Generation of three-cycle multi-millijoule laser pulses at 318 W average power

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**Optica**
**6 (2019) 1423-1424**

https://doi.org/10.1364/OPTICA.6.001423

The generation of three-cycle multi-millijoule pulses at 318 W power is reported by compressing pulses of a Yb-fiber chirped pulse amplifier in a 6 m long stretched flexible hollow fiber. This technique brings high-power lasers to the few-cycle regime.

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# Theoretical investigation of terahertz generation from two-color laser pulse ionized gases: the role of the thickness of the nonlinear crystal

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Proc. SPIE
11026 (2019) 110260I

https://doi.org/10.1117/12.2520406

We report on the numerical investigation of the generation of THz short pulses by two-color laser pulses in ionized gas plasma in ambient air. One of the major aspects of this study is to find the ideal conditions of second harmonic pulse generation in a suitable nonlinear crystal, where the generated THz pulse is the most intense at given input pulse parameters. We found that an optimal thickness can be found depending on the input pulse parameters, which is defined by two opposing phenomena, the frequency conversion and the linear dispersion. On one hand, a thick crystal can generate energetic second harmonic, but group velocity mismatch spoils the temporal overlap, while on the other hand, a thin crystal does not have enough conversion efficiency. The optimal thickness of one of the most common nonlinear material, BBO was investigated between 1 µm and 500 µm in regards two of the major laser pulse parameters, the pulse duration from 25 fs up to 100 fs and the fluence from 1 $Jcm^{-2}$ up to 5 $Jcm^{-2}$. Our investigation concluded that the optimal thickness increases with the pulse duration and decreases with the fluence.

# THz generation from mid-infrared two-color laser pulses in air and a simple method for controlling the THz intensity

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Proc. SPIE
11026 (2019) 1102611

https://doi.org/10.1117/12.2520408

In this work, we investigated the generation of THz short pulses by two-color mid-infrared laser pulses induced gas plasma in ambient air by numerical analysis. In the simulation, the central wavelength of the input laser varied from 2.5 µm up to 4.0 µm. Our result indicates that the generated THz pulse intensity increases at longer wavelengths significantly. In our simulations, the fundamental pulse intensity and duration were kept fixed yielding no significant difference between the generated electron densities indicating that the asymmetry of the electric field has a major role in the THz generation efficiency by increasing the velocity of electrons. Our results also show that the fundamental pulse and the THz pulse can spectrally overlap, which makes it difficult to separate them spectrally. The possibility of the relative phase control between the fundamental and the second harmonic pulses with a single plate is also examined. Our calculations show that the best materials are the fluorides for controlling the relative phase.

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# Control of THz field waveform emitted from air plasma by chirping two-color laser pulses

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Optics Communications
436 (2019) 222-226

https://
doi.org/10.1016/
j.optcom.2018.12.020

Few-cycle terahertz pulses can be effectively generated in air plasma by mixing an ultrashort laser pulse with its second harmonic. The temporal overlap between these two pulses is critical, which can degrade with the mismatch of group velocities. However, the overlap between the fundamental and second harmonic can be controlled by pre-stretching the temporally separated pulses. In this paper, we study the role of chirp and the relative phase between the fundamental and its second harmonic in the formation of the terahertz waveform. We demonstrate experimentally and explain within the context of transient photocurrent model, that the combination of chirping and relative phase shifting can provide a powerful tool to control the waveform of the terahertz pulse.

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# Single-shot surface ablation and transient reflectivity changes of optical glasses induced by 34 fs laser pulses

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**Proc. SPIE**
**11034 (2019) 110340T**

https://doi.org/10.1117/12.2523006

Results of a comparative study on single-shot surface ablation of commercial optical glasses together with the transient reflectivity enhancement during the process are reported. Three types of optical glasses: Schott's BOROFLOAT®, BK7 and B270 are ablated by single pulses of 34 fs duration at 800 nm central wavelength of the TeWaTi laser systems at University of Szeged, varying systematically both the pulse energy and the beam diameter on the surface, while recording the reflected signal. The depth and diameter of the ablated holes are characterized ex-situ by a DEKTAK profilometer. Very similar ablation characteristics have been determined: Above the ablation thresholds at 5.84±0.21 $Jcm^{-2}$ (1.72±0.06*$10^{14}$ $Wcm^{-2}$), 6.43±0.56 $Jcm^{-2}$ (1.89±0.16*$10^{14}$ $Wcm^{-2}$) and 5.86±0.31 $Jcm^{-2}$ (1.75±0.09*$10^{14}$ Wcm-2) for BOROFLOAT®, BK7 and B270, respectively, both the diameter and the depth of the holes produced show logarithmic increase as a function of pulse energy/fluence until saturating above ~18 $Jcm^{-2}$. On the contrary, significant differences have been obtained in the time integrated transient reflectivities, with the highest absolute values measured for the BOROFLOAT® glass. Strong spot size dependence has been revealed: The reflectivity increases monotonously with increasing pulse energy for all spot sizes, with decreasing absolute values/slopes with decreasing spot areas. Different reflectivities belong to the same fluence/intensity depending on the actual spot size, consequently the fluence/intensity alone does not define unambigously the characteristics of the plasma. The correct description of the changes in reflectivity requires the specification of the spot size together with the pulse energy/fluence/intensity.

# Reconstruction of attosecond pulses in the presence of interfering dressing fields using a 100kHz laser system at ELI-ALPS

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**J. Phys. B.**
**52 (2019) 23LT01**

https://doi.org/10.1088/1361-6455/ab486c

Attosecond Pulse Trains (APT) generated by high-harmonic generation (HHG) of high-intensity near-infrared (IR) laser pulses have proven valuable for studying the electronic dynamics of atomic and molecular species. However, the high intensities required for high-photon-energy, high-flux HHG usually limit the class of adequate laser systems to repetition rates below 10 kHz. Here, APT's generated from the 100 kHz, 160 W, 40 fs laser system (HR-1) currently under commissioning at the extreme light infrastructure attosecond light pulse source (ELI-ALPS) are reconstructed using the reconstruction of attosecond beating by interference of two-photon Transitions (RABBIT) technique. These experiments constitute the first attosecond time-resolved photoelectron spectroscopy measurements with attosecond pulses performed at 100 kHz repetition rate and one of the first experiments performed at ELI-ALPS in the framework of projects commissioning its newly installed technologies. These RABBIT measurements were taken with an additional IR field temporally locked to the extreme-ultraviolet APT, resulting in an atypical ω beating. We show that the phase of the 2ω beating recorded under these conditions is strictly identical to that observed in standard RABBIT measurements within second-order perturbation theory. This work highlights an experimental simplification for future experiments based on attosecond interferometry (or RABBIT), which is particularly useful when lasers with high average powers are used.

Balázs MAJOR
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Valér TOSA
Katalin VARJÚ

# Effect of plasma-core induced self-guiding on phase matching of high-order harmonic generation in gases

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**JOSA B**
**36 (2019), 1594-1601**

https://doi.org/10.1364/JOSAB.36.001594

In this work, we numerically study a self-guiding process in which ionization plays a dominant role and analyze its effect on high-order harmonic generation (HHG) in gases. Although this type of self-guiding—termed "plasma-core-induced self-guiding" in previous works—limits the achievable cutoff by regulating the intensity of the laser beam, it provides favorable conditions for phase matching, which is indispensable for high-flux-gas high-harmonic sources. To underline the role of self-guiding in efficient HHG, we investigate the time-dependent phase-matching conditions in the guided beam and show how the spatiotemporally constant fundamental intensity contributes to the constructive buildup of the harmonic field in a broad photon energy range up to the provided cutoff.

