ABSTRACTS

ELI ALPS | 2023

A comparative study of femtosecond pulsed laser ablation of meloxicam in distilled water and in air

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The increasing prevalence of water insoluble or poorly soluble drugs calls for the development of new formulation methods. Common approaches include the reduction of particle size and degree of crystallinity. Pulsed laser ablation is a clean technique for producing sub-micrometre sized drug particles and has the potential to induce amorphization. We studied the effect of femtosecond pulsed laser ablation (ELI ALPS THz pump laser system: λ_c=781 nm, τ=135 fs) on meloxicam in distilled water and in air. The ablated particles were characterized chemically, morphologically and in terms of crystallinity. We demonstrated that femtosecond laser ablation can induce partial amorphization of the particles in addition to a reduction in particle size. In the case of femtosecond pulsed laser ablation in air, the formation of pure meloxicam spheres showed that this technique can produce amorphous meloxicam without the use of excipients, which is a unique result. We also aimed to describe the ablation processes in both investigated media.

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The petawatt laser of ELI ALPS: reaching the 700 TW level at 10 Hz repetition rate

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Performance of the novel high repetition rate HF-PW laser system of ELI ALPS is presented in its first operation phase at 400 TW and 700 TW levels. Long-term operation was tested at 2.5 and 10 Hz repetition rates, where an exceptional 0.66% and 1.08% shot-to-shot energy stability was demonstrated, respectively. Thorough spatio-spectral and temporal measurements confirmed high quality output pulses with a Strehl ratio of >0.9 after compression at both repetition rates. Amplified pulses with an unprecedentedly high 240 W average power were reached for the first time from a PW-class amplifier chain by using novel pseudo-active mirror disk amplification-based pump lasers.

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Single thin-plate compression of multi-TW laser pulses to 3.9 fs

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Post-compression of 12-fs laser pulses with multi-TW peak power from an optical parametric chirped pulse amplification (OPCPA) system was performed by using a single thin fused silica plate in a vacuum. By optimizing the input pulses in both spatial and temporal domains, after compression with customized chirped mirrors, we achieved pulses as short as 3.87 fs, in combination with 12-mJ energy. The spatio-spectral quality of the post-compressed pulses was thoroughly analyzed. The generated 1.4-cycle pulses pave the way for next generation attosecond and particle acceleration experiments.

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Spectrally tunable ultrashort monochromatized extreme ultraviolet pulses at 100 kHz

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We present the experimental realization of spectrally tunable, ultrashort, quasi-monochromatic extreme ultraviolet (XUV) pulses generated at 100 kHz repetition rate in a user-oriented gas high harmonic generation beamline of the Extreme Light Infrastructure—Attosecond Light Pulse Source facility. Versatile spectral and temporal shaping of the XUV pulses is accomplished with a double-grating, time-delay compensated monochromator accommodating the two composing stages in a novel, asymmetrical geometry. This configuration supports the achievement of high monochromatic XUV flux ($2.8 \pm 0.9 \times 10^{10}$ photons/s at 39.7 eV selected with 700 meV full width at half maximum bandwidth) combined with ultrashort pulse duration (4.0 ± 0.2 fs using 12.1 ± 0.6 fs driving pulses) and small spot size (sub-100 μm). Focusability, spectral bandwidth, and overall photon flux of the produced radiation were investigated, covering a wide range of instrumental configurations. Moreover, complete temporal (intensity and phase) characterization of the few-femtosecond monochromatic XUV pulses—a goal that is difficult to achieve by conventional reconstruction techniques—has been realized using a ptychographic algorithm on experimentally recorded XUV-infrared pump–probe traces. The presented results contribute to in situ, time-resolved experiments, accessing direct information on the electronic structure dynamics of novel target materials.

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Ion acceleration with few cycle relativistic laser pulses from foil targets

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Ion acceleration resulting from the interaction of 11 fs laser pulses of ~35 mJ energy with ultrahigh contrast (<10^{-10}) and 10^{19} W cm^{-2} peak intensity with foil targets made of various materials and thicknesses at normal (0°) and 45° laser incidence is investigated. The maximum energy of the protons reached 1.4 MeV accelerated in the laser propagation direction and 1.2 MeV in the opposite direction from a formvar target. The energy conversion efficiency from the laser to the proton beam is estimated to be as high as 1.4% at 45° laser incidence using a 51 nm thick Al target. The high laser contrast indicates the predominance of vacuum heating via Brunel's effect as an absorption mechanism involving a tiny pre-plasma at the target front. The experimental results are in reasonable agreement with theoretical estimates, where proton acceleration from the target front side in the backward direction is well explained by the Coulomb explosion of a charged cavity formed in a tiny pre-plasma, while forward proton acceleration is likely to be a two-step process: protons are first accelerated in the target front-side cavity and then further boosted in energy through the target back side via the target normal sheath acceleration (TNSA) mechanism.

