ABSTRACTS

ELI ALPS

2020

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SYLOS Lasers – the frontier of few-cycle, multi-TW, kHz lasers for ultrafast applications at ELI ALPS

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The extreme light infrastructure attosecond light pulse source offers beamtime for users of various attosecond and particle sources driven by versatile laser systems. Here we report on the state of the art of a few-cycle, multi-TW, 1 kHz repetition rate laser system, now fully operational in the facility. The system is based on four stages of optical parametric amplifiers (OPAs) pumped by a total of 320 mJ, 80 ps frequency-doubled Nd:YAG laser pulses. All OPA stages utilize double crystal configuration, which design has been also confirmed by model calculations. The 1 kHz SYLOS 2 system produces 32 mJ laser pulses around a central wavelength of 891 nm with 6.6 fs (<2.3 optical cycles) pulse duration exceeding the peak power of 4.8 TW on a daily basis. The recorded best pulse duration is 6.3fs, which corresponds to 2.12 cycles and 5.1 TW peak power. During long-term (24h) performance tests, energy stability of 1.2%, carrier-envelope phase (CEP) stability of 210 mrad, and pointing stability of 0.4µrad were demonstrated, while the Strehl ratio of the beam is kept above 0.75. In order to help the alignment of all the different experiments at the facility and to reduce the workload on SYLOS 2 system, a second laser system has been developed. The so-called SYLOS Experimental Alignment (SEA) laser mimicks the performance of the SYLOS 2 laser, but at a repetition rate two orders of magnitude lower and without CEP-stabilization. The three single-crystal OPA stages of the SEA laser provide 42mJ pulse energy for the users, while having energy stability of 0.87% and sub-13 fs pulse duration at a repetition rate ranging from a single shot up to 10 Hz.

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2.3-cycle mid-infrared pulses from hybrid thin-plate post-compression at 7 W average power

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Optics Communications
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j.optcom.2020.126035

Output pulses of a 100 kHz mid-infrared OPCPA system are post-compressed from 4.7 cycles down to 2.3 cycles by using a combination of a dielectric and a semiconductor crystal in a hybrid thin plate setup. Efficient spectral broadening is demonstrated with 11 W average input power. After compression the output power reached 6.8 W with exceptional CEP and energy stability for a several hours. The post-compressed pulses were carefully characterized in both temporal and spatial domains, resulting in 2.3-cycle temporal duration at 3.1 μm central wavelength with a temporal Strehl ratio of 0.73 and a spatial Strehl ratio of 0.97. Thermal limitations due to multiphoton absorption of semiconductors present at this power level are explored by temperature measurements, which are supported by detailed numerical simulations. Upscaling for higher average powers was also investigated.

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Carrier-envelope offset stable, coherently combined ytterbium-doped fiber CPA delivering 1 kW of average power

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Optics Letters
45 (2020) 23,6350-6353

https://doi.org/10.1364/OL.409410

We present a carrier-envelope offset (CEO) stable ytterbium-doped fiber chirped-pulse amplification system employing the technology of coherent beam combining and delivering more than 1 kW of average power at a pulse repetition rate of 80 MHz. The CEO stability of the system is 220 mrad rms, characterized out-of-loop with an f-to-2f interferometer in a frequency offset range of 10 Hz to 20 MHz. The high-power amplification system boosts the average power of the CEO stable oscillator by five orders of magnitude while increasing the phase noise by only 100 mrad. No evidence of CEO noise deterioration due to coherent beam combining is found. Low-frequency CEO fluctuations at the chirped-pulse amplifier are suppressed by a “slow loop” feedback. To the best of our knowledge, this is the first demonstration of a coherently combined laser system delivering an outstanding average power and high CEO stability at the same time.

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Development of a defect recognition algorithm for visual laser-induced damage detection

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Laser Physics
30 (2020) 046002

https://doi.org/10.1088/1555-6611/ab7a24

Laser-induced damage is defined as a permanent detrimental change in the characteristics of an optical element caused by a laser beam. This change can be observed by many different inspection techniques, of which optical and phase imaging microscopic techniques have superior sensitivity. However, such examinations conducted by human operators are relatively slow and subjective—so they cannot be used for online damage monitoring purposes, whereas automatic inspection systems have advantages in terms of sensitivity, reliability, and speed. In this paper we introduce a new method for the computer -aided recognition of damaged sites based on visual images taken from the sample surface by a CCD camera. The evaluation procedure is performed by a computer algorithm, which consists of exact, statistically established steps. It includes noise reduction by considering the statistical behavior of photon noise. Besides, it takes into account the spatial extent of a damage spot by nonlinear image filtering to separate damage-indicating intensity changes from random noise. This mimics the ability of the human eye to distinguish features from their surroundings. The evaluation algorithm is built of computationally less demanding mathematical operations to enable fast execution which is vital for monitoring at high repetition rates. The proposed method was tested on a sizeable dataset of images yielding 98.8% of damage detection efficiency. It was also compared to a generally used visual laser damage detection procedure, which has a success rate of 88.6%. This yields one order of magnitude reduction in the number of undetected damaged sites.

LaSo

High-power ytterbium-doped fiber laser delivering few-cycle, carrier-envelope phase-stable 100 µJ pulses at 100 kHz

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Optics Letters
45 (2020) 1, 97-100

https://doi.org/10.1364/OL.45.000097

We present a carrier-envelope phase (CEP)-stable Yb-doped fiber laser system delivering 100 µJ few-cycle pulses at a repetition rate of 100 kHz. The CEP stability of the system when seeded by a carrier-envelope offset-locked oscillator is 360 mrad, as measured pulse-to-pulse with a stereographic above-threshold ionization (stereo -ATI) phase meter. Slow CEP fluctuations have been suppressed by implementing a feedback loop from the phase meter to the pulse picking acousto-optic modulator. To the best of our knowledge, this is the highest CEP stability achieved to date with a fiber-based, high-power few-cycle laser.

eli attosecond

Objective quantification and spatial mapping of cataract with a Shack-Hartmann wavefront sensor

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Scientific Reports
10 (2020) 12585

https://doi.org/10.1038/s41598-020-69321-3

Based on wavefront sensor images an objective and quantitative method is presented for characterising cataract. By separating direct and scattered light in the focal plane of the microlenses, the new procedure is able to make two-dimensional maps of the spatial variation of scattering properties in the crystalline lens, and also provides a single figure descriptive for the whole eye. The developed evaluation algorithm successfully quantifies cataract, especially that of nuclear type. To demonstrate its operation, a custom-built measurement setup was constructed using a Shack-Hartmann wavefront sensor with 40×3240×32 microlenses to capture 12-bit images of the pupil plane, and a superluminescent diode of 830 nm wavelength as a light source. Slit-lamp clinical measurements served as reference for calibration and to estimate the accuracy of the new method. The tests were carried out on 78 eyes with cataract in different progression state ranging from healthy to above 5 on the LOCS III scale. The residual error of the calibration (i.e. the standard deviation of difference between clinical reference and our algorithmic characterisation) turned out to be ±0.29 category on the LOCS III N scale, which approximates the ±0.33 precision of classic cataract measurements carried out with the greatest care.

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Attosecond delays in photoionization studied with coherent-controlled FEL

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J. Phys.: Conf. Ser.
1412 112006

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When an electron is ejected from an atom after absorption of a photon, the photoelectron wave packet has an extremely short group delay between the photon absorption and the electron emission. This interval, called the Eisenbud-Wigner-Smith delay, is on the order of a few attoseconds. Here, we present a new method to measure the photoemission delay, using coherent-controlled free-electron laser pulses .

eli attosecond

SeSo

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New method for measuring angle-resolved phases in photoemission

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Phys. Rev. X 10 (2020) 031070

https://doi.org/10.1103/PhysRevX.10.031070

Quantum mechanically, photoionization can be fully described by the complex photoionization amplitudes that describe the transition between the ground state and the continuum state. Knowledge of the value of the phase of these amplitudes has been a central interest in photoionization studies and newly developing attosecond science, since the phase can reveal important information about phenomena such as electron correlation. We present a new attosecond-precision interferometric method of angle-resolved measurement for the phase of the photoionization amplitudes, using two phase-locked extreme ultraviolet pulses of frequency ω and 2ω, from a free-electron laser. Phase differences Δ˜η between one- and two-photon ionization channels, averaged over multiple wave packets, are extracted for neon 2p electrons as a function of the emission angle at photoelectron energies 7.9, 10.2, and 16.6 eV. Δ˜η is nearly constant for emission parallel to the electric vector but increases at 10.2 eV for emission perpendicular to the electric vector. We model our observations with both perturbation and ab initio theory and find excellent agreement. In the existing method for attosecond measurement, reconstruction of attosecond beating by interference of two-photon transitions (RABBITT), a phase difference between two-photon pathways involving absorption and emission of an infrared photon is extracted. Our method can be used for extraction of a phase difference between single-photon and two-photon pathways and provides a new tool for attosecond science, which is complementary to RABBITT.