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# Multi-parameter optimization of a loose focusing high flux high-harmonic beamline

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**J. Phys. B**
**52 (2019) 055402**

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We perform a multi-dimensional parameter scan in the generation of high-order harmonics, with the main purpose to find the macroscopic conditions that optimize the harmonic yield in a specific spectral domain, around 40 eV for this particular case. The scanned parameters are the laser pulse energy, gas pressure, interaction cell position relative to focus and the cell length, while the fixed parameters are chosen to model a loose focusing configuration which is used in many existing laboratories. We performed the simulations with a 3D non-adiabatic model complemented by a detailed analysis of the phase matching mechanisms involved in an efficient harmonic generation. Based on the results we identify a range of parameter combinations that lead to a high yield in the specified spectral domain. The method and results presented here can be the framework for the design and construction of high flux high-order harmonic generation beamlines.

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# Single-shot extreme-ultraviolet wavefront measurements of high-order harmonics

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**Optics Express**
**27 (2019) 2656-2670**

https://doi.org/10.1364/OE.27.002656

We perform wavefront measurements of high-order harmonics using an extreme-ultraviolet (XUV) Hartmann sensor and study how their spatial properties vary with different generation parameters, such as pressure in the nonlinear medium, fundamental pulse energy and duration as well as beam size. In some conditions, excellent wavefront quality (up to λ/11) was obtained. The high throughput of the intense XUV beamline at the Lund Laser Centre allows us to perform single-shot measurements of both the full harmonic beam generated in argon and individual harmonics selected by multilayer mirrors. We theoretically analyze the relationship between the spatial properties of the fundamental and those of the generated high-order harmonics, thus gaining insight into the fundamental mechanisms involved in high-order harmonic generation (HHG).

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# Double-pulse characterization by self-referenced spectral interferometry

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**Appl. Phys. Lett. 115 (2019) 051106**

https://doi.org/10.1063/1.5089959

The reconstruction of ultrashort optical pulses with a complex intensity substructure is demonstrated using the Self-Referenced Spectral Interferometry (SRSI) pulse characterization technique with a modified phase retrieval algorithm. A correction spectral phase term is extracted by the manipulation of the temporal interferogram, allowing the treatment of scenarios with complicated pulse shapes, where the original algorithm fails. The improved SRSI algorithm is verified through the application on two temporally well-separated pulses having the same polarization direction and spectral shape, generated by duplicating 37 fs-long amplified pulses of a Ti:Sa based laser system. The spectral phase of highly chirped double pulses with equal or different amplitude ratios is numerically retrieved. The collinear and achromatic experimental arrangement results in a compact and easy-to-align system.

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# A detailed investigation of single-photon laser enabled Auger decay in neon

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New J. Phys.
21 (2019) 113036

https://doi.org/10.1088/1367-2630/ab520d

Single-photon laser enabled Auger decay (spLEAD) is an electronic de-excitation process which was recently predicted and observed in Ne. We have investigated it using bichromatic phase-locked free electron laser radiation and extensive angle-resolved photoelectron measurements, supported by a detailed theoretical model. We first used separately the fundamental wavelength resonant with the $Ne^+$ 2s–2p transition, 46.17 nm, and its second harmonic, 23.08 nm, then their phase-locked bichromatic combination. In the latter case the phase difference between the two wavelengths was scanned, and interference effects were observed, confirming that the spLEAD process was occurring. The detailed theoretical model we developed qualitatively predicts all observations: branching ratios between the final Auger states, their amplitudes of oscillation as a function of phase, the phase lag between the oscillations of different final states, and partial cancellation of the oscillations under certain conditions

# Complete dispersion characterization of microstructured optical fibers using windowed Fourier-transform spectral interferometry

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Attila P. KOVÁCS

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Proc. SPIE
11029 (2019) 110290Q

https://doi.org/10.1117/12.2520571

Dispersion measurements on a birefringent hollow-core (HC-800-02) and a solid-core (LMA-PM-5) photonic crystal fiber (PCF) are presented using a windowed Fourier-transform (WFT) spectral interferometric method. We investigate the optimal value of the spectral window function of the WFT method to reach the highest accuracy in the dispersion measurement. This requires the knowledge of the precise position of the polarization axes of the fibers. In order to determine the position of the polarization axes we have developed a method based on analyzing the WFT signals, which were obtained from a series of interferograms at different excitation ratios of the polarization modes of the PCFs.

Subhendu KAHALY

# Towards intense isolated attosecond pulses from relativistic surface high harmonics

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Optica
6 (2019) 280-287

https://doi.org/10.1364/OPTICA.6.000280

Relativistic surface high harmonics have been considered a unique source for the generation of intense isolated attosecond pulses in the extreme ultra-violet and x-ray spectral ranges. Their practical realization, however, is still a challenging task and requires identification of optimum experimental conditions and parameters. Here, we present measurements and particle-in-cell simulations to determine the optimum values for the most important parameters. In particular, we investigate the dependence of harmonics efficiency, divergence, and beam quality on the pre-plasma scale length as well as identify the optimum conditions for generation of isolated attosecond pulses by measuring the dependence of the harmonics spectrum on the carrier–envelope phase of the driving infrared field.

Subhendu KAHALY
Dimitris CHARALAMBIDIS
Paraskevas TZALLAS

# Imaging the source of high-harmonics generated in atomic gas media

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Optics Express
27 (2019) 9733-9739

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We report the application of the time gated ion microscopy technique in accessing online the position of the source of harmonics generated in atomic gas media. This is achieved by mapping the spatial extreme-ultraviolet (XUV)-intensity distribution of the harmonic source onto a spatial ion distribution, produced in a separate focal volume of the generated XUV beam through single photon ionization of atoms. It is found that the position of the harmonic source depends on the relative position of the harmonic generation gas medium and the focus of the driving infrared (IR) beam. In particular, by translating the gas medium with respect to the IR beam focus different “virtual” source positions are obtained online. Access to such online source positioning allows better control and provides increased possibilities in experiments where selection of electron trajectory is important. The present study gives also access to quantitative information which is connected to the divergence, the coherence properties and the photon flux of the harmonics. Finally, it constitutes a precise direct method for providing complementary experimental info to different attosecond metrology techniques.

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# Superior photo-thermionic electron emission from illuminated phosphorene surface

**S. MADAS, S. K. MISHRA, S. KAHALY, and M. UPADHYAY KAHALY**

**Scientific Reports**
**9 (2019) 10307**

https://doi.org/10.1038/s41598-019-44823-x

This work demonstrates that black phosphorene, a two dimensional allotrope of phosphorus, has the potential to be an efficient photo-thermionic emitter. To investigate and understand the novel aspects we use a combined approach in which *ab initio* quantum simulation tools are utilized along with semiclassical description for the emission process. First by using density functional theory based formalism, we study the band structure of phosphorene. From the locations of electronic bands, and band edges, we estimate the Fermi level and work function. This leads us to define a valid material specific parameter space and establish a formalism for estimating thermionic electron emission current from phosphorene. Finally we demonstrate how the emission current can be enhanced substantially under the effect of photon irradiation. We observe that photoemission flux to strongly dominate over its coexisting counterpart thermionic emission flux. Anisotropy in phosphorene structure plays important role in enhancing the flux. The approach which is valid over a much wider range of parameters is successfully tested against recently performed experiments in a different context. The results open up a new possibility for application of phosphorene based thermionic and photo-thermionic energy converters.

Sergei KÜHN
Mathieu DUMRGUE
Péter FÖLDI
Subhendu KAHALY
Eric CORMIER
Bálint KISS
Katalin VARJÚ
Sándor VARRÓ
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# Quantum optical signatures in a strong laser pulse after interaction with semiconductors

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Electrodynamical processes induced in complex systems like semiconductors by strong electromagnetic fields have traditionally been described using semiclassical approaches. Although these approaches allowed the investigation of ultrafast dynamics in solids culminating in multipetahertz electronics, they do not provide any access to the quantum-optical nature of the interaction, as they treat the driving field classically and unaffected by the interaction. Here, using a full quantum-optical approach, we demonstrate that the subcycle electronic response in a strongly driven semiconductor crystal is imprinted in the quantum state of the driving field resulting in nonclassical light states carrying the information of the interaction. This vital step towards strong-field ultrafast quantum electrodynamics unravels information inaccessible by conventional approaches and leads to the development of a new class of nonclassical light sources.