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High-order harmonic generation in a strongly overdriven regime

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High-order harmonic generation (HHG) normally requires a careful adjustment of the driving laser intensity (typically 10^{14}–10^{15} W/cm^2) and gas medium parameters to obtain a microscopically and macroscopically optimized output. In contrast to conventional wisdom, we present experimental results indicating efficient HHG in all rare gases, using a high-density medium and a driving laser intensity of around 10^{16} W/cm^2. The experimental results are corroborated by theoretical simulations, which indicate that ionization-induced self-phase modulation and plasma defocusing self-regulate the driver laser intensity to a level that allows efficient HHG. A tenfold broadening of the driving near-infrared spectrum is observed, which results in the generation of continuous spectra from 18 to 140 eV in spite of using 50-fs-long driving pulses. The presented scheme represents a simple and versatile concept for the generation of extreme-ultraviolet and soft-x-ray continua, which could be used for transient absorption and reflection spectroscopy.

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Time resolved investigation of high repetition rate gas jet target for high harmonic generation

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High-repetition-rate gas targets constitute an essential component in intense laser matter interaction studies. The technology becomes challenging as the repetition rate approaches the kilohertz regime. In this regime, cantilever-based gas valves are employed, which can open and close in tens of microseconds, resulting in a unique kind of gas characteristics in both the spatial and temporal domain. Here we characterize piezo cantilever-based kilohertz pulsed gas valves in the low density regime, where it provides sufficient peak gas density for high-harmonic generation while releasing a significantly smaller amount of gas reducing the vacuum load within the interaction chamber, suitable for high-vacuum applications. In order to obtain reliable information of the gas density in the target jet, space-time resolved characterization is performed. The gas-jet system is validated by conducting interferometric gas density estimations and high-harmonic generation measurements at the Extreme Light Infrastructure Attosecond Light Pulse Source facility. Our results demonstrate that while employing such targets for optimal high-harmonic generation, the high intensity interaction should be confined to a suitable time window, after the cantilever opening. The measured gas density evolution correlates well with the integrated high-harmonic flux and state-of-the-art three-dimensional simulation results, establishing the importance of such metrology.

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Two phase-matching regimes in high-order harmonic generation

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High-order harmonic generation (HHG) provides scalable sources of coherent extreme ultraviolet radiation with pulse duration down to the attosecond time scale. Efficient HHG requires the constructive interplay between microscopic and macroscopic effects in the generation volume, which can be achieved over a large range of experimental parameters from the driving field properties to those of the generating medium. Here, we present a systematic study of the harmonic yield as a function of gas pressure and medium length. Two regimes for optimum yield are identified, supporting the predictions of a recently proposed analytical model. Our observations are independent on the focusing geometry and, to a large extent, on the pulse duration and laser intensity, providing a versatile approach to HHG optimization.

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Attosecond coherent control of electronic wave packets in two-colour photoionization using a novel timing tool for seeded free-electron laser

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In ultrafast spectroscopy, the temporal resolution of time-resolved experiments depends on the duration of the pump and probe pulses, and on the control and characterization of their relative synchronization. Free-electron lasers operating in the extreme ultraviolet and X-ray spectral regions deliver pulses with femtosecond and attosecond duration in a broad array of pump–probe configurations to study a wide range of physical processes. However, this flexibility, together with the large dimensions and high complexity of the experimental set-ups, limits control of the temporal delay to the femtosecond domain, thus precluding a time resolution below the optical cycle. Here we demonstrate a novel single-shot technique able to determine the relative synchronization between an attosecond pulse train—generated by a seeded free-electron laser—and the optical oscillations of a near-infrared field, with a resolution of one atomic unit (24 as). Using this attosecond timing tool, we report the first example of attosecond coherent control of photoionization in a two-colour field by manipulating the phase of high-order near-infrared transitions.

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Quantum electrodynamics of ultra-intense laser-matter interactions

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Intense laser-matter interactions are at the center of interest in research and technology since the development of high-power lasers. They have been widely used for fundamental studies in atomic, molecular, and optical physics, and they are at the core of attosecond physics and ultrafast optoelectronics. Although the majority of these studies have been successfully described using classical electromagnetic fields, recent investigations based on fully quantized approaches have shown that intense laser-atom interactions can be used for the generation of controllable high-photon-number entangled coherent states and coherent state superpositions. In this tutorial, we provide a comprehensive fully quantized description of intense laser-atom interactions. We elaborate on the processes of high-harmonic generation, above-threshold ionization, and we discuss new phenomena that cannot be revealed within the context of semiclassical theories. We provide the description for conditioning the light field on different electronic processes, and their consequences for quantum state engineering of light. Finally, we discuss the extension of the approach to more complex materials, and the impact to quantum technologies for a new photonic platform composed of the symbiosis of attosecond physics and quantum information science.