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A 10-gigawatt attosecond source for non-linear XUV optics and XUV-pump-XUV-probe studies

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https://doi.org/10.1038/s41598-020-60331-9

The quantum mechanical motion of electrons and nuclei in systems spatially confined to the molecular dimensions occurs on the sub-femtosecond to the femtosecond timescales respectively. Consequently, the study of ultrafast electronic and, in specific cases, nuclear dynamics requires the availability of light pulses with attosecond (asec) duration and of sufficient intensity to induce two-photon processes, essential for probing the intrinsic system dynamics. The majority of atoms, molecules and solids absorb in the extreme-ultraviolet (XUV) spectral region, in which the synthesis of the required attosecond pulses is feasible. Therefore, the XUV spectral region optimally serves the study of such ultrafast phenomena. Here, we present a detailed review of the first 10-GW class XUV attosecond source based on laser driven high harmonic generation in rare gases. The pulse energy of this source largely exceeds other laser driven attosecond sources and is comparable to the pulse energy of femtosecond Free-Electron-Laser (FEL) XUV sources. The measured pulse duration in the attosecond pulse train is 650 ± 80 asec. The uniqueness of the combined high intensity and short pulse duration of the source is evidenced in non-linear XUV-optics experiments. It further advances the implementation of XUV-pump-XUV-probe experiments and enables the investigation of strong field effects in the XUV spectral region.

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Propagation-assisted generation of intense few-femtosecond high-harmonic pulses

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The ongoing development of intense high-harmonic generation (HHG) sources has recently enabled highly non-linear ionization of atoms by the absorption of at least 10 extreme-ultraviolet (XUV) photons within a single atom (Senfftleben et al, arXiv:1911.01375). Here we investigate how the generation of these very intense HHG pulses in our 18-m-long beamline is aided by the reshaping of the fundamental, few-cycle, near-infrared (NIR) driving laser within a 30-cm-long HHG Xe medium. Using an incident NIR intensity that is higher than what is required for phase-matched HHG, signatures of reshaping are found by measuring the NIR blueshift and the fluorescence from the HHG medium along the propagation axis. These results are well reproduced by numerical calculations that show temporal compression of the NIR pulses in the HHG medium. The simulations predict that after refocusing an XUV beam waist radius of 320 nm and a clean attosecond pulse train can be obtained in the focal plane, with an estimated XUV peak intensity of 9×1015 W cm−2. Our results show that XUV intensities that were previously only available at large-scale facilities can now be obtained using moderately powerful table-top light sources.

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SeSo

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Wavelength-dependent orientation of the principal axes of photonic crystal fibers measured by windowed Fourier-transform spectral interferometry

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Optics Express
28 (2020) 2, 2156-2165

https://doi.org/10.1364/OE.381769

We present a novel polarization alignment technique based on windowed Fourier-transform (WFT) spectral interferometry to determine the wavelength-dependent orientation of the principal polarization axes of photonic crystal fibers (PCFs). To test the technique, a commercially available, 82.5-cm-long HC-800-02 type hollow-core PCF was measured. The angles belonging to the fast and the slow principal axes of the fiber were determined from the peak intensity values of the ridges in the WFT map at different wavelengths. We demonstrate that the orientation of the principal polarization axes of the tested PCF is wavelength-dependent. The precision of the angle measurement was better than 0.3°.

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Attosecond pulse-shaping using a seeded free-electron laser

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Attosecond pulses are central to the investigation of valence- and core-electron dynamics on their natural timescales. The reproducible generation and characterization of attosecond waveforms has been demonstrated so far only through the process of high-order harmonic generation. Several methods for shaping attosecond waveforms have been proposed, including the use of metallic filters, multilayer mirrors and manipulation of the driving field. However, none of these approaches allows the flexible manipulation of the temporal characteristics of the attosecond waveforms, and they suffer from the low conversion efficiency of the high-order harmonic generation process. Free-electron lasers, by contrast, deliver femtosecond, extreme-ultraviolet and X-ray pulses with energies ranging from tens of microjoules to a few millijoules. Recent experiments have shown that they can generate subfemtosecond spikes, but with temporal characteristics that change shot-to-shot. Here we report reproducible generation of high-energy (microjoule level) attosecond waveforms using a seeded free-electron laser. We demonstrate amplitude and phase manipulation of the harmonic components of an attosecond pulse train in combination with an approach for its temporal reconstruction. The results presented here open the way to performing attosecond time-resolved experiments with free-electron lasers.

Nature
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Generation of high-order harmonics with tunable photon energy and spectral width using double pulses

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Phys. Rev. A
102 (2020) 013504

https://doi.org/10.1103/PhysRevA.102.013504

This work theoretically investigates high-order harmonic generation in rare-gas atoms driven by two temporally delayed ultrashort laser pulses. Apart from their temporal delay, the two pulses are identical. Using a single-atom model of the laser-matter interaction it is shown that the photon energy of the generated harmonics is controllable within the range of one eV—a bandwidth comparable to the photon energy of the fundamental field—by varying the time delay between the generating laser pulses. It is also demonstrated that high-order harmonics generated by double pulses have advantageous characteristics, which mimick certain properties of an extreme ultraviolet monochromator. With the proposed method, a simpler setup at a much lower cost and comparatively higher spectral yield can be implemented in contrast to other approaches.

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Non-linear processes in the extreme ultraviolet

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J. Physics: Photonics
2 (2020) 042003

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Recent developments in extreme ultraviolet (XUV) and x-ray radiation sources have pushed pulse energies and durations to unprecedented levels that opened up the era of non-linear XUV and x-ray optics. In this quest, laser driven high order harmonic generation sources providing attosecond resolution in the XUV spectral region enabled XUV-pump-XUV-probe experiments, while Free Electron Laser research infrastructures offer unique x-ray brilliances for highly non-linear interactions and since recently, they too entered the sub-fs temporal regime. This topical review discusses the conceptual intricacies of non-linear XUV and x-ray processes, addresses experimental particularities and highlights recent applications of such processes with emphasis to laser driven XUV-attosecond source related research

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Strong-field effects induced in the extreme ultraviolet domain

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High Power Laser Sci. & Eng. 8, e44 (2020)

https://doi.org/10.1017/hpl.2020.43

Motivated by the achieved high intensities of novel extreme ultraviolet (XUV) radiation sources, such as free electron lasers and laser-driven high harmonic generation beamlines, we elaborate on their perspective in inducing observable strong field effects. The feasibility of extending such effects from the infrared and visible spectral regimes in the XUV domain is supported through numerically calculated models of near-future experiments. We highlight the advancement of performing studies in the time domain, using ultra-short XUV pulses, which allows for the temporal evolution of such effects to be followed. Experimental and theoretical obstacles and limitations are further discussed.

SeSo

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A perspective on high photon flux nonclassical light and applications in nonlinear optics

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Phys. Rev. Research
2 (2020) 013338

http://doi:10.1017/hpl.2020.44

Nonclassical light sources have a vital role in quantum optics as they offer a unique resource for studies in quantum technology. However, their applicability is restricted by their low intensity, while the development of new schemes producing intense nonclassical light is a challenging task. In this perspective article, we discuss potential schemes that could be used towards the development of high photon flux nonclassical light sources and their future prospects in nonlinear optics.

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SeSo

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Ultrafast plasma electron dynamics: a route to terahertz pulse shaping

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Phys. Rev. Appl. 13 (2020) 034044

https://doi.org/10.1103/PhysRevApp-lied.13.034044

Intense ultrafast laser interaction with solid-density plasma can lead to relativistic transient electron dynamics that are favorable for the generation of high-field terahertz (THz) pulses. We investigate the temporal characteristics of such THz pulses. Single-shot electro-optic measurements enable us to record the complete temporal profiles of the THz pulses emanating from planar Cu and aligned Cu-nanorod-array plasma. The temporal properties of the THz pulses exhibit several transients. Fully relativistic two-dimensional particle-in-cell simulations corroborate experimental observations and reveal that these features originate from electron microbunch emission from the target. Our results demonstrate that target nanostructuring provides a route to control such ultrafast electron dynamics and hence THz-pulse properties in the time domain.

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Characterisation and modelling of ultrashort laser-driven electromagnetic pulses

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Scientific Reports
10 (2020) 3108

https://doi.org/10.1038/s41598-020-59882-8

Recent advances on laser technology have enabled the generation of ultrashort (fs) high power (PW) laser systems. For such large scale laser facilities there is an imperative demand for high repetition rate operation in symbiosis with beamlines or end-stations. In such extreme conditions the generation of electromagnetic pulses (EMP) during high intense laser target interaction experiments can tip the scale for the good outcome of the campaign. The EMP effects are several including interference with diagnostic devices and actuators as well as damage of electrical components. The EMP issue is quite known in the picosecond (ps) pulse laser experiments but no systematic study on EMP issues at multi-Joule fs-class lasers has been conducted thus far. In this paper we report the first experimental campaign for EMP-measurements performed at the 200 TW laser system (VEGA 2) at CLPU laser center. EMP pulse energy has been measured as a function of the laser intensity and energy together with other relevant quantities such as (i) the charge of the laser-driven protons and their maximum energy, as well as (ii) the X-ray Kα emission coming from electron interaction inside the target. Analysis of experimental results demonstrate (and confirm) a direct correlation between the measured EMP pulse energy and the laser parameters such as laser intensity and laser energy in the ultrashort pulse duration regime. Numerical FEM (Finite Element Method) simulations of the EMP generated by the target holder system have been performed and the simulations results are shown to be in good agreement with the experimental ones.