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# Saddle point approaches in strong field physics and generation of attosecond pulses

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Physics Reports
833 (2019) 1-52

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Attoscience is the emerging field that accesses the fastest electronic processes occurring at the atomic and molecular length scales with attosecond (1 as = 10−18 s) time resolution having wide ranging physical, chemical, material science and biological applications. The quintessential and one of the most fundamental processes in this domain is the generation of phase locked XUV attosecond pulses. The theoretical approach to understand the process incorporates a fully quantum or semi classical or relativistic description of coherent charge dynamics in intense ultrashort electromagnetic fields driving a quantum system (an atom, a molecule, solid band gap materials or surface plasmas). Modelling of such physical and dynamical systems in science and also in many other branches often leads to equations represented in terms of complex multi-dimensional integrals. These integrals can often be solved using the stationary phase approximation, which leads to a series of equations identifying the points in the multi-dimensional space, having most significant contributions in their evaluation. These points are usually indicated as saddle points. The description of the dynamics of quantum mechanical or relativistic systems that results from such an approach enables near to classical physics intuitive perceptions of the processes under investigation. Thus, the saddle point methods are very powerful and valuable general theoretical tools to obtain asymptotic expressions of such solutions and help also to gain physical insights on the underlying phenomena. Such techniques developed in the past have been adapted to study the emission of as pulses by different physical systems and have been widely employed in calculating and estimating the response of matter to intense electromagnetic pulses on ultrafast time scales. Here we provide an extensive disposition of the saddle point approaches unifying their ubiquitous applications within the domain of attoscience valid for simple atomic to more complex condensed matter systems undergoing ultrafast dynamics and present current trends and advancements in the field. In this review we would delineate the methodology, present a synthesis of seminal works and describe the state of the art applications. Finally we also address ultrashort time dynamics of novel materials that have gained much attention recently, namely lower dimensional material systems and micro-plasma systems.

# Numerical investigation of imaging-free terahertz generation setup using segmented tilted-pulse-front excitation

József A. FÜLÖP

GY. TÓTH, L. PÁLFALVI, J. A. FÜLÖP, G. KRIZSÁN, N. MATLIS, G. ALMÁSI, and J. HEBLING

Optics Express
27 (2019) 5, 7762-7775

https://doi.org/10.1364/OE.27.007762

Recently a hybrid-type terahertz (THz) pulse source was proposed for high energy terahertz pulse generation. It is the combination of the conventional tilted-pulse-front setup and a nonlinear crystal with a transmission stair-step echelon of period in the hundred-micrometer range etched into the front face. The tilt angle introduced by the conventional tilted-pulse-front setup (pre-tilt) was chosen to be equal to the tilt-angle needed inside the nonlinear crystal (62° for lithium niobate (LN)) in order to fulfill velocity-matching. In this case, plane-parallel nonlinear optical crystals can be used. The possibility of using a plane-parallel nonlinear optical crystal for producing good-quality, symmetric THz beams was considered the most important advantage of this setup. In the present paper, a thorough numerical investigation of a modified version of that setup is presented. In the new version, the tilted pulse-front is created by a transmission grating without any imaging optics, and a wedged nonlinear optical crystal with a small wedge angle is supposed. According to a 1D numerical code, significantly higher THz generation efficiency can be achieved with a transmission stair-step echelon-faced nonlinear crystal having a $5^{-15}$-degree wedge angle than with a plane-parallel one or with the conventional tilted-pulse-front setup. Because of the spatially-dependent group-delay dispersion introduced by the transmission grating, a small wedge in the nonlinear crystal improves the spatial homogeneity of the THz-generation process, resulting in higher efficiencies and better beam profiles. At 100 K temperature, and by using 800 nm pump pulses with 20 mJ pulse energy, 100 fs pulse length and 8 mm beam spot radius, approximately 4.5% conversion efficiency and close to 1 mJ terahertz pulse energy can be reached with the newly-proposed setup.

Song LI
Christos KAMPERIDIS
Nasr A. M. HAFZ

# A laser-plasma accelerator driven by two-color relativistic femtosecond laser pulses

**S. LI, G. LI, Q. AIN, M. S. HUR, A. C. TING, V. V. KULAGIN, CH. KAMPERIDIS, and N. A. M. HAFZ**

**Science Advances**
**5 (2019), 11 eaav7940**

https://doi.org/10.1126/sciadv.aav7940

A typical laser-plasma accelerator (LPA) is driven by a single, ultrarelativistic laser pulse from terawatt- or petawatt-class lasers. Recently, there has been some theoretical work on the use of copropagating two-color laser pulses (CTLP) for LPA research. Here, we demonstrate the first LPA driven by CTLP where we observed substantial electron energy enhancements. Those results have been further confirmed in a practical application, where the electrons are used in a bremsstrahlung-based positron generation configuration, which led to a considerable boost in the positron energy as well. Numerical simulations suggest that the trailing second harmonic relativistic laser pulse is capable of sustaining the acceleration structure for much longer distances after the preceding fundamental pulse is depleted in the plasma. Therefore, our work confirms the merits of driving LPAs by two-color pulses and paves the way toward a downsizing of LPAs, making their potential applications in science and technology extremely attractive and affordable.

PaTHz

József A. FÜLÖP

# Laser-driven strong-field terahertz sources

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Adv. Optical Mater.
8 (2019) 3, 1900681

https://doi.org/10.1002/adom.201900681

A review on the recent development of intense laser-driven terahertz (THz) sources is provided here. The technologies discussed include various types of sources based on optical rectification (OR), spintronic emitters, and laser-filament-induced plasma. The emphasis is on OR using pump pulses with tilted intensity front. Illustrative examples of newly emerging applications are briefly discussed, in particular strong-field THz control of materials and acceleration and manipulation of charged particles.

eli attosecond

PaTHz

Abhishek GUPTA

# A polarization-resolved study of nanopatterned photoconductive antenna for enhanced Terahertz emission

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IEEE Trans. THz Sci. Tech. 9 (2019) 2, 193-199

https://doi.org/10.1109/TTHZ.2019.2891022

Terahertz (THz) frequencies, despite having the potential for several important applications, have been relatively underexplored in the past owing to the unavailability of proper sources and detectors. The scenario has been changing over the past few decades due to the advent of convenient THz sources and detectors. THz photoconductive antennas (PCA), due to their attractive features, such as cost effectiveness and room temperature operation, are playing a key role in current and future research prospect in the field of THz spectroscopy, both as sources and detectors. Complex PCA designs have been proposed and studied to boost the THz emission efficiencies. Elucidating the underlying physics in such devices requires a thorough investigation of a few physical parameters. This requires the integration of several experimental techniques under identical conditions. In this paper, we show such a study, including a parametric variation of pump polarization, conducted on a PCA with a nanopatterned active region, which boosts the emitted THz radiation. Through the set of measurements, we unravel the subtle interplay of the various physical processes responsible for the emission of THz radiation from the device.

eli attosecond

# Demonstration of a tilted-pulse-front pumped plane-parallel slab terahertz source

József A. FÜLÖP

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Optics Letters
44 (2019) 4, 1023

https://doi.org/10.1364/OL.44.001023

A new type of tilted-pulse-front pumped terahertz (THz) source has been demonstrated, which is based on a LiNbO3LiNbO3plane-parallel slab with an echelon structure on its input surface. Single-cycle pulses of 1 μJ energy and 0.30 THz central frequency have been generated with $5\times10^{-4}$ efficiency from such a source. One order-of-magnitude increase in efficiency is expected by pumping a cryogenically cooled echelon of increased size and thickness with a Ti:sapphire laser. The use of a plane-parallel nonlinear optical crystal slab enables straightforward scaling to high THz pulse energies and the production of a symmetric THz beam with a uniform pulse shape for good focusability and high field strength.