SeSo

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Strong–laser–field physics, non–classical light states and quantum information science

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Strong–laser–field physics is a research direction that relies on the use of high-power lasers and has led to fascinating achievements ranging from relativistic particle acceleration to attosecond science. On the other hand, quantum optics has been built on the use of low photon number sources and has opened the way for groundbreaking discoveries in quantum technology, advancing investigations ranging from fundamental tests of quantum theory to quantum information processing. Despite the tremendous progress, until recently these directions have remained disconnected. This is because the majority of the interactions in the strong-field limit have been successfully described by semi-classical approximations treating the electromagnetic field classically, as there was no need to include the quantum properties of the field to explain the observations. The link between strong–laser–field physics, quantum optics, and quantum information science has been developed in the recent past. Studies based on fully quantized and conditioning approaches have shown that intense laser–matter interactions can be used for the generation of controllable entangled and non-classical light states. These achievements open the way for a vast number of investigations stemming from the symbiosis of strong–laser–field physics, quantum optics, and quantum information science. Here, after an introduction to the fundamentals of these research directions, we report on the recent progress in the fully quantized description of intense laser–matter interaction and the methods that have been developed for the generation of non-classical light states and entangled states. Also, we discuss the future directions of non-classical light engineering using strong laser fields, and the potential applications in ultrafast and quantum information science.

SeSo

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Generation of micro-Joule level coherent quasi-continuum extreme ultraviolet radiation using multi-cycle intense laser-atom interactions

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In the present work we report on the current progress of the recently constructed GW attosecond extreme ultraviolet (XUV) source developed at the Institute of Electronic Structure and Laser of the Foundation for Research and Technology-Hellas (I.E.S.L-FO.R.T.H.). By the implementation of a compact-collinear polarization gating arrangement, the generation of a broadband, coherent XUV quasi-continuum produced by the interaction of a many-cycle infrared field with a gas phase medium is achieved. The spectral width of the XUV emission generated in Xenon, is spanning in the range of 17–32 eV and can support isolated pulses of duration in the range from 0.4 fs to 1.3 fs and pulse energy in the 1 μJ level. Theoretical calculations, taking into account the experimental conditions of this work, are supporting the observations, offering also an insight regarding the temporal profile of the emitted radiation. Finally, the high intensity of the produced XUV pulses has been confirmed by investigating the two-XUV-photon double ionization process of Argon atoms. The demonstrated results inaugurate the capability of the beamline of producing intense coherent quasi-continuum XUV radiation, supporting isolated as pulses, that can be exploited in studies of non-linear XUV processes, attosecond pulse metrology and XUV-pump–XUV-probe experiments.

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Numerical representation of tightly focused ultra-short laser pulses

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A scheme for numerical representation for the fields of tightly focused laser pulses is presented going beyond the paraxial framework that gives accurate values up to near single-cycle pulses. The laser field is defined by a solution of the paraxial Helmholtz equation and a temporal pulse profile in light-cone coordinates, the fourth- and second-order corrections are provided for short pulses and vector beams, and the absolute error of these terms is presented. The method can give an accurate field description for any transverse electromagnetic mode, and it is suited for providing initial fields for existing solvers for Maxwell's equations, such as in the particle-in-cell codes.

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Hybrid acceleration of compact ion bunches by few-cycle laser pulses in gas jets of two atomic species

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Extreme states of matter that exist for a short time need probing with compact proton sources. Here, the generation of a compact MeV-energy proton source from few-cycle laser pulse interaction with narrow gas-jet target is demonstrated numerically. We realize such proton source by incorporating a gas mixture which is optimal for generation of quasimonoenergetic proton bunches. In the presented laser-plasma interaction we identify the ion acceleration from magnetic vortex, charge separation, and collisionless shock wave. The proposed particle source is excellent for applications where moderate-energy proton bunches are used at high repetition rate. We prove the applicability of this scheme for generating a pulsed spherical neutron burst with ~100 ps duration.

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Three-stage laser wakefield accelerator scheme for sub-Joule few-cycle laser pulses

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Laser-driven electron acceleration in underdense plasma is a promising route towards the realization of reliable sources of relativistic electrons in the 0.1–1 GeV energy range. Generation of such electron bunches at high repetition rates is hindered by the limited energy per pulse, which inevitably results in very short pulse duration and tight focusing. Compressing the laser energy in time and space allows scientists to use higher plasma density to drive wakefieds, which in turn results in enhanced diffraction and dispersion of the broadband laser pulse. These features make difficult to control the acceleration in the plasma wave and to improve the beam quality. Here we propose a mm-long three-stage acceleration scheme, which allows for tunable injection and optimal acceleration of high-quality electron bunches. The full interaction length is modeled by 3D particle-in-cell simulations.

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Controlled transition to different proton acceleration regimes: Near-critical-density plasmas driven by circularly polarized few-cycle pulses

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A controlled transition between two different ion acceleration mechanisms would pave the way to achieving different ion energies and spectral features within the same experimental set up, depending on the region of operation. Based on numerical simulations conducted over a wide range of experimentally achievable parameter space, reported here is a comprehensive investigation of the different facets of ion acceleration by relativistically intense circularly polarized laser pulses interacting with thin near-critical-density plasma targets. The results show that the plasma thickness, exponential density gradient, and laser frequency chirp can be controlled to switch the interaction from the transparent operating regime to the opaque one, thereby enabling the choice of a Maxwellian-like ion energy distribution with a cutoff energy in the relativistically transparent regime or a quasi-monoenergetic spectrum in the opaque regime. Next, it is established that a multispecies target configuration can be used effectively for optimal generation of quasi-monoenergetic ion bunches of a desired species. Finally, the feasibility is demonstrated for generating monoenergetic proton beams with energy peak at E≈20–40 MeV and a narrow energy spread of ΔE/E≈18%–28.6% confined within a divergence angle of ~175 mrad at a reasonable laser peak intensity of $I_0 \approx 5.4 \times 10^{20}$ W/cm^2.