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SeSo

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New injection and acceleration scheme of positrons in the laser-plasma bubble regime

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Phys. Rev. Acc. & B. 23 (2020) 091301

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A novel approach for positron injection and acceleration in the laser driven plasma wakefield is proposed. One ring-shaped beam and one coaxially propagating Gaussian beam drive wakefields in a preformed plasma volume filled with both electrons and positrons. The laser's ponderomotive force as well as the charge separation force in the front bucket of the first bubble are utilized to provide the transverse momenta of injected positrons and those positrons can be trapped by the focusing field and then accelerated by the wakefield. Theoretical analysis of the process is presented and verified by particle-in-cell simulations. The simulations show that relatively high-charge, quasimonoenergetic positron beams can be obtained.

eli attosecond

SeSo

Control of electron beam energy - spread by beam loading effects in a laser-plasma accelerator

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Plasma Phys. Contr. F. 62 (2020) 055004

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We present experimental results from a laser wakefield electron accelerator driven by 70 TW ultrashort laser pulses in helium and helium–nitrogen gaseous plasmas with two different nitrogen concentrations, showing distinct electron-beam qualities. In order to get a clear view of the involved phenomenon, two-dimensional particle-in-cell simulations are performed which not only agreed with the experimental results but also provided an investigation on the evolution of accelerating structures. The experimental and simulation results depict that the beam loading effect can strongly modify the longitudinal accelerating electric field of the wake wave, imposing diametrically opposite effects on the final electron-beam qualities, especially the energy-spread, in the helium–nitrogen gas mixtures with different nitrogen concentrations. In the helium–nitrogen-mixed plasma with a lower nitrogen concentration (0.5%), if appropriately controlled, the beam loading effect can be employed to flatten the accelerating electric field for reducing the electron-beam energy spread. In contrast, in the helium–nitrogen-mixed plasmas with a higher nitrogen concentration (5%), the accelerating electric field of the wake is locally reversed by the self-fields of the overloaded electron bunch, and the correspondingly generated negative-slope region of electric field increases the electron-beam energy-spread.

eli attosecond

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High stability positron beam generation based on ultra-intense laser

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Acta Physica Polonica A
137 (2020)

http://
doi.org/10.12693/
APhysPolA.137.156

Relativistic positron beams were generated by laser wakefield electrons bombarding on solid target. Very stable positron beams were generated in our experiments. The total yield of positrons is about 4.4×108 /shot. The energy spectra of positrons and electrons obey quasi-Maxwell distribution. Compared with the direct method, the indirect method produces positrons (38.5 MeV) and electrons (50.5 MeV) with much higher slope temperature.

SeSo

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Enhanced laser wakefield acceleration using dual-color relativistic pulses

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Plasma Phys. Contr. F.
62 (2020) 095012

https://doi.org/10.1088/1361-6587/aba481

In a recent article by Li *et al* (2019 *Sci. Adv.* 5. eaav7940), experimental results from a dual-color laser wakefield acceleration (LWFA) were presented. In the present paper we, primarily, focus on detailed simulation studies of such a scheme in the self-injection and ionization injection regimes, respectively. The spatiotemporally-overlapped 30 fs dual-color laser pulses are at fundamental (FL, 800 nm, 'red') and second-harmonic (SH, 400 nm, 'blue') wavelengths. They are (a) co-propagating in an under-dense plasma, (b) relativistically intense ($I > 10^{18}$ W cm^{-2}) and (c) having relatively high-energy (multi-Joule, loose focusing) and low-energy (sub-Joule, tight focusing), respectively. The basic concept of the scheme is the fact that the depletion length (L_{pd}) for a relativistic laser pulse in an under-dense plasma has an inverse quadratic dependence on the laser wavelength (~$1/\lambda^2$). Here, first by using a single FL 77 TW/30 fs laser pulse to drive a LWFA, an electron beam was accelerated up to ~400 MeV from a background plasma having an electron density of 10^{19} cm^{-3}. Then, by driving the same LWFA by co-propagating 'blue' 7 TW/30 fs and 'red' 70 TW/30 fs laser pulses, the electron energy reached ~700–800 MeV (maximum). The simulations confirm that in such a dual-color LWFA scheme, the role of the SH laser pulse is post-accelerating electrons after a rapid depletion of the FL laser pulse in the plasma. Furthermore, the SH pulse assists the ionization-injection of the electrons which is an additional benefit of the dual-color LWFA scheme.

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SeSo

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Angular dispersion boost of high order laser harmonics with Carbon nano-rods

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Optics Express
28 (2020) 4, 5355-5366

https://doi.org/10.1364/OE.3840788

Periodic surface gratings or photonic crystals are excellent tools for diffracting light and to collect information about the spectral intensity, if the target structure is known, or about the diffracting object, if the light source is well defined. However, this method is less effective in the case of extreme ultraviolet (XUV) light due to the high absorption coefficient of any material in this frequency range. Here we propose a nanorod array target in the plasma phase as an efficient dispersive medium for the intense XUV light which is originated from laser-plasma interactions where various high harmonic generation processes take place. The scattering process is studied with the help of particle-in-cell simulations and we show that the angular distribution of different harmonics after scattering can be perfectly described by a simple interference theory.

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Relativistic electron acceleration by surface plasma waves in the nonlinear regime

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High Power Laser Sci. & Eng. 8 (2020) e15

https://doi.org/10.1017/hpl.2020.14

The process of high energy electron acceleration along the surface of grating targets (GTs) that were irradiated by a relativistic, high-contrast laser pulse at an intensity $I=2.5\times10^{20}$ W/cm^2 was studied. Our experimental results demonstrate that for a GT with a periodicity twice the laser wavelength, the surface electron flux is more intense for a laser incidence angle that is larger compared to the resonance angle predicted by the linear model. An electron beam with a peak charge of ~2.7 nC/sr, for electrons with energies >1.5 MeV, was measured. Numerical simulations carried out with parameters similar to the experimental conditions also show an enhanced electron flux at higher incidence angles depending on the preplasma scale length. A theoretical model that includes ponderomotive effects with more realistic initial preplasma conditions suggests that the laser-driven intensity and preformed plasma scale length are important for the acceleration process. The predictions closely match the experimental and computational results.

eli attosecond

SeSo

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Bunching of light ions driven by heavy-ion front in multispecies ion beam accelerated by laser

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Phys. Rev. E 102 (2020) 023212

https://doi.org/10.1103/PhysRevE.102.023212

Deeply modulated ion spectra from contaminants present on the target surface were measured at the interaction of ultraintense $(2–5)\times10^{20}$W/cm^2 and high-contrast laser pulses (<~10−10) with thin (~μm) and ultrathin (~nm) targets. This phenomenon, observed over a wide range of laser and target parameters, suggests that it is a generic feature of multispecies ion acceleration at high laser pulse contrast. The modulation is ascribed to the acceleration of various ion species at the rear of the target with steplike density profiles which provide well-separated ion species in the accelerated beam. The observed coincidence of the velocity of the modulated region in the ion spectra with the maximum velocity of another ion with a lower mass-to-charge ratio is consistent with this model. The impact of heavy ions on light ions leads to a spectral "bunching" of light ions. Two-dimensional modeling has shown that high laser contrast prevents backside plasma expansion, which provides a well separated ion species with a steplike density profile that allows for the additional acceleration of "light" ions by the slower moving "heavy"-ion front. Spectral modulations can be controlled by tuning the ratio of heavy to light ions in future experiments with ultrathin rear coatings.

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Diagnostic of peak laser intensity by pair production from thin foil target

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Laser Phys. Lett.
17 (2020) 056101

https://doi.org/10.1088/1612-202X/ab83fe

It is known that in the small quantum parameter regime the Breit-Wheeler electron-positron pair creation cross section is extremely sensitive on the photon energy and on the background field. We present the dependence of positron yield on laser intensity in laser-foil interactions with the help of analytical theory and particle-in-cell simulaitons. These particles are emitted in a relatively narrow range of cone angles close to perpendicular direction relative to the laser beam axis. This allows for establishing an in situ intensity diagnostic by measuring the positron current in experiments where peta-watt class lasers are used.

SeSo

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Substantial enhancement of betatron radiation in cluster targets

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Phys. Rev. E
102 (2020) 053205

https://
doi.org/10.1103/
PhysRevE.102.053205

Betatron radiation generated by relativistic electrons during their wiggling motion in an ion channel is a well-studied source of x-ray photons. Due to the highly collimated emission such compact laser-driven sources have attracted significant attention in various laser or plasma-based applications, but the spectral intensity is still too low. The high repetition rate is also demanded, thus the pulse energy is strongly limited. Here, based on theory and computer simulations, we present a different method to enhance the radiation power by increasing the number of betatron oscillations along the acceleration path of electrons. A stronger wiggling of electrons is achieved by using clusterized gas targets, which allows one to achieve three orders of magnitude higher x-ray yield than in optimized uniform gas target with similar average electron density.