# High energy proton micro-bunches from a laser plasma accelerator

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Scientific Reports
9 (2019) 13840

https://doi.org/10.1038/s41598-019-50348-0

Recent advances on laser-driven ion accelerators have sparked an increased interest in such energetic particle sources, particularly towards the viability of their usage in a breadth of applications, such as high energy physics and medical applications. Here, we identify a new ion acceleration mechanism and we demonstrate, via particle-in-cell simulations, for the first time the generation of high energy, monochromatic proton micro-bunches while witnessing the acceleration and self-modulation of the accelerated proton beam in a dual-gas target, consisting of mixed ion species. In the proposed ion acceleration mechanism due to the interaction of an ultra-short, ultra-intense (2 PW, 20 fs) laser pulses with near-critical-density partially ionized plasmas (C & H species), we numerically observed high energy monochromatic proton microbunches of high quality (peak proton energy 350 MeV, laser to proton conversion efficiency ~$10^{-4}$ and angular divergence <10 degree), which can be of high relevance for medical applications. We envisage that through this scheme, the range of attained energies and the monochromaticity of the accelerated protons can be increased with existing laser facilities or allow for laser-driven ion acceleration investigations to be pursued at moderate energies in smaller scale laser laboratories, hence reducing the size of the accelerators. The use of mixed-gas targets will enable high repetition rate operation of these accelerators, free of plasma debris and electromagnetic pulse disruptions.

Dániel PAPP

# Investigation of wire-array Z-pinches by laser probing diagnostics

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Matter Radiat. Extremes
4 (2019) 017401

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Laser diagnostics provides powerful tools for the investigation of dense Z-pinches. In this paper, wire-array Z-pinches are investigated at the 1 MA Zebra generator using laser diagnostics at different wavelengths coupled with x-ray diagnostics. Plasma dynamics during the ablation, implosion, and stagnation stages are observed by multiframe diagnostics. Cascading and nonprecursor implosions are studied in wire arrays. Ultraviolet diagnostics allows deep penetration into the Z-pinch plasma at stagnation. End-on probing reveals the complicated structure of the precursor. Strong magnetohydrodynamic instabilities are found in a dense pinch hidden in the trailing plasma. Small-scale instabilities are seen in the Z-pinch plasma with micrometer resolution. Probing of the pinch from four directions shows asymmetrical trailing plasma in some configurations of wire arrays. Faraday rotation diagnostics reveals the magnetic fields and the current distribution in the plasma of the precursor and Z-pinch. Redistribution of current in the trailing plasma is seen during kink and sausage instabilities in the stagnation stage. The formation of micropinches and hot spots in the Z-pinch is analyzed with coupled laser and x-ray diagnostics. Different laser diagnostics allow the study of Z-pinch plasmas in all stages, including fast dynamics and instabilities.

# Terahertz electric field modulated mode coupling in graphene-metal hybrid metamaterials

József A. FÜLÖP

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Optics Express
27 (2019) 3, 2317-2326

https://doi.org/10.1364/OE.27.002317

Taking advantage of the tunable conductivity of graphene under high terahertz (THz) electric field, a graphene-metal hybrid metamaterial consisting of an array of three adjoined orthogonally oriented split-ring resonators (SRRs) is proposed and experimentally demonstrated to show a maximum modulation depth of 23% in transmission when the THz peak field reaches 305 kV/cm. The transmission of the sample is dominated by the antisymmetric and symmetric resonant modes originating from the strong magneto-inductive and conductive coupling among the three SRRs, respectively. Numerical simulations and model calculations based on a coupled oscillator theory were performed to explain the modulation process. It is found that the graphene coating impairs the resonances by increasing the damping of the modes and decreasing the coupling between the SRRs whereas the strong THz field restores the resonances by decreasing the conductivity of graphene.

PaTHz

Nasr A. M. HAFZ

# Ultrafast dynamics of magnetic vortices and pulse collapse in a laser-under dense plasma interaction

G. LI, S. LI, Q. AIN, K. GAO, M. MIRZAIE, and N. A. M. HAFZ

Phys. Plasmas
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https://doi.org/10.1063/1.5053636

The energy of an intense ultrashort laser pulse interacting with high density (still under dense) plasma is typically transformed into electron heating and in excitation of nonlinear coherent structures such as magnetic vortices, solitons, or post-solitons. Using 33 TW 30 fs laser pulses and a high-density nitrogen gas jet, we experimentally investigated magnetic vortices in the laser pulse collapse region where electrons are efficiently accelerated and heated. Those vortices, which are associated with rapidly decaying magnetic fields, are found to be immobile and persist for several picoseconds. A collisionless plasma was formed due to the quasi-static field ionization of the gas associated with the hot and fast electron currents. The evolution dynamics of such nonlinear plasma phenomena have been monitored by using a 30-fs probe laser beam through employing the polarimetric and shadowgraphic techniques. Our experimental results are also supported by particle-in-cell simulations.

eli attosecond

SA

Károly TŐKÉSI

# Relaxation dynamics of charge patches formed inside an insulating capillary by ion impact

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Nucl. Inst. and Meth. In Phys. Res.
460 (2019) 234-239

https://doi.org/10.1016/j.nimb.2018.12.027

The dynamics of the charge distribution produced at the inner surface of an insulating capillary by low energy ion impact are investigated theoretically in the case where the entrance, exit and outer capillary surface are grounded. Starting with the surface continuity equation, that describes the charge dynamics at the inner surface, we deduced an analytical solution of the continuity equation in the form of a linear combination of surface charge moments that satisfy the given boundary conditions. We determined the relaxation rate of each moment, which is given as a function of the dimensions and electrical properties of the capillary. We found an approximate expression of the time evolution of the electric potential and field generated by a charge patch during and after beam irradiation. The time evolution of the total charge is also described in detail. Our findings are illustrated with examples.

eli attosecond

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# Optoelectronic properties of CuI photoelectrodes

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J. Phys. Chem. Lett.
10 (2019) 259–264

https://
doi.org/10.1021/
acs.jpclett.8b03242

Detailed mechanistic understanding of the optoelectronic features is a key factor in designing efficient and stable photoelectrodes. In situ spectroelectrochemical methods were employed to scrutinize the effect of trap states on the optical and electronic properties of CuI photoelectrodes and to assess their stability against (photo) electrochemical corrosion. The excitonic band in the absorption spectrum and the Raman spectral features were directly influenced by the applied bias potential. These spectral changes exhibit a good correlation with the alterations observed in the charge-transfer resistance. Interestingly, the population and depopulation of the trap states, which are responsible for the changes in both the optical and electronic properties, occur in a different potential/energy regime. Although cathodic photocorrosion of CuI is thermodynamically favored, this process is kinetically hindered, thus providing good stability in photoelectrochemical operation.

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# Electrochemical hole injection selectively expels iodide from mixed halide perovskite films

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J. Am. Chem. Soc. 141
(2019) 27, 10812-10820

https://
doi.org/10.1021/
jacs.9b04568

Halide ion mobility in metal halide perovskites remains an intriguing phenomenon, influencing their optical and photovoltaic properties. Selective injection of holes through electrochemical anodic bias has allowed us to probe the effect of hole trapping at iodide (0.9 V) and bromide (1.15 V) in mixed halide perovskite ($CH_3NH_3PbBr_{1.5}I_{1.5}$) films. Upon trapping holes at the iodide site, the iodide gradually gets expelled from the mixed halide film (as iodine and/or triiodide ion), leaving behind re-formed $CH_3NH_3PbBr_3$ domains. The weakening of the Pb–I bond following the hole trapping (oxidation of the iodide site) and its expulsion from the lattice in the form of iodine provided further insight into the photoinduced segregation of halide ions in mixed halide perovskite films. Transient absorption spectroscopy revealed that the iodide expulsion process leaves a defect-rich perovskite lattice behind as charge carrier recombination in the re-formed lattice is greatly accelerated. The selective mobility of iodide species provides insight into the photoinduced phase segregation and its implication in the stable operation of perovskite solar cells.