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Energetic, tunable, highly elliptically polarized higher harmonics generated by intense two-color counter-rotating laser fields

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In this work, we demonstrate experimentally the efficient generation and tunability of energetic highly elliptical high harmonics in Ar gas, driven by intense two-color counter-rotating laser electric fields. A bichromatic beam tailored by a Mach-Zehnder-Less for Threefold Optical Virginia spiderwort (MAZEL-TOV) apparatus generates high-order harmonic generation (HHG), where the output spectrum of the highly elliptical HHG radiation can be tuned for an energy range of ΔE≈150 meV in the spectral range of-20 eV with energy per pulse E^{XUV}≈400 nJ at the source. Furthermore, we employ time-dependent density-functional simulations to probe the dependence of the harmonic ellipticity and the strength of the attosecond pulses on the driving-field parameters and demonstrate the robustness of the HHG with the bichromatic field. We show how, by properly tuning the central frequency of the second harmonic, the central frequency of the extreme ultraviolet (XUV) high-harmonic radiation is continuously tuned. The demonstrated energy values largely exceed the output energy from many other laser-driven attosecond sources reported so far and prove to be sufficient for inducing nonlinear processes in an atomic system. We envisage that such tunable energetic highly elliptical HHG spectra can remove the facility restrictions from requirements of few-cycle driving pulses for isolated circular attosecond-pulse generation.

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Subcycle surface electron emission driven by strong-field terahertz waveforms

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The advent of intense terahertz (THz) sources opened a new era when the demonstration of the acceleration and manipulation of free electrons by THz pulses became within reach. THz-field-driven electron emission was predicted to be confined to a single burst due to the single-cycle waveform. Here we demonstrate the confinement of single-cycle THz-waveform-driven electron emission to one of the two half cycles from a solid surface emitter. Either the leading or the trailing half cycle was active, controlled by reversing the field polarity. THz-driven single-burst surface electron emission sources, which do not rely on field-enhancement structures, will impact the development of THz-powered electron acceleration and manipulation devices, all THz compact electron sources, THz waveguides and telecommunication, THz-field-based measurement techniques and solid-state devices.

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Photo-ionization initiated differential ultrafast charge migration: impact of molecular symmetries and tautomeric forms

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Photo-ionization induced ultrafast electron dynamics is considered as a precursor for the slower nuclear dynamics associated with molecular dissociation. Here, using the ab initio multielectron wave-packet propagation method, we study the overall many-electron dynamics, triggered by ionizing the outer-valence orbitals of different tautomers for a prototype molecule with more than one symmetry element. From the time evolution of the initially created averaged hole density of each system, we identify distinctly different charge dynamics responses in the tautomers. We observe that the keto form shows a charge migration direction away from the nitrogen bonded with hydrogen, while in enol–U – away from oxygen bonded to hydrogen. Additionally, the dynamics following the ionization of molecular orbitals with different symmetries reveals that a′ orbitals show a fast and highly delocalized charge density in comparison to a′′ symmetry. These observations indicate why different tautomers respond differently to an XUV ionization,and might explain the subsequent different fragmentation pathways. An experimental schematics allowing the detection and reconstruction of such charge dynamics is also proposed. Although the present study uses a simple, prototypical bio-relevant molecule, it reveals the explicit role of molecular symmetry and tautomerism in the ionization-triggered charge migration that controls many ultrafast physical, chemical, and biological processes, making tautomeric forms a promising tool of molecular design for desired charge migration.

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Atomic scale interfacial magnetism and origin of metal-insulator transition in $(LaNiO_3)_n/(CaMnO_3)_m$ superlattices: a first principles study

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Interfacial magnetism and metal-insulator transition at $LaNiO_3$-based oxide interfaces have triggered intense research efforts, because of the possible implications in future heterostructure device design and engineering. Experimental observation lack in some points a support from an atomistic view. In an effort to fill such gap, we hereby investigate the structural, electronic, and magnetic properties of $(LaNiO_3)_n/(CaMnO_3)_m$ superlattices with varying $LaNiO_3$ thickness (n) using density functional theory including a Hubbard-type effective on-site Coulomb term. We successfully capture and explain the metal-insulator transition and interfacial magnetic properties, such as magnetic alignments and induced Ni magnetic moments which were recently observed experimentally in nickelate-based heterostructures. In the superlattices modeled in our study, an insulating state is found for n=1 and a metallic character for n=2, 4, with major contribution from Ni and Mn 3d states. The insulating character originates from the disorder effect induced by sudden environment change for the octahedra at the interface, and associated to localized electronic states; on the other hand, for larger n, less localized interfacial states and increased polarity of the $LaNiO_3$ layers contribute to metallicity. We discuss how the interplay between double and super-exchange interaction via complex structural and charge redistributions results in interfacial magnetism. While $(LaNiO_3)_n/(CaMnO_3)_m$ superlattices are chosen as prototype and for their experimental feasibility, our approach is generally applicable to understand the intricate roles of interfacial states and exchange mechanism between magnetic ions towards the overall response of a magnetic interface or superlattice.