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SeSo

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Magnetic dipole moment generated in nano-droplets irradiated by circularly polarized laser pulse

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Phys. Rev. Research 2 (2020) 023088

https://doi.org/10.1364/OL.409410

We present a carrier-envelope offset (CEO) stable ytterbium-doped fiber chirped-pulse amplification system employing the technology of coherent beam combining and delivering more than 1 kW of average power at a pulse repetition rate of 80 MHz. The CEO stability of the system is 220 mrad rms, characterized out-of-loop with an f-to-2f interferometer in a frequency offset range of 10 Hz to 20 MHz. The high-power amplification system boosts the average power of the CEO stable oscillator by five orders of magnitude while increasing the phase noise by only 100 mrad. No evidence of CEO noise deterioration due to coherent beam combining is found. Low-frequency CEO fluctuations at the chirped-pulse amplifier are suppressed by a “slow loop” feedback. To the best of our knowledge, this is the first demonstration of a coherently combined laser system delivering an outstanding average power and high CEO stability at the same time.

eli attosecond

SeSo

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Measurement of four-photon absorption in GaP and ZnTe semiconductors

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Optics Express
28 (2020) 8, 12352-12362

https://doi.org/10.1364/OE.382388

Intensity-dependent effective four-photon absorption (4PA) coefficients in GaP and ZnTe semiconductors were measured by the z-scan method using pump pulses of 1.75 µm wavelength, 135 fs duration, and up to 500 GWcm^{-2} intensity. A nonlinear pulse propagation model, including linear dispersion and 4PA was used to obtain the 4PA coefficients from measurements. The intensity-dependent effective 4PA coefficients vary from 2.6×10^{-4} to 65×10^{-4} cm^{5}GW^{-3} in GaP, and from 3.5×10^{-4} to 9.1×10^{-4} cm^{5}GW^{-3} in ZnTe. The anisotropy in 4PA was shown in GaP. The knowledge of 4PA coefficients is important for the design of semiconductor photonics devices.

SeSo

Characterization of aminopyridinium based organic crystals for THz generation

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Opt. & Laser Techn. 131 (2020) 106394

https://doi.org/10.1016/j.optlastec.2020.106394

Two aminopyridinium based novel organic materials, 2-amino-5-nitropyridinium p-phenolsulfonate (2A5NPP) and 2,6-diaminopyridinium-4-nitrophenolate-4-nitrophenol (DAP+NP-NP) were characterized in the 0–2 THz range by time-domain terahertz (THz) spectroscopy. Density functional theory calculations were carried out to find the molecular vibrational modes of the materials in 0–15 THz range and partially compared with the experimental results. THz absorption peaks were assigned to molecular vibration modes. THz pulse generation by optical rectification in both materials was studied using optical pump pulses of 1030 nm wavelength. The anisotropy of THz generation was investigated in detail by rotating the polarization of the optical pump with respect to the crystal axes.

eli attosecond

SeSo

Programmable generation of terahertz bursts in chirped-pulse laser amplification

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Optica
7 (2020) 1758-1763

https://doi.org/10.1364/OPTICA.403184

Amplified bursts of laser pulses are sought for various machining, deposition, spectroscopic, and strong-field applications. Standard frequency- and time-domain techniques for pulse division become inadequate when intraburst repetition rates reach the terahertz (THz) range as a consequence of inaccessible spectral resolution, requirement for interferometric stability, and collapse of the chirped-pulse amplification (CPA) concept due to the loss of usable bandwidth needed for safe temporal stretching. Avoiding the burst amplification challenge and resorting to lossy post-division of an isolated laser pulse after CPA leaves the limitations of frequency- and time-domain techniques unsolved. In this Letter, we demonstrate an approach that successfully combines amplitude and phase shaping of THz bursts, formed using the Vernier effect, with active stabilization of spectral modes and efficient energy extraction from a CPA regenerative amplifier. As proof of concept, the amplified bursts of femtosecond near-infrared pulses are down-converted into tunable THz-frequency pulses via optical rectification.

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SeSo

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Laser-driven strong-field terahertz sources

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Adv. Opt. Mat.
8(2020) 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.

US

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Striking generic impact of light -induced non-adiabaticity in polyatomic molecules

CS. FÁBRI, B. LASORNE, G. J. HALÁSZ, L. S. CEDERBAUM, and Á. VIBÓK

J. Phys. Chem. Lett. 11 (2020) 5324−5329

https://doi.org/10.1021/acs.jpclett.0c01574

Non-adiabaticity, i.e., the effect of mixing electronic states by nuclear motion, is a central phenomenon in molecular science. The strongest nonadiabatic effects arise due to the presence of conical intersections of electronic energy surfaces. These intersections are abundant in polyatomic molecules. Laser light can induce in a controlled manner new conical intersections, called light-induced conical intersections, which lead to strong nonadiabatic effects similar to those of the natural conical intersections. These effects are, however, controllable and may even compete with those of the natural intersections. In this work we show that the standard low-energy vibrational spectrum of the electronic ground state can change dramatically by inducing non-adiabaticity via a light-induced conical intersection. This generic effect is demonstrated for an explicit example by full-dimensional high-level quantum calculations using a pump-probe scheme with a moderate-intensity pump laser and a weak probe laser.

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US

Three-player polaritons: nonadiabatic fingerprints in an entangled atom-molecule-photon system

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New J. Phys.
22 (2020) 053001

https://doi.org/10.1088/1367-2630/ab8264

A quantum system composed of a molecule and an atomic ensemble, confined in a microscopic cavity, is investigated theoretically. The indirect coupling between atoms and the molecule, realized by their interaction with the cavity radiation mode, leads to a coherent mixing of atomic and molecular states, and at strong enough cavity field strengths hybrid atom–molecule–photon polaritons are formed. It is shown for the Na2 molecule that by changing the cavity wavelength and the atomic transition frequency, the potential energy landscape of the polaritonic states and the corresponding spectrum could be changed significantly. Moreover, an unforeseen intensity borrowing effect, which can be seen as a strong nonadiabatic fingerprint, is identified in the atomic transition peak, originating from the contamination of the atomic excited state with excited molecular rovibronic states.

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Robust field-dressed spectra of diatomics in an optical lattice

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Phys. Chem. Chem. Phys. 2020, Advance Article

https://doi.org/10.1039/C9CP06587C

The absorption spectra of the cold Na_2 molecule dressed by a linearly polarized standing laser wave is investigated with a theoretical model incorporating translational, electronic, vibrational as well as rotational degrees of freedom. In such a situation a light-induced conical intersection (LICI) can be formed (J. Phys. B: At. Mol. Opt. Phys., 2008, 41, 221001). To measure the spectra a weak field is used whose propagation direction is perpendicular to the direction of the dressing field but has identical polarization direction. Although LICIs are present in our model, the simulations demonstrate a very robust absorption spectrum, which is insensitive to the intensity and the wavelength of the dressing field and which does not reflect clear signatures of light-induced nonadiabatic phenomena related to the strong mixing between the electronic, vibrational, rotational and translational motions. However, by widening artificially the very narrow translational energy level gaps, the fingerprint of the LICI appears to some extent in the spectrum.

US

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Control of photodissociation with the dynamic Stark effect induced by THz pulses

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Phys. Rev. Research
2 (2020) 013338

https://doi.org/10.1103/PhysRevResearch.2.013338

We demonstrate how dynamic Stark control can be achieved on molecular photodissociation in the dipole limit, using single-cycle (full width at half maximum) laser pulses in the terahertz (THz) regime. As the laser-molecule interaction follows the instantaneous electric field through the permanent dipoles, the molecular potentials dynamically oscillate and so do the crossings between them. In this paper, we consider rotating-vibrating diatomic molecules (two-dimensional description) and reveal the interplay between the dissociating wave packet and the dynamically fluctuating crossing seam located in the configuration space of the molecules spanned by the R vibrational and θ rotational coordinates. Our showcase example is the widely studied lithium fluoride molecule for which the two lowest Σ states are nonadiabatically coupled at an avoided crossing (AC); furthermore a low-lying pure repulsive Π state is energetically close. Optical pumping of the system in the ground state thus results in two dissociation channels: one indirect route via the AC in the ground Σ state and one direct path in the Π state. We show that applying THz control pulses

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Quantum light-induced nonadiabatic phenomena in the absorption spectrum of formaldehyde: Full- and reduced-dimensionality studies

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J. Chem. Phys.
153 (2020) 234302

https://doi.org/10.1063/5.0035870

The coupling of a molecule to a cavity can induce conical intersections of the arising polaritonic potential energy surfaces. Such intersections give rise to the strongest possible nonadiabatic effects. By choosing an example that does not possess nonadiabatic effects in the absence of the cavity, we can study, for the first time, the emergence of these effects in a polyatomic molecule due to its coupling with the cavity taking into account all vibrational degrees of freedom. The results are compared with those of reduced-dimensionality models, and the shortcomings and merits of the latter are analyzed.