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# Tuning the excited-state dynamics of CuI films with electrochemical bias

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ACS Energy Lett.
3 (2019) 702-708

https://doi.org/10.1021/acsenergylett.9b00182

Owing to its high hole conductivity and ease of preparation, CuI was among the first inorganic hole-transporting materials that were introduced early on in metal halide perovskite solar cells, but its full potential as a semiconductor material is still to be realized. We have now performed ultrafast spectroelectrochemical experiments on ITO/CuI electrodes to show the effect of applied bias on the excited-state dynamics in CuI. Under operating conditions, the recombination of excitons is dependent on the applied bias, and it can be accelerated by decreasing the potential from +0.6 to −0.1 V vs Ag/AgCl. Prebiasing experiments show the persistent and reversible "memory" effect of electrochemical bias on charge carrier lifetimes. The excitation of CuI in a CuI/$CsPbBr_3$ film provides synergy between both CuI and $CsPbBr_3$ in dictating the charge separation and recombination.

SA

Péter DOMBI

# High harmonic generation on noble gas clusters

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Optics Express
27 (2019) 19 ,26721

https://doi.org/10.1364/OE.27.026721

High order harmonics (HHG) were generated on noble gas cluster targets with different cluster sizes. The independently characterized cluster sources enabled experimental investigation of the recombination mechanism. HHG spectra were recorded for different backing pressures and gases (Ar, Xe) as a function of driver pulse ellipticity. Since the ellipticity-dependent HHG decay is essentially the same for the different gas-pressure pairs, we can conclude that the recombination process is dominated by atom-to-itself recollisions irrespective of cluster size and material.

eli attosecond

Zsuzsanna PÁPA

# Monitoring of evolving laser induced periodic surface structures

A. LÜBCKE, ZS. PÁPA, and M. SCHNÜRER

Appl. Sci.
9 (2019) 3636

https://
doi.org/10.3390/
app9173636

Laser induced periodic surface structures (LIPSS) are generated on titanium and silicon nitride surfaces by multiple femtosecond laser pulses. An optical imaging system is used to observe the backscattered light during the patterning process. A characteristic fringe pattern in the backscattered light is observed and evidences the surface modification. Experiments are complemented by finite difference time domain numerical simulations which clearly show that the periodic surface modulation leads to characteristic modulations in the coherently scattered light field. It is proposed that these characteristic fringe pattern can be used as a very fast and low-cost monitor of LIPSS formation formation during the manufacturing process.

# Simulation of photoelectron emission from metallic nanoparticles under laser irradiation

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Péter DOMBI
Károly TŐKÉSI

L. BUDAI, ZS. MÁRTON, P. DOMBI, and K. TŐKÉSI

Eur. Phys. J. D
73 (2019) 138

https://doi.org/10.1140/epjd/e2019-90686-x

We developed a new computer simulation code that calculates trajectories of photoelectrons emitted from nanoparticles by laser excitation. The code uses the pre-calculated electric field obtained by finite-difference time-domain simulations as input. The photoelectron trajectories emitted from silver nanoparticles were calculated using the classical trajectory Monte Carlo method, where the image force towards the surface is taken into account. We show that our present code is suitable to describe the recent experimental findings reasonably well. Significant effect of the image acceleration to the calculated electron spectra was observed. We found that the calculated energy distributions of photoelectrons are in agreement with the recent experiments.

Károly TŐKÉSI

# Surface and bulk plasmon excitations of silver by electron impact

J. GONG, L. YANG, K. TŐKÉSI, and Z. DING

Eur. Phys. J. D
73 (2019) 24

https://doi.org/10.1140/epjd/e2018-90603-y

We present an analysis of electron incident angle-dependent reflected electron energy loss spectroscopy spectra of silver by using a Monte Carlo simulation method with which the separated contributions from surface and bulk excitations can be identified. The simulations were performed at several different incident electron energies and with various incident angles with respect to the surface normal. We found that the surface plasmon excitation plays a dominant role in the loss peak at around 3.7 and 7.5 eV.

SA

# Determination of electron inelastic mean free path of three transition metals from reflection electron energy loss spectroscopy spectrum measurement data

Károly TŐKÉSI

L. YANG, K. TŐKÉSI, K., B. DA, and Z. DING

Eur. Phys. J. D
73 (2019) 21

https://doi.org/10.1140/epjd/e2018-90551-6

The energy loss functions (ELFs) of three transition metals (Cr, Co and Pd) have been derived from reflection electron energy loss spectroscopy spectrum with a theoretical analysis of the measured data [Xu et al., J. Appl. Phys. 123, 043306 (2018)]. In this work, we update our previous ELFs in a wider photon energy region (0–200 eV) with a better accuracy, which is verified by sum rules and a root-mean-square deviation. The electron inelastic mean free paths (IMFPs) of Cr, Co and Pd have been calculated with the obtained ELFs by adopting a dielectric response theory. We employ both the single-pole approximation and full Penn algorithm for the calculation of IMFPs, and the calculated results are compared with other references.

Károly TŐKÉSI

# Photoelectron holography of atomic targets

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Phys. Rev. A
99 (2019) 013413

https://
doi.org/10.1103/
PhysRevA.99.013413

We study the spatial interference effects appearing during the ionization of atoms (H, He, Ne, and Ar) by few-cycle laser pulses using single-electron ab initio calculations. The spatial interference is the result of the coherent superposition of the electronic wave packets created during one half cycle of the driving field following different spatial paths. This spatial interference pattern may be interpreted as the hologram of the target atom. With the help of a wave-function analysis (splitting) technique and approximate (strong-field and Coulomb-Volkov) calculations, we directly show that the hologram is the result of the electronic-wave-packet scattering on the parent ion. On the He target we demonstrate the usefulness of the wave-function splitting technique in the disentanglement of different interference patterns. Further, by performing calculations for the different targets, we show that the pattern of the hologram does not depend on the angular symmetry of the initial state and it is strongly influenced by the atomic species of the target: A deeper bounding potential leads to a denser pattern.

Károly TŐKÉSI

# Semiclassical two-step model for ionization of the hydrogen molecule by a strong laser field

N. I. SHVETSOV-SHILOVSKI, M. LEIN, and K. TŐKÉSI

Eur. Phys. J. D
73 (2019) 37

https://doi.org/10.1140/epjd/e2018-90527-6

We extend the semiclassical two-step model for strong-field ionization that describes quantum interference and accounts for the Coulomb potential beyond the semiclassical perturbation theory to the hydrogen molecule. In the simplest case of the molecule oriented along the polarization direction of a linearly polarized laser field, we predict significant deviations of the two-dimensional photoelectron momentum distributions and the energy spectra from the case of atomic hydrogen. Specifically, for the hydrogen molecule the electron energy spectrum falls off slower with increasing energy, and the holographic interference fringes are more pronounced than for the hydrogen atom at the same parameters of the laser pulse.

# Elastic electron scattering cross sections for triethyl phosphate molecule at intermediate electron energies from 50 eV to 250 eV

Károly TŐKÉSI

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Eur. Phys. J. D
73 (2019) 27

https://doi.org/10.1140/epjd/e2019-90631-1

We present a combined experimental and theoretical study of the electron elastic differential cross sections of triethyl phosphate molecule (C2H5)3PO4 (TEP). The experimental setup based on a crossed beam technique comprising of an electron gun, a single capillary gas needle and a detection system with a channeltron was used to measure differential cross sections. The absolute scale for the cross sections is obtained by relative-flow method using argon gas as a reference. For the interpretation of the measured data we applied the partial expansion method to calculate the elastic cross sections of TEP. We found excellent agreement between the shapes of measured and calculated data.