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Inorganic drugs as a tool for protein structure solving and studies on conformational changes

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Inorganic drugs are capable of tight interactions with proteins through coordination towards aminoacidic residues, and this feature is recognized as a key aspect for their pharmacological action. However, the “protein metalation process” is exploitable for solving the phase problem and structural resolution. In fact, the use of inorganic drugs bearing specific metal centers and ligands capable to drive the binding towards the desired portions of the protein target could represent a very intriguing and fruitful strategy. In this context, a theoretical approach may further contribute to solve protein structures and their refinement. Here, we delineate the main features of a reliable experimental-theoretical integrated approach, based on the use of metallodrugs, for protein structure solving.

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Chemistry Europe
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Effect of reduced graphene oxide hybridization on ZnO nanoparticles sensitivity to NO_2 gas: A DFT study

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In the present work, a density functional theory (DFT) calculation to simulate reduced graphene oxide (rGO) hybrid with zinc oxide (ZnO) nanoparticle's sensitivity to NO_2 gas is performed. In comparison with the experiment, DFT calculations give acceptable results to available bond lengths, lattice parameters, X-ray photoelectron spectroscopy (XPS), energy gaps, Gibbs free energy, enthalpy, entropy, etc. to ZnO, rGO, and ZnO/rGO hybrid. ZnO and rGO show n-type and p-type semiconductor behavior, respectively. The formed p-n heterojunction between rGO and ZnO is of the staggering gap type. Results show that rGO increases the sensitivity of ZnO to NO_2 gas as they form a hybrid. ZnO/rGO hybrid has a higher number of vacancies that can be used to attract oxygen atoms from NO_2 and change the resistivity of the hybrid. The combined reduction of oxygen from NO_2 and NO can give a very high value of the Gibbs free energy of reaction that explains the ppb level sensitivity of the ZnO/rGO hybrid. The dissociation of NO_2 in the air reduces the sensitivity of the ZnO/rGO hybrid at temperatures higher than 300˚C.

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Enhancing the strength, electrical properties and versatility of epoxy polymer composite foam with carbon nanostructures

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Epoxy resins (epoxy), also known as polyepoxides, are an extensive class of reactive polymers containing epoxide groups, and a very popular material is used in various applications such as marine, automotive, aircraft and aerospace architectures and electronic applications. However, epoxy is brittle and has high coefficient of thermal expansion, poor mechanical strength and electrical conductivity. Incorporating nano and micro filler in epoxy is an established approach to enhance all these properties. However, obviously also depends on the properties of the filler. In this mini review article, we will briefly describe how carbon nanomaterials, depending on their intrinsic structure and properties, can affect the epoxy composite foams, in terms of their fabrication methods, resulting mechanical, dielectric, and physical properties.

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Journal of Polymer Science
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Prospective utilization of boron nitride and beryllium oxide nanotubes for Na, Li, and K-ion batteries: a DFT-based analysis

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Context
In the present work, we investigated the adsorption mechanism of natural sodium (Na), potassium (K), and lithium (Li) atoms and their respective ion on two nanostructures: boron-nitride nanotubes (BNNTs) and beryllium-oxide nanotubes (BeONTs). The main goal of this research is to calculate the gain voltage for Na, K, and Li ionic batteries. Density function theory (DFT) calculations indicated that the adsorption energy between Na+is higher than that of the other cations, and this is particularly clear in the BeONT. Furthermore, gain voltage calculations showed that BNNTs generate a higher potential than BeONTs, with the most significant difference observed in BNNT/Na+. This research provides theoretical insights into the potential uses of these nanostructures as anodes in Na, K, and Li-ion batteries.

Method
Density function theory used to compute the ground state properties for BeONT and BNNT with and without selected atoms and their ions (Li, K, and Na). B3LYP used for exchange correlation between electrons and ions, and 6-31G* basis set used for all atoms and ions. Gauss Sum 2.2 software used for estimate the density of state (DOS) for all structure under investigation.