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On the preservation of coherence in the electronic wavepacket of a neutral and rigid polyatomic molecule

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J. Phys. B
53 (2020) 184005

https://doi.org/10.1088/1361-6455/aba10e

We present various types of reduced models including five vibrational modes and three electronic states for the pyrazine molecule in order to investigate the lifetime of electronic coherence in a rigid and neutral system. Using ultrafast optical pumping in the ground state (1^1A_g), we prepare a coherent superposition of two bright excited states, 1^1B_{2u} and 1^1B_{1u}, and reveal the effect of the nuclear motion on the preservation of the electronic coherence induced by the laser pulse. More specifically, two aspects are considered: the anharmonicity of the potential energy surfaces and the dependence of the transition dipole moments (TDMs) with respect to the nuclear coordinates. To this end, we define an 'ideal model' by making three approximations: (i) only the five totally symmetric modes move, (ii) which correspond to uncoupled harmonic oscillators, and (iii) the TDMs from the ground electronic state to the two bright states are constant (Franck–Condon approximation). We then lift the second and third approximations by considering, first, the effect of anharmonicity, second, the effect of coordinate-dependence of the TDMs (first-order Herzberg–Teller contribution), third, both. Our detailed numerical study with quantum dynamics is meant to be realistic for pyrazine over about 20 femtoseconds, and was further extended so as to probe the effect of such approximations on a model system. We show that long-term revivals of the electronic coherence persist up to the picosecond time range even for the most realistic model.

eli attosecond

US

Ambient pressure CO_2 hydrogenation over a cobalt/manganese-oxide nanostructured interface: A combined in situ and ex situ study

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Proc. SPIE
11493, 114930Z

https://doi.org/10.1016/j.jcat.2020.03.028

We report on a cobalt/manganese-oxide interface catalyst with outstanding activity and selectivity towards methane even at high temperatures and ambient pressure in CO_2 hydrogenation. The catalyst was formed from a $MnCo_2O_4$-based spinel structure during the oxidative-reductive pretreatment process just before the catalytic tests. Several Mn-, Fe- and Ni-containing cobaltite spinel and reverse spinel structures were tested to find the best overall performer. The reusable $MnCo_2O_4$-based structure featured a CO_2 consumption rate of ~8500 $nmol \cdot g^{-1} \cdot s^{-1}$. Even though methane is not the thermodynamically favoured product, it was produced with ~80% and ~50% selectivity at ambient pressure at 673 K and 823 K, respectively. This unexpected finding is linked to the presence of a unique nanostructured Co/Mn(II)O catalyst with a surface composition of $Mn_{3.3}Co_{2.0}O_{4.7}$ formed after the pretreatment activation step. Over this phase, the reduction of CO_2 progresses through bridge bonded formate located at the Co/Mn^{2+} interface and this is mostly responsible for high temperature methane formation. This hypothesis is proven here by the reported combination of ex-situ XRD, TPR, HRTEM-ED, HAADF-EDX and in-situ NAP-XPS and DRIFTS techniques.

eli attosecond

US

Adsorption of azobenzene on hexagonal boron nitride nanomesh supported by Rh(111)

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J. Phys. Chem. C
124 (2020) 14182−14194

https://doi.org/10.1021/acs.jpcc.0c01725

Adsorption properties of azobenzene, the prototypical molecular switch, were investigated on a hexagonal boron nitride (h-BN) monolayer (“nanomesh”) prepared on Rh(111). The h-BN layer was produced by decomposing borazine (B3N3H6) at 1000–1050 K. Temperature-programmed desorption (TPD) studies revealed that azobenzene molecules adsorbed on the “wire” and “pore” regions desorb at slightly different temperatures. Angle-resolved high-resolution electron energy loss spectroscopy (HREELS) measurements demonstrated that the first molecular layer is characterized predominantly by an adsorption geometry with the molecular plane parallel to the surface. Scanning tunneling microscopy (STM) indicated a clear preference for adsorption in the pores, manifesting a templating effect, but in some cases one-dimensional molecular stripes also form, implying attractive molecule–molecule interaction. Density functional theory (DFT) calculations provided further details regarding the adsorption energetics and bonding and confirmed the experimental findings that the molecules adsorb with the phenyl rings parallel to the surface, preferentially in the pores, and indicated also the presence of an attractive molecule–molecule interaction.

US

Iodine (I) expulsion at photoirradiated mixed halide perovskite interface. Should I stay or should I go?

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ACS Energy Lett.
5 (2020) 6, 1872–188

https://doi.org/10.1021/acsenergylett.0c00925

Visible light irradiation of a mixed halide perovskite film in contact with a solvent (dichloromethane, DCM) in which the film otherwise is stable leads to selective expulsion of iodide (I) from the film with a concurrent shift in the band edge to lower wavelengths. We have now employed mixed halide perovskites to uncover the influence of A-site cation [methylammonium (MA) and cesium (Cs)] on the mobility of iodide ions under photoirradiation. In the absence of solvent contact, the mixed halide perovskite films undergo photoinduced segregation with a rate constant that decreases with increasing Cs content. Interestingly, the iodide expulsion rate in DCM is strongly dependent on the rate of photoinduced segregation. At Cs atomic concentrations greater than 50%, the films become stable as the iodide expulsion is largely suppressed. The role of the A-site cation in dictating the mobility of halide ions is discussed.

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US

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Photocorrosion at irradiated perovskite/electrolyte interfaces

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JACS 142 (2020)
52, 21595–21614

https://
doi.org/10.1021/
jacs.0c10348

Metal–halide perovskites transformed optoelectronics research and development during the past decade. They have also gained a foothold in photocatalytic and photoelectrochemical processes recently, but their sensitivity to the most commonly applied solvents and electrolytes together with their susceptibility to photocorrosion hinders such applications. Understanding the elementary steps of photocorrosion of these materials can aid the endeavor of realizing stable devices. In this Perspective, we discuss both thermodynamic and kinetic aspects of photocorrosion processes occurring at the interface of perovskite photocatalysts and photoelectrodes with different electrolytes. We show how combined in situ and operando electrochemical techniques can reveal the underlying mechanisms. Finally, we also discuss emerging strategies to mitigate photocorrosion (such as surface protection, materials and electrolyte engineering, etc.).

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Hybrid FeNiOOH/α-Fe_2O_3/ graphene photoelectrodes with advanced water oxidation performance

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Adv. Funct. Materials (2020) 2002124

https://doi.org/10.1002/adfm.202002124

In this study, the photoelectrochemical behavior of electrodeposited FeNiOOH/Fe_2O_3/graphene nanohybrid electrodes is investigated, which has precisely controlled structure and composition. The photoelectrode assembly is designed in a bioinspired manner where each component has its own function: Fe_2O_3 is responsible for the absorption of light, the graphene framework for proper charge carrier transport, while the FeNiOOH overlayer for facile water oxidation. The effect of each component on the photoelectrochemical behavior is studied by linear sweep photovoltammetry, incident photon-to-charge carrier conversion efficiency measurements, and long-term photoelectrolysis. 2.6 times higher photocurrents are obtained for the best-performing FeNiOOH/Fe_2O_3/graphene system compared to its pristine Fe_2O_3 counterpart. Transient absorption spectroscopy measurements reveal an increased hole-lifetime in the case of the Fe_2O_3/graphene samples. Long-term photoelectrolysis measurements in combination with Raman spectroscopy, however, prove that the underlying nanocarbon framework is corroded by the photogenerated holes. This issue is tackled by the electrodeposition of a thin FeNiOOH overlayer, which rapidly accepts the photogenerated holes from Fe_2O_3, thus eliminating the pathway leading to the corrosion of graphene.

eli attosecond

US

E. Rita SZABÓ

Spectral and spatial shaping of laser-driven proton beams using a pulsed high-field magnet beamline

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Scientific Reports 10 (2020) 9118

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Intense laser-driven proton pulses, inherently broadband and highly divergent, pose a challenge to established beamline concepts on the path to application-adapted irradiation field formation, particularly for 3D. Here we experimentally show the successful implementation of a highly efficient (50% transmission) and tuneable dual pulsed solenoid setup to generate a homogeneous (laterally and in depth) volumetric dose distribution (cylindrical volume of 5 mm diameter and depth) at a single pulse dose of 0.7 Gy via multi-energy slice selection from the broad input spectrum. The experiments were conducted at the Petawatt beam of the Dresden Laser Acceleration Source Draco and were aided by a predictive simulation model verified by proton transport studies. With the characterised beamline we investigated manipulation and matching of lateral and depth dose profiles to various desired applications and targets. Using an adapted dose profile, we performed a first proof-of-technical-concept laser-driven proton irradiation of volumetric in-vitro tumour tissue (SAS spheroids) to demonstrate concurrent operation of laser accelerator, beam shaping, dosimetry and irradiation procedure of volumetric biological samples.

eli attosecond

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A feasibility study of zebrafish embryo irradiation with laser-accelerated protons

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Rev. Sci. Instrum.
91 (2020) 063303

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The development from single shot basic laser plasma interaction research toward experiments in which repetition rated laser-driven ion sources can be applied requires technological improvements. For example, in the case of radio-biological experiments, irradiation duration and reproducible controlled conditions are important for performing studies with a large number of samples. We present important technological advancements of recent years at the ATLAS 300 laser in Garching near Munich since our last radiation biology experiment. Improvements range from target positioning over proton transport and diagnostics to specimen handling. Exemplarily, we show the current capabilities by performing an application oriented experiment employing the zebrafish embryo model as a living vertebrate organism for laser-driven proton irradiation. The size, intensity, and energy of the laser-driven proton bunches resulted in evaluable partial body changes in the small (<1 mm) embryos, confirming the feasibility of the experimental system. The outcomes of this first study show both the appropriateness of the current capabilities and the required improvements of our laser-driven proton source for in vivo biological experiments, in particular the need for accurate, spatially resolved single bunch dosimetry and image guidance.