# $^{11}$Boron delivery agents for boron proton-capture enhanced proton therapy (BPCEPT)

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E. Rita SZABÓ
Róbert POLANEK
Tünde TŐKÉS
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Anticancer Res.
39 (2019) 5, 2265-2276

https://doi.org/10.21873/anticanres.13343

The aim of this review was to define appropriate $^{11}$B delivery agents for boron proton-capture enhanced proton therapy (BPCEPT) taking into account the accumulated knowledge on boron compounds used for boron neutron capture therapy (BNCT). BPCEPT is a promising treatment approach which uses a high linear energy transfer (LET) dose component in conjunction with conventional proton therapy to increase the relative biological effectiveness of highly-selective charged particle therapy. Boron proton fusion reactions occur with highest cross section at certain proton energy level and thus can be tailored to the target volume with careful treatment planning that defines the 675 MeV proton distribution with high accuracy. Appropriate $^{11}$B compounds are required in order to achieve relevant high LET dose contribution from the boron proton-capture reaction. Previous scientific results and experiences with BNCT provide background knowledge and information regarding the optimization of boronated compound development, their characterization, measurement and imaging. However, there are substantial differences between BNCT and BPCEPT, which in turn places special unique chemical, physical and biological demands on $^{11}$B-carrier compounds for BPCEPT. In this review, we evaluate well-known and recently developed boron compounds for BPCEPT.

Katalin HIDEGHÉTY
E. Rita SZABÓ

# Feasibility of proton FLASH effect tested by zebrafish

**E. BEYREUTHER, M. BRAND, S. HANS, K. HIDEGHÉTY, L. KARSCH, E. LEßMANN, M. SCHÜRER, E. R. SZABÓ, and J. PAWELKE**

Radioth.and Oncology
139 (2019) 46-50

https://
doi.org/10.1016/
j.radonc.2019.06.024

Background and purpose: Motivated by first animal trials showing the normal tissue protecting effect of electron and photon Flash irradiation, i.e. at mean dose rates of 100 Gy/s and higher, relative to conventional beam delivery over minutes the feasibility of proton Flash should be assessed.

Materials and methods: A setup and beam parameter settings for the treatment of zebrafish embryo with proton Flash and proton beams of conventional dose rate were established at the University Proton Therapy Dresden. Zebrafish embryos were treated with graded doses and the differential effect on embryonic survival and the induction of morphological malformations was followed for up to four days after irradiation.

Results: Beam parameters for the realization of proton Flash were set and tested with respect to controlled dose delivery to biological samples. Analyzing the dose dependent embryonic survival and the rate of spinal curvature as one type of developmental abnormality, no significant influence of proton dose rate was revealed. For the rate of pericardial edema as acute radiation effect, a significant difference ($p < 0.05$) between proton Flash and protons delivered at conventional dose rate of 5 Gy/min was observed for one dose point only.

Conclusion: The feasibility of Flash proton irradiation was successfully shown, whereas more experiments are required to confirm the presence or absence of a protecting effect and to figure out the limits and requirements for the Flash effect.

Imre F. BARNA
Károly TŐKÉSI

# Photoionisation of rubidium in strong laser fields

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The photoionisation of rubidium in strong infra-red laser fields based on ab initio calculations was investigated. The bound and the continuum states are described with Slater orbitals and Coulomb wave packets, respectively. The bound state spectra were calculated with the variational method and we found it reproduced the experimental data within a few percent accuracy. Using the similar approach, ionisation of Rb was also successfully investigated. The effects of the shape and the parameters of the pulse to the photoionisation probabilities and the energy spectrum of the ionised electron are shown. These calculations may provide a valuable contribution at the design of laser and plasma based novel accelerators, the CERN AWAKE experiment.

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# Femtosecond wave-packet revivals in ozone

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**Phys. Rev. A**
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Photodissociation of ozone following absorption of biologically harmful solar ultraviolet radiation is the key mechanism for the life protecting properties of the atmospheric ozone layer. Even though ozone photolysis is described successfully by post-Hartree-Fock theory, it has evaded direct experimental access so far, due to the unavailability of intense ultrashort deep ultraviolet radiation sources. The rapidity of ozone photolysis with predicted values of a few tens of femtoseconds renders both ultrashort pump and probe pulses indispensable to capture this manifestation of ultrafast chemistry. Here, we present the observation of femtosecond time-scale electronic and nuclear dynamics of ozone triggered by ~10-fs, ~2-μJ deep ultraviolet pulses and, in contrast to conventional attochemistry experiments, probed by extreme ultraviolet isolated pulses. An electronic wave packet is first created. We follow the splitting of the excited B-state related nuclear wave packet into a path leading to molecular fragmentation and an oscillating path, revolving around the Franck-Condon point with 22-fs wave-packet revival time. Full quantum-mechanical ab initio multiconfigurational time-dependent Hartree simulations support this interpretation.

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# Photodissociation dynamics of the LiF molecule: Two- and three-state descriptions

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The photodissociation of the lithium fluoride molecule gathered much attention lately. Theoretical works treating this subject usually consider the 1 $^1\Sigma^+$ and 2 $^1\Sigma^+$ electronic states, which are coupled nonadiabatically. As a continuation of a previous work, we intend to highlight the importance of the 1 $^1\Pi$ state by investigating the kinetic energy and angular distribution of the photofragments. Besides, our results pointed out the importance of the molecular rotation.

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# Finding intersections between electronic excited potential energy surfaces with simultaneous ultrafast X-ray scattering and spectroscopy

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**Chemical Science**
**10 (2019) 5749-5760**

https://doi.org/10.1039/c8sc04023k

Vacancy-ordered lead-free perovskites with more-stable crystalline structures have been intensively explored as the alternatives for resolving the toxic and long-term stability issues of lead halide perovskites (LHPs). The dispersive energy bands produced by the closely packed halide octahedral sublattice in these perovskites are meanwhile anticipated to facility the mobility of charge carriers. However, these perovskites suffer from unexpectedly poor charge carrier transport. To tackle this issue, we have employed the ultrafast, elemental-specific X-ray transient absorption (XTA) spectroscopy to directly probe the photoexcited electronic and structural dynamics of a prototypical vacancy-ordered lead-free perovskite ($Cs_3Bi_2Br_9$). We have discovered that the photogenerated holes quickly self-trapped at Br centers, simultaneously distorting the local lattice structure, likely forming small polarons in the configuration of $V_k$ center ($Br_2$ - dimer). More significantly, we have found a surprisingly long-lived, structural distorted state with a lifetime of ~59 μs, which is ~3 orders of magnitude slower than that of the charge carrier recombination. Such long-lived structural distortion may produce a transient "background" under continuous light illumination, influencing the charge carrier transport along the lattice framework.

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# Visualizing the coordination-spheres of photoexcited transition metal complexes with ultrafast hard X-rays

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**Phys. Chem. Chem. Phys. 21 (2019) 9277 - 9284**

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The concept of coordination sphere (CS) is central to the rational development of hierarchical molecular assemblies in modern chemistry. Manipulating the organization around transition metal ions with covalent and supramolecular interactions is a general strategy that underlies most synthetic protocols. Achieving similar control for photoexcited molecular complexes is necessary to advance the design of light-driven functionalities. This objective calls for monitoring the ultrafast dynamics of the primary (1-CS) and the secondary (2-CS) coordination spheres on the atomic scale, which remains to date an important experimental challenge for short-lived species. In this work, transient wide-angle scattering of hard X-rays (25 keV) is employed with state-of-the-art AIMD simulations in order to visualize the 1-CS (solute-only) and the 2-CS (solvation cage) of the photoinduced high-spin (HS) state for $[Fe(bpy)_3]^{2+}$ (bpy = 2,2′-bipyridine) in aqueous solution. Correlating this structural information in real-space reveals the interlacing of the two CS, which in turn explains why solvation affects the photoinduced electronic and structural dynamics in this class of complexes. More generally, these results obtained for a prominent prototypical system in ultrafast X-ray sciences demonstrate the unique perspectives offered by this technique to gain the crucial knowledge about the multiscale solvation dynamics that is currently missing for controlling the solute–solvent interactions in advanced functional nano and biomaterials employed for photoconversion.