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Holy Water: Photo-brightening in quasi-2D perovskite films under ambient enables highly performing light-emitting diodes

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Quasi-2D perovskites provide new opportunities for lighting and display applications due to their high radiative recombination and excellent stability. However, seldom attention has been placed on their self-stability/working operation under ambient storage. Herein, quasi-2D perovskites/Polyethylene oxide (PEO) films are studied, showing an unforeseen photo-brightening effect under ambient storage (i.e., an increase of the photoluminescence quantum yield from 55% to 74% after 100 days). In stark contrast, those stored under a dark/inert atmosphere show a significant decrease down to 38%. This counterintuitive phenomenon responds to the increasing radiative recombination rate caused by the passivation of the surface Br vacancies in the presence of physically adsorbed water molecules, as corroborated by in situ/ex situ X-ray photoelectron spectroscopy and density functional theory calculations. Capitalizing on this surprising effect, stable light-emitting diodes (LEDs) using quasi-2D perovskites/PEO color filters are fabricated, realizing high stabilities of ≈400 h@10 mA under operating ambient conditions, representing a 20-fold enhancement compared to LEDs with 3D counter partners. Hence, this study reveals a unique insight into the impact of water passivation on the optical/structural properties of quasi-2D perovskite films, broadening their applications under operating ambient conditions.

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Decisive role of Cu/Co interfaces in copper cobaltite derivatives for high performance CO_2 methanation catalyst

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Thermo-catalytic bio-SNG (CH_4) production is one of the useful tools for converting waste to gaseous fuels through CO_2 conversion. To abundant properly, however, efficient, robust and cost-effective catalysts would be required. Bimetallic systems based on transition metals seem to be promising candidates for this task. The CoCu bimetallic system with in-situ generated interfaces was synthesized and used as a catalyst for CO_2 methanation. The in-depth analysis of the structure-activity-selectivity relationships involving XRD, (NAP-)XPS, EXAFS and TEM-EDX revealed that the co-existence of Co^0, CoO, and Cu^0 in the proper distribution on the surface can ensure the selective production of methane. To fine-tune the surface composition of the bimetallic systems, a systematic alteration of the Cu:Co ratio in the precursor spinel structures must be performed. $Cu_{0.4}Co_{2.6}O_4$ derivative, stabilizing subsurface Cu(I)–O specimen, showed the best performance with high activity (12,800 nmol g^{-1} s^{-1}) and a remarkable selectivity of 65–85% for methane in a wide temperature range (250–425 °C). In studying the mechanistic aspects of methanation, it has been shown that the hydrogenation of active carbon at the surface or below the surface is the key step for the production of methane. So far, this cobalt-catalyzed sub-step has been proposed in catalytic Fischer-Tropsch syntheses.

US

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New insights into thermal processes of metal deposits on h-BN/Rh(1 1 1): A comparison of Au and Rh

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In this paper the thermal properties of Au and Rh deposits are compared on hexagonal boron nitride (h-BN) “nanomesh” prepared on Rh(111), applying STM, XPS, low energy ion scattering (LEIS), LEED, and DFT. At room temperature, both metals essentially follow Volmer-Weber (3D) growth. Upon subsequent annealing, agglomeration (sintering), intercalation, and desorption are competing surface processes for both metals. For the more reactive Rh, we suggest an additional encapsulation mechanism: between 600 K and 750 K, fragments of decomposed h-BN diffuse locally from the bottom onto the metal clusters covering them partially. STM data indicates that agglomeration of gold nanoparticles proceeds faster compared to rhodium. At higher temperatures (1050 K–1100 K), all non-desorbing gold atoms diffuse below h-BN, even for large initial coverages. On the other hand, for larger Rh deposits (≥5 ML), the outermost layer always contains Rh. Accumulation of gold at the interface between h-BN and Rh(111) significantly enhances the thermal stability of h-BN, attributed to the lower reactivity of Au in the decomposition of h-BN compared to Rh(111). At elevated substrate temperatures, intercalation of individual adatoms takes place during deposition, which requires higher temperatures for rhodium due to its slower diffusion and higher probability of nucleation.

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Applied Surface Science 623 (2023) 157041

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Noble metals-deposited TiO_2 photocatalysts for photoreduction of CO_2: Exploration of surface chemistry and a reflection on the importance of wavelength dependence

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We report the enhanced photocatalytic performance of CO_2 reduction by introducing noble metal-deposited TiO_2 photocatalysts (NMD/TiO_2) under both visible light irradiation (VLI) and UV. It was found that NMD-TiO_2 samples demonstrated an expanded ability for light absorption, resulting in an improved photoefficiency for CO_2 reduction and selectivity towards carbon monoxide (CO). The conversion yields of CO_2 to CO followed the order: Rh >Ru >Pt and with Rh/TiO_2 reached 10.65 µmol/gcat (under visible light) and 7.84 µmol/gcat (under UV), which are ~3 and 10 times more as compared to pristine anatase TiO_2 (0, and 2.43 µmol/gcat). Various factors were addressed including theoretical studies via DFT revealed that the formation of new impurity energy state levels in NMD/TiO_2, which act as acceptor levels. Moreover, DRIFTS studies showed that the H-O-C-O and formyl (H_2CO) species were the important intermediates towards the hydrogenation of CO_2 to CO. Also, a charge transfer mechanism toward CO_2 reduction was elucidated.