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Improved FBX chemical dosimeter system with enhanced radiochemical yield for reference dosimetry in radiobiology and radiotherapy

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Rad. Phys. and Chem. 174 (2020) 108899

https://doi.org/10.1016/j.radphyschem.2020.108899

Radiation dosimetry plays important role in the reproducibility of radiobiology experiments, in the replicability of results, as well as in the successful and safe use of radiotherapy procedures. The consistency and accuracy of the applied dosimetry methods pre-define the outcomes of these applications. This paper presents a version of the well-known ferrous sulphate – benzoic acid – xylenol orange (FBX) chemical dosimeter with improved sensitivity, accuracy and precision. Sensitivity is increased due to a slight modification in composition and the preparation procedures. We use stock solutions for the preparation of the dosimeter solution, which consists of 1 mM ferrous sulphate and 16 mM benzoic acid with 0.25 mM xylenol orange added post-irradiation. The nonlinear response to the absorbed dose of this system is eliminated by the increased ferrous sulphate concentration, permitting the calculation of the absorbed dose by a linear relationship between the absorbed dose and the optical absorbance of the solution. The measured chemical yield of our dosimeter is $9.08 \cdot 10^{-6}$ mol/J for 6 MV photon beams and $6.42 \cdot 10^{-6}$ mol/J for 250 kVp x-rays. This is a 24% enhancement over the original FBX solution, which permits a finer dose resolution. The accuracy and precision of our method is assured by a well-designed and consistently used practice. A custom designed multipurpose PMMA slab phantom was used for irradiation in reference conditions. This phantom can be used for irradiation in reference conditions of dosimetric solutions, dosimetric films and chemical or biological samples. The combined standard uncertainty of this system is 1.12%, which can be improved by using an appropriate temperature correction factor. Furthermore, a working protocol has been established which allows dosimetry measurements using less than 1 mL dosimetric solutions.

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Origin of vibrational wavepacket dynamics in Fe carbene photosensitizer determined with femtosecond X-ray emission and scattering

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Nature Communications 11 (2020) 634

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The non-equilibrium dynamics of electrons and nuclei govern the function of photoactive materials. Disentangling these dynamics remains a critical goal for understanding photoactive materials. Here we investigate the photoinduced dynamics of the $[Fe(bmip)_2]^{2+}$ photosensitizer, where bmip = 2,6-bis(3-methyl-imidazole-1-ylidine)-pyridine, with simultaneous femtosecond-resolution Fe Kα and Kβ X-ray emission spectroscopy (XES) and X-ray solution scattering (XSS). This measurement shows temporal oscillations in the XES and XSS difference signals with the same 278 fs period oscillation. These oscillations originate from an Fe-ligand stretching vibrational wavepacket on a triplet metal-centered (3MC) excited state surface. This 3MC state is populated with a 110 fs time constant by 40% of the excited molecules while the rest relax to a 3MLCT excited state. The sensitivity of the Kα XES to molecular structure results from a 0.7% average Fe-ligand bond length shift between the 1 s and 2p core-ionized states surfaces.

Sophie E. CANTON

Hot branching dynamics in a light-harvesting iron carbene complex revealed by ultrafast X-ray emission spectroscopy

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Angewandte Chemie 59 (2020) 1, 354-372

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Iron N-heterocyclic carbene (NHC) complexes have received a great deal of attention recently because of their growing potential as light sensitizers or photocatalysts. We present a sub-ps X-ray spectroscopy study of an FeIINHC complex that identifies and quantifies the states involved in the deactivation cascade after light absorption. Excited molecules relax back to the ground state along two pathways: After population of a hot ^{3}MLCT state, from the initially excited ^{1}MLCT state, 30 % of the molecules undergo ultrafast (150 fs) relaxation to the ^{3}MC state, in competition with vibrational relaxation and cooling to the relaxed ^{3}MLCT state. The relaxed ^{3}MLCT state then decays much more slowly (7.6 ps) to the ^{3}MC state. The ^{3}MC state is rapidly (2.2 ps) deactivated to the ground state. The ^{5}MC state is not involved in the deactivation pathway. The ultrafast partial deactivation of the ^{3}MLCT state constitutes a loss channel from the point of view of photochemical efficiency and highlights the necessity to screen transition-metal complexes for similar ultrafast decays to optimize photochemical performance.

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Attosecond spectroscopy reveals alignment dependent core-hole dynamics in the ICl molecule

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Nature Communications 11 (2020) 1 5810

https://doi.org/10.1038/s41467-020-19496-0

The removal of electrons located in the core shells of molecules creates transient states that live between a few femtoseconds to attoseconds. Owing to these short lifetimes, time-resolved studies of these states are challenging and complex molecular dynamics driven solely by electronic correlation are difficult to observe. Here, we obtain few-femtosecond core-excited state lifetimes of iodine monochloride by using attosecond transient absorption on iodine $4d^{-1}6p$ transitions around 55 eV. Core-level ligand field splitting allows direct access of excited states aligned along and perpendicular to the ICl molecular axis. Lifetimes of 3.5 ± 0.4 fs and 4.3 ± 0.4 fs are obtained for core-hole states parallel to the bond and 6.5 ± 0.6 fs and 6.9 ± 0.6 fs for perpendicular states, while nuclear motion is essentially frozen on this timescale. Theory shows that the dramatic decrease of lifetime for core-vacancies parallel to the covalent bond is a manifestation of non-local interactions with the neighboring Cl atom of ICl.

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Electronic decay through non-linear carbon chains

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J. Phys. B
53 (2020) 184006

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A multielectron wave-packet propagation method was used to calculate the electronic decay of oxygen and fluorine 2*s* vacancies for a group of trifluoroalkyl alcohols, $HOC_nH_{(2n-1)}F_3$, with *n* between 1 and 5. Whether ionizing O2*s* or F2*s* orbitals, it is shown that an electron can be emitted non-locally from the opposite terminus of the molecule. The decay of the $O(2s^{-1})$ state is found to be about 2–3 times faster than that of the $F(2s^{-1})$, but in both cases the process takes only a few femtoseconds, demonstrating a highly efficient energy transfer through the carbon bridge. A comparison to the previously reported non-local decay in linear difluorocumulenone systems shows that the non-linearity of the trifluoroalkyl alcohols does not appear to dramatically influence the decay efficiency. These results shed light onto the nature of the scaling of electron correlation and open the door to the potential design of molecules that take advantage of this mechanism.

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Strong-field nano-optics

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Rev. Mod. Phys. 92 (2020) 025003

https://doi.org/10.1103/RevModPhys.92.025003

The present status and development of strong-field nano-optics, an emerging field of nonlinear optics, is discussed. A nonperturbative regime of light-matter interactions is reached when the amplitude of the external electromagnetic fields that are driving a material approach or exceed the field strengths that bind the electrons inside the medium. In this strong-field regime, light-matter interactions depend on the amplitude and phase of the field, rather than its intensity, as in more conventional perturbative nonlinear optics. Traditionally such strong-field interactions have been intensely investigated in atomic and molecular systems, and this has resulted in the generation of high-harmonic radiation and laid the foundations for contemporary attosecond science. Over the past decade, however, a new field of research has emerged, the study of strong-field interactions in solid-state nanostructures. By using nanostructures, specifically those made out of metals, external electromagnetic fields can be localized on length scales of just a few nanometers, resulting in signficantly enhanced field amplitudes that can exceed those of the external field by orders of magnitude in the vicinity of the nanostructures. This leads not only to dramatic enhancements of perturbative nonlinear optical effects but also to significantly increased photoelectron yields. It resulted in a wealth of new phenomena in laser-solid interactions that have been discovered in recent years. These include the observation of above-threshold photoemission from single nanostructures, effects of the carrier-envelope phase on the photoelectron emission yield from metallic nanostructures, and strong-field acceleration of electrons in optical near fields on subcycle timescales. The current state of the art of this field is reviewed, and several scientific applications that have already emerged from the fundamental discoveries are discussed. These include, among others, the coherent control of localized electromagnetic fields at the surface of solid-state nanostructures and of free-electron wave packets by such optical near fields, resulting in the creation of attosecond electron bunches, the coherent control of photocurrents on nanometer length and femtosecond timescales by the electric field of a laser pulse, and the development of new types of ultrafast electron microscopes with unprecedented spatial, temporal, and energy resolution. The review concludes by highlighting possible future developments, discussing emerging topics in photoemission and potential strong-field nanophotonic devices, and giving perspectives for coherent ultrafast microscopy techniques. More generally, it is shown that the synergy between ultrafast science, plasmonics, and strong-field physics holds promise for pioneering scientific discoveries in the upcoming years.

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Tuning plasmonic field enhancement and transients by far-field coupling between nanostructures

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Appl. Phys. Lett. 117 (2020) 081105

https://doi.org/10.1063/5.0015374

We study how the collective effects of nanoparticles arranged in rectangular arrays influence their temporal plasmon response and field enhancement property. By systematically changing the lattice constant for arrays containing identical metal nanorods, we experimentally demonstrate how grating-induced effects affect the position and, more importantly, the broadening of extinction spectra. We correlate these effects with the achievable field enhancement and the temporal duration of plasmon transients and formulate criteria for the generation of enhanced few-cycle localized plasmon oscillations.

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Few-cycle localized plasmon oscillations

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Scientific Reports
10 (2020) 12986

https://doi.org/10.1038/s41598-020-69761-x

The generation of few-cycle laser pulses proved to be a key enabling technology in strong-field physics and ultrafast science. The question naturally arises whether one can induce few-cycle localized plasmon oscillations in optical near-fields. Here, we perform a comparative study of different plasmonic nanoresonators illuminated by few-cycle pulses. We analyze the number of cycles (NOC) of the plasmonic field, the near-field enhancement (NFE) as well as the figure of merit NFE/NOC. The pulse length dependence of these quantities is also investigated. Throughout the inspected pulse-length interval silica-gold and silica-silver core–shell monomers have the potential to preserve the NOC of the incoming pulse, silver bow-ties result in the highest NFE, whereas gold core–shell dimers have the highest NFE/NOC. Based on the analysis, silver bow-ties, gold core–shell and silver nanorod dimers proved to be the most suitable for few-cycle near-field amplification.