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# Resonant photoelectron confinement in the $SF_6$ molecule

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J. Phys. Chem. A
123 (2019) 1062

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Recent thorough experimental activity aiming to generate high harmonics in the $SF_6$ molecules requires the knowledge of, on one hand, accurate valence-shell photoionization cross sections and phases, from a threshold up to a few tens of eV, where resonances are likely to appear, and, on the other hand, the effect of the nuclear vibrational dynamics on the process. In this work, we have experimentally determined and theoretically evaluated vibrationally resolved photoionization cross sections of $SF_6$ up to 80 eV photon energies, with an emphasis on the $E^2T_{1u}$ channel, for which vibrational progressions are fully resolved in the experiment. Our results reveal the presence of shape resonances due to excitation to $SF_6$ virtual states lying just above the ionization threshold, in agreement with previous synchrotron radiation work and theoretical calculations. More interestingly, our calculations also disclose resonance features at photoelectron energies as high as 40–50 eV, which are due to the transient confinement of the ejected electron in the octahedral cage formed by the peripheral F atoms. In the vicinity of all resonances, including those due to confinement, the calculated ionization phases experience an excursion of about π or π/2 and significantly depend on the final vibrational state of the remaining cation. Both effects should be taken into account to correctly interpret ongoing high-harmonic generation work in $SF_6$. A similar behavior is expected for other symmetric molecules containing a central atom, such as $BF_3$, $CF_4$, and the like.

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# Resolving the ultrafast changes of chemically-inequivalent metal-ligand bonds in photoexcited molecular complexes with transient X-ray absorption spectroscopy

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Photoactive transition-metal complexes that incorporate heteroleptic ligands present a first coordination shell, which is asymmetric. Although it is generally expected that the metal–ligand bond lengths respond differently to photoexcitation, resolving these fine structural changes remains experimentally challenging, especially for flexible multidentate ligands. In this work, ultrafast X-ray absorption spectroscopy is employed to capture directly the asymmetric elongations of chemically inequivalent metal–ligand bonds in the photoexcited spin-switching $Fe^{II}$ complex $[Fe^{II}(tpen)]^{2+}$ solvated in acetonitrile, where tpen denotes *N*,*N*,*N*′,*N*′-tetrakis(2-pyridylmethyl)-1,2-ethylenediamine. The possibility to correlate precisely the nature of the donor/acceptor coordinating atoms to specific photoinduced structural changes within a binding motif will provide advanced diagnostics for optimizing numerous photoactive chemical and biological building blocks.

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# Asynchronous photoexcited electronic and structural relaxation in lead free perovskites

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Vacancy-ordered lead-free perovskites with more-stable crystalline structures have been intensively explored as the alternatives for resolving the toxic and long-term stability issues of lead halide perovskites (LHPs). The dispersive energy bands produced by the closely packed halide octahedral sublattice in these perovskites are meanwhile anticipated to facility the mobility of charge carriers. However, these perovskites suffer from unexpectedly poor charge carrier transport. To tackle this issue, we have employed the ultrafast, elemental-specific X-ray transient absorption (XTA) spectroscopy to directly probe the photoexcited electronic and structural dynamics of a prototypical vacancy-ordered lead-free perovskite ($Cs_3Bi_2Br_9$). We have discovered that the photogenerated holes quickly self-trapped at Br centers, simultaneously distorting the local lattice structure, likely forming small polarons in the configuration of $V_k$ center ($Br_2^-$ dimer). More significantly, we have found a surprisingly long-lived, structural distorted state with a lifetime of ~59 μs, which is ~3 orders of magnitude slower than that of the charge carrier recombination. Such long-lived structural distortion may produce a transient “background” under continuous light illumination, influencing the charge carrier transport along the lattice framework.

# Minimum requirements for electron–positron pair creation in the interaction of ultra-short laser pulses with thin foils

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Current laser technology allows the production of extremely high laser intensities and brings ever closer experimental probing of quantum electro-dynamic effects e.g. radiation reaction and electron–positron pair creation via the multiphoton Breit–Wheeler process. The exponential dependence of the latter process on laser intensity means that the process appears suddenly above some threshold, which is still not well defined in the case of laser-plasma interactions. The threshold intensity for the generation of a significant number of positrons is shown to be in the order $10^{22}$ W cm$^{-2}$, when optimal target properties, as presented in this paper, are considered. With the help of a modified particle-in-cell code, the detailed angular-energy distribution of positrons is presented, which is in good agreement with our simple analytical model.

# Features of the generation of fast particles from microstructured targets irradiated by high intensity, picosecond laser pulses

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Laser and Particle Beams
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The use of targets with surface structures for laser-driven particle acceleration has potential to significantly boost the particle and radiation energies because of enhanced laser absorption. We investigate, via experiment and particle-in-cell simulations, the impact of micron-scale surface-structured targets on the spectrum of electrons and protons accelerated by a picosecond laser pulse at relativistic intensity. Our results show that, compared with flat-surfaced targets, structures on this scale give rise to a significant enhancement in particle and radiation emission over a wide range of laser–target interaction parameters. This is due to the longer plasma scale length when using micro-structures on the target front surface. We do not observe an increase in the proton cutoff energy with our microstructured targets, and this is due to the large volume of the relief.

# Attosecond bunches of gamma photons and positrons generated in nanostructure targets

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https://
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PhysRevE.99.013202

The subatomic experimental exploration of physical processes on extremely short time scales has become possible by the generation of high-quality electron bunches and x-ray pulses with subfemtosecond durations. Increasing the photon energy from the x-ray to gamma-ray regime makes probing of extremely small space-time domains accessible. Here, a mechanism for generating attosecond gamma photon and positron bunches with small divergence using laser intensities below $10^{23}$W/cm$^2$ is proposed. In contrast with previous works, in our scheme a single laser pulse is sufficient instead of two counterpropagating pulses. Numerical simulations are used to formulate the conditions for confined radiation and to characterize the generated photon and positron bunches.

# Insights into the mechanisms and dynamics of energy transfer in plant light-harvesting complexes from two-dimensional electronic spectroscopy

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BBA Bioenergetics
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During the past two decades, two-dimensional electronic spectroscopy (2DES) and related techniques have emerged as a potent experimental toolset to study the ultrafast elementary steps of photosynthesis. Apart from the highly engaging albeit controversial analysis of the role of quantum coherences in the photosynthetic processes, 2DES has been applied to resolve the dynamics and pathways of energy and electron transport in various light-harvesting antenna systems and reaction centres, providing unsurpassed level of detail. In this paper we discuss the main technical approaches and their applicability for solving specific problems in photosynthesis. We then recount applications of 2DES to study the exciton dynamics in plant and photosynthetic light-harvesting complexes, especially light-harvesting complex II (LHCII) and the fucoxanthin-chlorophyll proteins of diatoms, with emphasis on the types of unique information about such systems that 2DES is capable to deliver.