US

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Development of magnetizable, nickel–ferrite-decorated carbon nanocomposites as hydrogenation catalyst for aniline synthesis

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Catalysts with magnetic properties can be easily recovered from the reaction medium without loss by using a magnetic field, which highly improves their applicability. To design such systems, we have successfully combined the magnetic properties of nickel ferrite nanoparticles with the positive properties of carbon-based catalyst supports. Amine-functionalized NiFe2O4 nanoparticles were deposited on the surfaces of nitrogen-doped bamboo-like carbon nanotubes (N-BCNT) and carbon nanolayers (CNL) by using a coprecipitation process. The magnetizable catalyst supports were decorated by Pd nanoparticles, and their catalytic activity was tested through the hydrogenation of nitrobenzene (NB). By using the prepared catalysts, high nitrobenzene conversion (100% for 120 min at 333 K) and a high aniline yield (99%) were achieved. The Pd/$NiFe_2O_4$-CNL catalyst was remarkable in terms of stability during the reuse tests due to the strong interaction formed between the catalytically active metal and its support (the activity was retained during four cycles of 120 min at 333 K). Furthermore, despite the long-lasting mechanical stress, no significant palladium loss (only 0.08 wt%) was detected.

US

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Pt/MnO interface induced defects for high reverse water gas shift activity

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The implementation of supported metal catalysts heavily relies on the synergistic interactions between metal nanoparticles and the material they are dispersed on. It is clear that interfacial perimeter sites have outstanding skills for turning catalytic reactions over, however, high activity and selectivity of the designed interface-induced metal distortion can also obtain catalysts for the most crucial industrial processes as evidenced in this paper. Herein, the beneficial synergy established between designed Pt nanoparticles and MnO in the course of the reverse water gas shift (RWGS) reaction resulted in a Pt/MnO catalyst having ≈10 times higher activity compared to the reference Pt/SBA-15 catalyst with >99% CO selectivity. Under activation, a crystal assembly through the metallic Pt (110) and MnO evolved, where the plane distance differences caused a mismatched-row structure in softer Pt nanoparticles, which was identified by microscopic and surface-sensitive spectroscopic characterizations combined with density functional theory simulations. The generated edge dislocations caused the Pt lattice expansion which led to the weakening of the Pt–CO bond. Even though MnO also exhibited an adverse effect on Pt by lowering the number of exposed metal sites, rapid desorption of the linearly adsorbed CO species governed the performance of the Pt/MnO in the RWGS.

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Local hydrophobicity allows high-performance electrochemical carbon monoxide reduction to C_{2+} products

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While CO can already be produced at industrially relevant current densities via CO_2 electrolysis, the selective formation of C_{2+} products seems challenging. CO electrolysis, in principle, can overcome this barrier, hence forming valuable chemicals from CO_2 in two steps. Here we demonstrate that a mass-produced, commercially available polymeric pore sealer can be used as a catalyst binder, ensuring high rate and selective CO reduction. We achieved above 70% faradaic efficiency for C_{2+} products formation at j = 500 mA cm^{-2} current density. As no specific interaction between the polymer and the CO reactant was found, we attribute the stable and selective operation of the electrolyzer cell to the controlled wetting of the catalyst layer due to the homogeneous polymer coating on the catalyst particles' surface. These results indicate that sophistically designed surface modifiers are not necessarily required for CO electrolysis, but a simpler alternative can in some cases lead to the same reaction rate, selectivity and energy efficiency; hence the capital costs can be significantly decreased.

US

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Diatomic molecule in a strong infrared laser field: level-shifts and bond-length change due to laser-dressed Morse potential

S. Varró, Sz. Hack, G. Paragi, P. Földi, I. F. Barna, and A. Czirják

We present a general mathematical procedure to handle interactions described by a Morse potential in the presence of a strong harmonic excitation. We account for permanent and field-induced terms and their gradients in the dipole moment function, and we derive analytic formulae for the bond-length change and for the shifted energy eigenvalues of the vibrations, by using the Kramers–Henneberger frame. We apply these results to the important cases of H_2 and LiH, driven by a near- or mid-infrared laser in the 10^{13} W cm^{-2} intensity range.

US

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Dynamic interference in below-threshold ionization

A. Tóth, S. Borbély and A. Csehi

We study the temporal interference of photoelectrons emitted during the rising and falling edges of intense femtosecond laser pulses, that can ionize atoms via near-resonant transitions. Due to the near-resonant coupling with the laser pulse, the emerging atomic dressed states repel each other, giving rise to the Rabi splitting that is primarily controlled by the laser intensity and detuning. Our numerical and analytical analysis reveals that dynamic interference is observed when the Rabi splitting of the dressed states is maximized while the ionization of the atom remains small. We demonstrate that these two conditions are fulfilled when the atom is driven off resonantly within the one-Rabi-cycle regime at perturbative laser intensities. As a result, the single peak of the photoelectron spectrum found in the weak-field limit is replaced by a pronounced multipeak pattern.