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Time-resolved momentum microscopy with a 1 MHz high-harmonic extreme ultraviolet beamline

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Rev. Sci. Instr. 91 (2020) 063905

https://doi.org/10.1063/5.0006531

Recent progress in laser-based high-repetition rate extreme ultraviolet (EUV) light sources and multidimensional photoelectron spectroscopy enables the build-up of a new generation of time-resolved photoemission experiments. Here, we present a setup for time-resolved momentum microscopy driven by a 1 MHz fs EUV table-top light source optimized for the generation of 26.5 eV photons. The setup provides simultaneous access to the temporal evolution of the photoelectron's kinetic energy and in-plane momentum. We discuss opportunities and limitations of our new experiment based on a series of static and time-resolved measurements on graphene.

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Dose-dependent changes after proton and photon irradiation in a zebrafish model

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Anticancer Res.
40 (2020) 11 6123-6135

https://doi.org/10.21873/anticanres.14633

Background/Aim: The importance of hadron therapy in the cancer management is growing. We aimed to refine the biological effect detection using a vertebrate model. Materials and Methods: Embryos at 24 and 72 h postfertilization were irradiated at the entrance plateau and the mid spread-out Bragg peak of a 150 MeV proton beam and with reference photons. Radiation-induced DNA double-strand breaks (DSB) and histopathological changes of the eye, muscles and brain were evaluated; deterioration of specific organs (eye, yolk sac, body) was measured. Results: More and longer-lasting DSBs occurred in eye and muscle cells due to proton versus photon beams, albeit in different numbers. Edema, necrosis and tissue disorganization, (especially in the eye) were observed. Dose-dependent morphological deteriorations were detected at ≥10 Gy dose levels, with relative biological effectiveness between 0.99±0.07 (length) and 1.12±0.19 (eye). Conclusion: Quantitative assessment of radiation induced changes in zebrafish embryos proved to be beneficial for the radiobiological characterization of proton beams.

US

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Near-field-induced femtosecond breakdown of plasmonic nanoparticles

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Plasmonics
15 (2020)

https://doi.org/10.1007/s11468-019-01043-3

We studied the evolution of femtosecond breakdown in lithographically produced plasmonic nanoparticles with increasing laser intensity. Localized plasmons were generated with 40-fs laser pulses with up to $1.4 \cdot 10^{12}$ W/cm^2 peak intensity. The damage morphology shows substantial variation with intensity, starting with the detachment of hot spots and stochastic nanoparticle removal. For higher intensities, we observe precise nanolithographic mapping of near-field distributions via ablation. The common feature of these phenomena is the central role played by the single plasmonic hot spot of the triangular nanoparticles used. We also derive a damage threshold value from stochastic damage trends on the arrays fostering the optimization of novel nanoarchitectures for nonlinear plasmonics.

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Photoelecton emission from silver nanoparticles after laser irradiation

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J. Phys.: Conf. Ser. 1412 092022 (2020)

https://doi.org/10.1088/1742-6596/1412/9/092022

We present theoretical study of photoelectron emission from silver nanoparticles after laser irradiation based on the classical simulation of the electron trajectories. The individual electron trajectories were analized with and without taken into account the image force between the emitted electron and its mirror charge. We show also the calculated energy spectra in comparision with the available experimental data. We observed a significant effect of the image acceleration to the calculated electron spectra.

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Oxygen vacancies induced photoluminescence in $SrZnO_2$ nanophosphors probed by theoretical and experimental analysis

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Scientific Reports
10 (2020) 17364

https://doi.org/10.1038/s41598-020-74436-8

We report, for the first time, the influence of oxygen vacancies on band structure and local electronic structure of $SrZnO_2$ (SZO) nanophosphors by combined first principle calculations based on density functional theory and full multiple scattering theory, correlated with experimental results obtained from X-ray absorption and photoluminescence spectroscopies. The band structure analysis from density functional theory revealed the formation of new energy states in the forbidden gap due to introduction of oxygen vacancies in the system, thereby causing disruption in intrinsic symmetry and altering bond lengths in SZO system. These defect states are anticipated as origin of observed photoluminescence in SZO nanophosphors. The experimental X-ray absorption near edge structure (XANES) at Zn and Sr K-edges were successfully imitated by simulated XANES obtained after removing oxygen atoms around Zn and Sr cores, which affirmed the presence and signature of oxygen vacancies on near edge structure.

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Dimethylammonium iodide stabilized bismuth halide perovskite photocatalyst for hydrogen evolution

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Nano Research
14 (2020) 4, 1116–1125

https://doi.org/10.1007/s12274-020-3159-0

Metal halide perovskites have emerged as novel and promising photocatalysts for hydrogen generation. Currently, their stability in water is a vital and urgent research question. In this paper a novel approach to stabilize a bismuth halide perovskite $[(CH_3)_2NH_2]_3[BiI_6]$ (DA_3BiI_6) in water using dimethylammonium iodide (DAI) without the assistance of acids or coatings is reported. The DA_3BiI_6 powder exhibits good stability in DAI solutions for at least two weeks. The concentration of DAI is found as a critical parameter, where the I-ions play the key role in the stabilization. The stability of DA_3BiI_6 in water is realized via a surface dissolution–recrystallization process. Stabilized DA3BiI6 demonstrates constant photocatalytic properties for visible light-induced photo-oxidation of I- ions and with $PtCl_4$ as a co-catalyst (Pt-DA_3BiI_6), photocatalytic H_2 evolution with a rate of 5.7 $\mu mol \cdot h^{-1}$ from HI in DAI solution, obtaining an apparent quantum efficiency of 0.83% at 535 nm. This study provides new insights on the stabilization of metal halide perovskites for photocatalysis in aqueous solution.

US

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High-order harmonic generation as induced by a quantized field: phase-space picture

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Phys. Rev. A
101 (2020) 013418

https://doi.org/10.1103/PhysRevA.101.013418

The interaction of matter with a quantized electromagnetic mode is considered. Representing a strong exciting field, the mode is assumed to contain a large number of photons. As a result, the material response is highly nonlinear: the completely quantized description results in generation of high harmonics. In order to understand the essence of the physical processes that are involved, we consider a finite dimensional model for the material system. Using an appropriate description in phase space, this approach leads to a transparent picture showing that the interaction splits the initial, exciting coherent state into parts, and the rapid change of the populations of these parts (that are coherent states themselves) results in the generation of high-order harmonics as secondary radiation. The method we use is an application of the discrete lattice of coherent states that was introduced by von Neumann.

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Density-based one-dimensional model potentials for strong-field simulations in He, H_2^+, and H_2

Attila CZIRJÁK

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Phys. Rev. A 101 (2020) 023405

https://doi.org/10.1103/PhysRevA.101.023405

We present results on the accurate one-dimensional (1D) modeling of simple atomic and molecular systems excited by strong laser fields. We use atomic model potentials that we derive from the corrections proposed earlier using the reduced ground-state density of a three-dimensional (3D) single-active electron atom. The correction involves a change of the asymptotics of the 1D Coulomb model potentials while maintaining the correct ground-state energy. We present three different applications of this method: we construct correct 1D models of the hydrogen molecular ion, the helium atom, and the hydrogen molecule using improved parameters of existing soft-core Coulomb potential forms. We test these 1D models by comparing the corresponding numerical simulation results with their 3D counterparts in typical strong-field physics scenarios with near- and mid-infrared laser pulses, having peak intensities in the 10^{14}–10^{15} W/cm^2 range, and we find an impressively increased accuracy in the dynamics of the most important atomic quantities on the time scale of the excitation. We also present the high-order harmonic spectra of the He atom, computed using our 1D atomic model potentials. They show a very good match with the structure and phase obtained from the 3D simulations in an experimentally important range of excitation amplitudes.

eli attosecond

LaSo SeSo

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Attosecond pulse generation at ELI-ALPS 100 kHz repetition rate beamline

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Phys. Rev. Research 2 (2020) 013338

https://doi.org/10.1088/1361-6455/ab92bf

We generate attosecond pulse train (APT) in argon driven by the high repetition rate (HR) laser of the Extreme Light Infrastructure – Attosecond Light Pulse Source (ELI-ALPS), providing 100 kHz, 80 W, 1030 nm, 40 fs pulses from a fiber chirped-pulse amplification (fiber-CPA) laser system. Under the current operating conditions of the high harmonic generation beamline (HR-GHHG), we observed the average pulse duration to be 395 as measured using the technique of reconstruction of attosecond beating by interference of two-photon transitions. The beamline uses an annular-shape laser beam so that the main part of the driving laser co-propagating with the APT can be eliminated by reflection on a holey mirror. An additional 100 nm aluminum foil is used to filter out the remaining laser and the low order harmonics, allowing 2 pJ APT with a bandwidth from 25 eV to 50 eV to be transported to the target position where the APT interacts with matter. The implementation of the HR-GHHG beamline in ELI-ALPS delivering attosecond pulse trains at 100 kHz paves the way for time-resolved experiments in the infrastructure, especially those that involve rare events and coincidence analysis, both of which need high statistics.

eli attosecond

Laser-induced inner-shell excitations through direct electron re-collision versus indirect collision

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Optics Express
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The dynamics and the decay processes of inner-shell excited atoms are of great interest in physics, chemistry, biology, and technology. The highly excited state decays very quickly through different channels, both radiative and non-radiative. It is therefore a long-standing goal to study such dynamics directly in the time domain. Using few-cycle infrared laser pulses, we investigated the excitation and ionization of inner-shell electrons through laser-induced electron re-collision with the original parent ions and measured the dependence of the emitted x-ray spectra on the intensity and ellipticity of the driving laser. These directly re-colliding electrons can be used as the initiating pump step in pump/probe experiments for studying core-hole dynamics at their natural temporal scale. In our experiment we found that the dependence of the x-ray emission spectrum on the laser intensity and polarization state varies distinctly for the two kinds of atomic systems. Relying on our data and numerical simulations, we explain this behavior by the presence of different excitation mechanisms that are contributing in different ratios to the respective overall x-ray emission yields. Direct re-collision excitation competes with indirect collisions with neighboring atoms by electrons having “drifted away” from the original parent ion.