# Temperature dependence of the energy transfer in LHCII revealed by two-dimensional electronic spectroscopy

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J. Phys. Chem. B.
123 (2019) 31, 6765-6775

https://doi.org/10.1021/acs.jpcb.9b05421

We measured two-dimensional electronic spectra of light-harvesting complex II (LHCII) at various temperatures (77, 110, 150, 230, and 295 K) under conditions free from singlet–singlet annihilation. We elucidated the temperature-dependent excitation energy transfer dynamics in the Chl a manifold of LHCII. Global analysis revealed that the dynamics can be summarized in distinct time scales from 200 fs up to 15 ps. While the fastest dynamics with a decay time of ~0.2–0.3 ps are relatively temperature-independent, the lifetimes and relative contributions of slower components showed considerable temperature dependence. The slowest time scale of equilibration with the lowest-energy Chl a increased from ~5 ps at 295 K to ~15 ps at 77 K. The final excited state is independent of initial excitation at 230 K and above, whereas static energy disorder is apparent at lower temperatures. A clear temperature dependence of uphill energy transfer processes was also discerned, which is consistent with the detailed-balance condition.

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# Anisotropic circular dichroism of light-harvesting complex II in oriented lipid bilayers: theory meets experiment

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**J. Phys. Chem. B**
**123 (2019) 5, 1090-1098**

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Anisotropic circular dichroism (ACD) spectroscopy of macroscopically aligned molecules reveals additional information about their excited states that is lost in the CD of randomly oriented solutions. ACD spectra of light-harvesting complex II (LHCII)—the main peripheral antenna of photosystem II in plants—in oriented lipid bilayers were recorded from the far-UV to the visible wavelength region. ACD spectra show a drastically enhanced magnitude and level of detail compared to the isotropic CD spectra, resolving a greater number of bands and weak optical transitions. Exciton calculations show that the spectral features in the chlorophyll $Q_y$ region are well-reproduced by an existing Hamiltonian for LHCII, providing further evidence for the identity of energy sinks at chlorophylls a603 and a610 in the stromal layer and chlorophylls a604 and a613 in the luminal layer. We propose ACD spectroscopy to be a valuable tool linking the three-dimensional structure and the photophysical properties of pigment–protein complexes.

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# Dependence of chlorophyll fluorescence quenching on the lipid-to-protein ratio in reconstituted light-harvesting complex II membranes containing lipid labels

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Chemical Physics
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The quenching of chlorophyll-a fluorescence was investigated in plant light-harvesting complex II (LHCII) embedded in reconstituted membranes containing thylakoid lipids and a lipid label. The proteoliposomes were further separated by density and the protein and lipid contents of the fractions were quantified spectrophotometrically, allowing tighter control over the L/P ratios in a wide range of values. Using time-resolved fluorescence, we found a strong correlation between the fluorescence quenching and the L/P ratio, in line with other studies reporting progressive quenching at low L/P ratios, presumably triggered by self-clustering in the membrane. The average fluorescence lifetimes decreased to 0.3 ns at L/P ratios below 50:1; these values are comparable to the quenching observed in plants under excess-light conditions and are accompanied by a similar far-red fluorescence signature. It is hypothesized that plants can exploit the intrinsic quenching propensity of LHCII-only membrane domains to safely store extra antenna units.

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# Revealing the excitation energy transfer network of light-harvesting complex II by phenomenological analysis of two-dimensional electronic spectra at 77 K

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**J. Chem. Phys. 151 (2019) 205101**

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Energy equilibration in light-harvesting antenna systems normally occurs before energy is transferred to a reaction center. The equilibration mechanism is a characteristic of the excitation energy transfer (EET) network of the antenna. Characterizing this network is crucial in understanding the first step of photosynthesis. We present our phenomenology-based analysis procedure and results in obtaining the excitonic energy levels, spectral linewidths, and transfer-rate matrix of Light-Harvesting Complex II directly from its 2D electronic spectra recorded at 77 K with waiting times between 100 fs to 100 ps. Due to the restriction of the models and complexity of the system, a unique EET network cannot be constructed. Nevertheless, a recurring pattern of energy transfer with very similar overall time scales between spectral components (excitons) is consistently obtained. The models identify a “bottleneck” state in the 664–668 nm region although with a relatively shorter lifetime (⍰4–6 ps) of this state compared to previous studies. The model also determines three terminal exciton states at 675, 677–678, and 680–681 nm that are weakly coupled to each other. The excitation energy equilibration between the three termini is found to be independent of the initial excitation conditions, which is a crucial design for the light-harvesting complexes to ensure the energy flow under different light conditions and avoid excitation trapping. We proposed two EET schemes with tentative pigment assignments based on the interpretation of the modeling results together with previous structure-based calculations and spectroscopic observables .

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# Size effects in charge migration in alkyne chains

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Theor. Chem. Accounts
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The charge migration dynamics initiated by an outer-valence ionization of alkyne chains containing between 4 and 12 carbon atoms is studied using ab initio methods only. It is shown that the removal of a HOMO–1 electron from each of the molecules in the series leads to the population of the same structure of a main state and correlation satellites in their ionization spectra. The resulting pure electron dynamics manifest as an ultrafast charge migration oscillations between the center of the molecule and its extremities. The charge migration follows the same pattern in all studied cases with a slight monotonic increase of the time scale with the system size. This shows that the correlation-driven charge migration can exhibit scaling properties and constitutes a proof of concept for the possibility of molecular design for charge migration. As alkynes are used as molecular wires, it is argued that the present results may open the door for ultrafast molecular electronics.

# Real-time observation of X-ray-induced intramolecular and interatomic electronic decay in $CH_2I_2$

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Nature Communications
10 (2019) 2186

https://doi.org/10.1038/s41467-019-10060-z

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The increasing availability of X-ray free-electron lasers (XFELs) has catalyzed the development of single-object structural determination and of structural dynamics tracking in real-time. Disentangling the molecular-level reactions triggered by the interaction with an XFEL pulse is a fundamental step towards developing such applications. Here we report real-time observations of XFEL-induced electronic decay via short-lived transient electronic states in the diiodomethane molecule, using a femtosecond near-infrared probe laser. We determine the lifetimes of the transient states populated during the XFEL-induced Auger cascades and find that multiply charged iodine ions are issued from short-lived (~20 fs) transient states, whereas the singly charged ones originate from significantly longer-lived states (~100 fs). We identify the mechanisms behind these different time scales: contrary to the short-lived transient states which relax by molecular Auger decay, the long-lived ones decay by an interatomic Coulombic decay between two iodine atoms, during the molecular fragmentation.

# The role of the time delay in the reflection and transmission of ultrashort electromagnetic pulses on a system of parallel current sheets

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Phys. Scr.
94 (2019) 045205

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The reflection and transmission of a few-cycle laser pulse impinging on two parallel thin metal layers have been analyzed. The two layers, with a thickness much smaller than the skin depth of the incoming radiation field, are represented by current sheets embedded in three dielectrics, all with different index of refraction. The dynamics of the surface currents and the scattered radiation field are described by the coupled system of Maxwell–Lorentz equations. When applying the plane wave modeling assumptions, these reduce to a hybrid system of two delay differential equations for the electron motion in the layers and a recurrence relation for the scattered field. The solution is given as the limit of a singularly perturbed system and the effects of the time delay on the dynamics is analyzed.

# Isolated attosecond pulses of μJ energy via coherent Thomson-backscattering, driven by a chirped laser pulse

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Eur. Phys. J. D
73 (2019) 77

https://doi.org/10.1140/epjd/e2019-90535-0

New theoretical and numerical results are presented regarding isolated attosecond XUV – soft X-ray pulses, that can be generated by Thomson-backscattering of a high-intensity single-cycle near-infrared laser pulse on a suitable nanobunch of MeV electons. A simple approximate formula is derived for the cut-off frequency of the collective radiation spectrum, which is then employed to find the length of the nanobunch which emits an isolated pulse of 16 as length. Detailed analysis of the spectral, temporal and spatial features of this attosecond pulse is given. It is also shown that the 100 nJ pulse energy, corresponding to $2.1 \times 10^{18}$ W/cm$^2$ peak intensity of the laser pulse, can be increased to reach the μJ pulse energy both by increasing the intensity or by setting a suitable down-chirp of the laser pulse.