US

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Physical Review A
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Role of dynamic Stark shifts in strong-field excitation and subsequent ionization

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The energy levels of atoms and molecules exposed to intense, short, high-frequency laser pulses undergo time-dependent distortion known as dynamic Stark shift (DSS). These level shifts are induced by the coupling with nonresonant (nonessential) states and follow the temporal intensity profile of the laser pulse. Owing to the different DSSs of the individual atomic levels, transient resonance suppressions and enhancements significantly modify the multiphoton ionization pathway, leaving clear fingerprints (asymmetry, splitting, and shifting) in the energy spectrum of emitted photoelectrons. Here we investigate this phenomenon by solving the time-dependent Schrödinger equation of the valence electron of Na and Li in increasing levels of complexity: (i) developing minimal models for two-photon transition and (2+1)-photon ionization, (ii) applying a spectral method without continuum-continuum couplings, and (iii) propagating the electron wave packet on a large spatial grid accounting for all possible couplings. We show that appropriately detuned transform limited pulses provide a high level of control in strong-field excitation, when the atomic levels are dynamically shifted. Furthermore, we demonstrate the role of DSSs in the multipeak structure of the Autler-Townes doublet, found when several Rabi oscillations are induced during the strong-field resonant ionization.

US

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The DRESDEN PLATFORM is a research hub for ultra-high dose rate radiobiology

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The recently observed FLASH effect describes the observation of normal tissue protection by ultra-high dose rates (UHDR), or dose delivery in a fraction of a second, at similar tumor-killing efficacy of conventional dose delivery and promises great benefits for radiotherapy patients. Dedicated studies are now necessary to define a robust set of dose application parameters for FLASH radiotherapy and to identify underlying mechanisms. These studies require particle accelerators with variable temporal dose application characteristics for numerous radiation qualities, equipped for preclinical radiobiological research. Here we present the DRESDEN PLATFORM, a research hub for ultra-high dose rate radiobiology. By uniting clinical and research accelerators with radiobiology infrastructure and know-how, the DRESDEN PLATFORM offers a unique environment for studying the FLASH effect. We introduce its experimental capabilities and demonstrate the platform's suitability for systematic investigation of FLASH by presenting results from a concerted in vivo radiobiology study with zebrafish embryos. The comparative pre-clinical study was conducted across one electron and two proton accelerator facilities, including an advanced laser-driven proton source applied for FLASH-relevant in vivo irradiations for the first time. The data show a protective effect of UHDR irradiation up to 105 Gy/s and suggests consistency of the protective effect even at escalated dose rates of 109 Gy/s. With the first clinical FLASH studies underway, research facilities like the DRESDEN PLATFORM, addressing the open questions surrounding FLASH, are essential to accelerate FLASH's translation into clinical practice.

US

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Carrier-envelope phase on-chip scanner and control of laser beams

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The carrier-envelope phase (CEP) is an important property of few-cycle laser pulses, allowing for light field control of electronic processes during laser-matter interactions. Thus, the measurement and control of CEP is essential for applications of few-cycle lasers. Currently, there is no robust method for measuring the non-trivial spatial CEP distribution of few-cycle laser pulses. Here, we demonstrate a compact on-chip, ambient-air, CEP scanning probe with 0.1 μm^3 resolution based on optical driving of CEP-sensitive ultrafast currents in a metal–dielectric heterostructure. We successfully apply the probe to obtain a 3D map of spatial changes of CEP in the vicinity of an oscillator beam focus with pulses as weak as 1 nJ. We also demonstrate CEP control in the focal volume with a spatial light modulator so that arbitrary spatial CEP sculpting could be realized.

US

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Nature Communications
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Tailoring surface topographies on solids with Mid-IR femtosecond laser pulses

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The recently observed FLASH effect describes the observation of normal tissue protection by ultra-high dose rates (UHDR), or dose delivery in a fraction of a second, at similar tumor-killing efficacy of conventional dose delivery and promises great benefits for radiotherapy patients. Dedicated studies are now necessary to define a robust set of dose application parameters for FLASH radiotherapy and to identify underlying mechanisms. These studies require particle accelerators with variable temporal dose application characteristics for numerous radiation qualities, equipped for preclinical radiobiological research. Here we present the DRESDEN PLATFORM, a research hub for ultra-high dose rate radiobiology. By uniting clinical and research accelerators with radiobiology infrastructure and know-how, the DRESDEN PLATFORM offers a unique environment for studying the FLASH effect. We introduce its experimental capabilities and demonstrate the platform's suitability for systematic investigation of FLASH by presenting results from a concerted in vivo radiobiology study with zebrafish embryos. The comparative pre-clinical study was conducted across one electron and two proton accelerator facilities, including an advanced laser-driven proton source applied for FLASH-relevant in vivo irradiations for the first time. The data show a protective effect of UHDR irradiation up to 105 Gy/s and suggests consistency of the protective effect even at escalated dose rates of 109 Gy/s. With the first clinical FLASH studies underway, research facilities like the DRESDEN PLATFORM, addressing the open questions surrounding FLASH, are essential to accelerate FLASH's translation into clinical practice.

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