LaSo SeSo

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Carrier-envelope-phase measurement of few-cycle mid-infrared laser pulses using high harmonic generation in ZnO

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High-harmonic generation (HHG) in crystals offers a simple, affordable and easily accessible route to carrier-envelope phase (CEP) measurements, which scales favorably towards longer wavelengths. We present measurements of HHG in ZnO using few-cycle pulses at 3.1µm. Thanks to the broad bandwidth of the driving laser pulses, spectral overlap between adjacent harmonic orders is achieved. The resulting spectral interference pattern provides access to the relative harmonic phase, and hence, the CEP.

eli attosecond

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1 kHz laser accelerated electron beam feasible for radiotherapy uses: a PIC–Monte Carlo based study

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Laser wakefield acceleration (LWFA) offers a promising compact solution for the production of high and very high energy electron (VHEE) beams, which have an ultrashort pulse duration with a high instantaneous dose rate and small source size. These unique properties are of radiobiological as well as clinical interest. In this paper we focus on the potential application of high repetition rate LWFA electron beams for radiobiology and radiotherapy. On the basis of particle-in-cell (PIC) and Monte Carlo simulations we propose that, using a commercially available 1 kHz laser system one can generate electron beams with 35.7 MeV mean energy and 3 pC electron bunch charge at 1 kHz repetition rate to deliver a dose rate of 18 Gy/min, which could be extremely useful for real radiotherapy applications. Thanks to the high repetition rate, dose delivery can be performed with high precision making this system a potential alternative to conventional clinical electron accelerators.

SeSo US

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VINYL: The VIrtual Neutron and x-raY Laboratory and its applications

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Experiments conducted in large scientific research infrastructures, such as synchrotrons, free electron lasers and neutron sources become increasingly complex. Such experiments, often investigating complex physical systems, are usually performed under strict time limitations and may depend critically on experimental parameters. To prepare and analyze these complex experiments, a virtual laboratory which provides start-to-end simulation tools can help experimenters predict experimental results under real or close to real instrument conditions. As a part of the PaNOSC (Photon and Neutron Open Science Cloud) project, the VIrtual Neutron and x-raY Laboratory (VINYL) is designed to be a cloud service framework to implement start-to-end simulations for those scientific facilities. In this paper, we present an introduction of the virtual laboratory framework and discuss its applications to the design and optimization of experiment setups as well as the estimation of experimental artifacts in an X-ray experiment.

eli attosecond

Thickness of natural contaminant layers on metal surfaces and its effects on laser-driven ion acceleration

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In the laser-driven ion acceleration studies, the naturally deposited contaminant layer on the target surface is thought to be a source of energetic ions and protons. Using ellipsometric measurements, we found that the thickness of the surface natural contaminant layer, which cannot be modified without external surface treatment, is on the order of a few nanometers. A conceptual approach is developed where “thick” and “thin” contaminant layer regimes of acceleration are identified and parameterized by the normalized thickness of the contaminant layer. These studies may also help in developing an ion acceleration concept using multilayered targets or through modifications of the target surface.

LaSo US

Processing of optical glasses by single, 34 fs pulses in the strong field ionization domain: ablation characteristics and crater morphology

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We demonstrate how dynamic Stark control can be achieved on molecular photodissociation in the dipole limit, using single-cycle (full width at half maximum) laser pulses in the terahertz (THz) regime. As the laser-molecule interaction follows the instantaneous electric field through the permanent dipoles, the molecular potentials dynamically oscillate and so do the crossings between them. In this paper, we consider rotating-vibrating diatomic molecules (two-dimensional description) and reveal the interplay between the dissociating wave packet and the dynamically fluctuating crossing seam located in the configuration space of the molecules spanned by the R vibrational and θ rotational coordinates. Our showcase example is the widely studied lithium fluoride molecule for which the two lowest Σ states are nonadiabatically coupled at an avoided crossing (AC); furthermore a low-lying pure repulsive Π state is energetically close. Optical pumping of the system in the ground state thus results in two dissociation channels: one indirect route via the AC in the ground Σ state and one direct path in the Π state. We show that applying THz control pulses with specific time delays relative to the pumping can significantly alter the population dynamics, as well as the kinetic energy and angular distribution of the photofragments.

eli attosecond

Time integrated transient reflectivity versus ablation characteristics of Borofloat, BK7, and B270 optical glasses ablated by 34 fs pulses

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Ablation and plasma mirror characteristics of Borofloat, BK7, and B270 glasses processed with 34 fs pulses of 800 nm central wavelength are compared in the 10^{14}–10^{15} W/cm^2 intensity domain. With thresholds of 1.7-1.9 × 10^{14} W/cm^2, higher than those of fused silica, and depths saturating above 5×10^{14} W/cm^2, the three glasses behave similarly from the point of view of ablation. With reflectivity enhancements comparing favorably with that of fused silica, the glasses prove to be good plasma mirror hosts. With the steepest increase in time integrated transient reflectivity with intensity, Borofloat is the most promising candidate.

Phase-controlled, second harmonic optimized THz pulse generation in nitrogen by infrared two-color laser pulses

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Broadband terahertz radiation can be efficiently produced by mixing laser pulses of different colors in the mid-infrared (MIR) and longwave -infrared (LWIR) spectral region. In this paper, we report on a numerical investigation of ultrashort terahertz pulse generation from plasmas created in nitrogen gas by two-color laser pulses with the fundamental laser pulse wavelength between 2.15 and 15.15 µm, in order to explore the efficiency of the terahertz pulse generation process. The results show that the electron acceleration efficiency increases monotonically with the fundamental laser pulse wavelength. The most intense terahertz pulse generation is observed at 12.30 µm with four optical-cycle laser pulses with 2.5 GW peak power. The results show that the terahertz pulse generation with a MIR laser is one order of magnitude and with a LWIR laser is two orders of magnitude more efficient than the terahertz pulse generation with Ti:Sapphire lasers using the exact same pulse parameters. The terahertz pulse generation efficiency is also known to be very sensitive to the relative phase between the components of the two-color laser pulses. One of the most useful tools to control the relative phase and optimize the terahertz pulse intensity is thin dielectric plates. It has been shown that alkaline halides and alkaline earth halides have suitable optical properties for the relative phase control for efficient terahertz pulse generation in the MIR spectral range.

Formation of CN radical from nitrogen and carbon condensation and from photodissociation in femtosecond laser-induced plasmas: time-resolved FT-UV–Vis spectroscopic study of the violet emission of CN radical

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Exploring the formation of diatomic radicals in femtosecond plasmas is important to establish the most dominant kinetic pathways following ionization and dissociation of small molecules. In this work, cyano radical formation has been studied from bromoform, acetonitrile, and methanol in nitrogen and argon plasmas created with a focused femtosecond laser beam operating at 100 kHz repetition rate and 1030 nm wavelength with 43 fs pulse length and 250 μJ pulse energy. Time-resolved Fourier transform fluorescence spectroscopy was applied in the ultraviolet–visible (UV–vis) spectral range for the characterization of the rotational and vibrational temperatures of the CN(B) radicals via fitting the experimental data. The high repetition rate of the laser allows efficient coupling with the step-scan Fourier transform spectroscopy method. Coulomb explosion at the very high intensity (~10^{16} W/cm^2) resulted in the formation of nascent atoms, ions, and electrons. The condensation reactions of carbon and reactive nitrogen species resulted in the formation of CN($B^2\Sigma^+$) radicals and $C_2(d^3\Pi_g)$ dicarbon molecules/radicals. The CN(B) radicals were formed at the highest concentration in the case of bromoform because the weak carbon–bromine bonds resulted in reactive carbon atoms and CH radicals, which are reactive precursors for the CN(B) radical formation. In the case of acetonitrile, immediate production of CN(B) is observed with nanosecond resolution, which suggests that the CN is formed either via photodetachment or via roaming reaction associated with the Coulomb explosion of the parent molecule. The nascent rotational temperature was very high (~6000–8500 K) and rapidly decreased in all instances within 40 ns with bromoform and acetonitrile. The highest vibrational temperature (~7800 K) was observed in an acetonitrile/Ar mixture that decreased in about 30 ns and then increased in the observed time window. The vibrational temperature increased in all samples between 30 and 200 ns. The time dependence of fluorescence is described with a monoexponential decay in the case of acetonitrile/Ar and with biexponential decays in all other instances in the 0–250 mbar total pressure range. The shorter time constant is close to the radiative lifetime of CN(B) emission (~60–80 ns), which can be attributed to the CN(B) radicals produced in the first few collisions at lower pressures. The longer CN(B) emission is from CN(B) created by slower chemical reactions involving carbon atoms, C_2 radicals, and reactive nitrogen-containing species.