ABSTRACTS

ELI ALPS

2022

attosecond

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Carrier-envelope phase stable few-cycle laser system delivering more than 100W, 1mJ, sub-2-cycle pulses

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Optics Letters
47 (2022) 6, 1537-1540

http://dx.doi.org/10.1364/OL.450991

Two-stage multipass-cell compression of a fiber–chirped-pulse amplifier system to the few-cycle regime is presented. The output delivers a sub-2-cycle (5.8 fs), 107 W average power, 1.07 mJ pulses at 100 kHz centered at 1030 nm with excellent spatial beam quality ($M^2 = 1.1$, Strehl ratio $S = 0.98$), pointing stability (2.3 µrad), and superior long-term average power stability of 0.1% STD over more than 8 hours. This is combined with a carrier-envelope phase stability of 360 mrad in the frequency range from 10 Hz to 50 kHz, i.e., measured on a single-shot basis. This unique system will serve as an HR1 laser for the Extreme Light Infrastructure Attosecond Light Pulse Source Research facility to enable high repetition rate isolated attosecond pulse generation.

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Sub-50 fs pulses at 2050 nm from a picosecond Ho:YLF laser using a two-stage Kagome-fiber-based compressor

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Optics Letters
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The high-energy few-cycle mid-infrared laser pulse beyond 2 µm is of immense importance for attosecond science and strong-field physics. However, the limited gain bandwidth of laser crystals such as Ho:YLF and Ho:YAG allows the generation of picosecond (ps) long pulses and, hence, makes it challenging to generate few-cycle pulse at 2 µm without utilizing an optical parametric chirped-pulse amplifier (OPCPA). Moreover, the exclusive use of the near-infrared wavelength has limited the generation of wavelengths beyond 4 µm (OPCPA). Furthermore, high harmonic generation (HHG) conversion efficiency reduces dramatically when driven by a long-wavelength laser. Novel schemes such as multi-color HHG have been proposed to enhance the harmonic flux. Therefore, it is highly desirable to generate few-cycle to femtosecond pulses from a 2 µm laser for driving these experiments. Here, we utilize two-stage nonlinear spectral broadening and pulse compression based on the Kagome-type hollow-core photonic crystal fiber (HC-PCF) to compress few-ps pulses to sub-50 fs from a Ho:YLF amplifier at 2 µm at 1 kHz repetition rate. We demonstrate both experimentally and numerically the compression of 3.3 ps at 140 µJ pulses to 48 fs at 11 µJ with focal intensity reaching 10^{13} W/cm^2. Thereby, this system can be used for driving HHG in solids at 2 µm. In the first stage, the pulses are spectrally broadened in Kagome fiber and compressed in a silicon-based prism compressor to 285 fs at a pulse energy of 90 µJ. In the second stage, the 285 fs pulse is self-compressed in air-filled HC-PCF. With fine-tuning of the group delay dispersion (GDD) externally in a 3 mm window, a compressed pulse of 48 fs is achieved. This leads to a 70-fold compression of the ps pulses at 2050 nm. We further used the sub-50 fs laser pulses to generate white light by focusing the pulse into a thin medium of YAG.

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Ultrafast Science
2022, 9823783

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High-flux 100-kHz attosecond pulse source driven by a high average power annular laser beam

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High-repetition rate attosecond pulse sources are indispensable tools for time-resolved studies of electron dynamics, such as coincidence spectroscopy and experiments with high demands on statistics or signal-to-noise ratio, especially in the case of solid and big molecule samples in chemistry and biology. Although with the high-repetition rate lasers, such attosecond pulses in a pump-probe configuration are possible to achieve, until now, only a few such light sources have been demonstrated. Here, by shaping the driving laser to an annular beam, a 100 kHz attosecond pulse train (APT) is reported with the highest energy so far (51 pJ/shot) on target (269 pJ at generation) among the high-repetition rate systems (>10 kHz) in which the attosecond pulses were temporally characterized. The on-target pulse energy is maximized by reducing the losses from the reflections and filtering of the high harmonics, and an unprecedented 19% transmission rate from the generation point to the target position is achieved. At the same time, the probe beam is also annular and low loss of this beam is reached by using another holey mirror to combine with the APT. The advantages of using an annular beam to generate attosecond pulses with a high-average power laser are demonstrated experimentally and theoretically. The effect of nonlinear propagation in the generation medium on the annular-beam generation concept is also analyzed in detail.

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Attosecond investigation of extreme-ultraviolet multi-photon multi-electron ionization

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Optica
9 (2022) 6, 639-644

https://doi.org/10.1364/OPTICA.456596

Multi-electron dynamics in atoms and molecules very often occur on sub- to few-femtosecond time scales. The available intensities of extreme-ultraviolet (XUV) attosecond pulses have previously allowed the time-resolved investigation of two-photon, two-electron interactions. Here we study double and triple ionization of argon atoms involving the absorption of up to five XUV photons using a pair of intense attosecond pulse trains (APTs). By varying the time delay between the two APTs with attosecond precision and the spatial overlap with nanometer precision, we obtain information on complex nonlinear multi-photon ionization pathways. Our experimental and numerical results show that Ar^{2+} is predominantly formed by a sequential two-photon process, whereas the delay dependence of the Ar^{3+} ion yield exhibits clear signatures of the involvement of a simultaneous two-photon absorption process. Our experiment suggests that it is possible to investigate multi-electron dynamics using attosecond pulses for both pumping and probing the dynamics.

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4D spatio-temporal characterization of ultrashort light pulses undergoing filamentation

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Optics Express
30 (2022) 15, 27938

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We present an experimental method capable of capturing the complete spatio-temporal dynamics of filamenting ultrashort laser pulses. By employing spatially resolved Fourier transform spectrometry in combination with the capability to terminate the filament at any length, we can follow the nonlinear dynamics in four dimensions, i.e. the transverse domain, time and filament length. Our method thus not only enables the full characterization of the filamentation process throughout its evolution, but also allows to identify and select laser pulses with desired parameters.

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Liquid-cooled modular gas cell system for high-order harmonic generation using high average power laser systems

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Rev. Sci. Instruments 93 (2022) 073002

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We present the design and implementation of a new, modular gas target suitable for high-order harmonic generation using high average power lasers. To ensure thermal stability in this high heat load environment, we implement an appropriate liquid cooling system. The system can be used in multiple-cell configurations, allowing us to control the cell length and aperture size. The cell design was optimized with heat and flow simulations for thermal characteristics, vacuum compatibility, and generation medium properties. Finally, the cell system was experimentally validated by conducting high-order harmonic generation measurements using the 100 kHz high average power HR-1 laser system at the Extreme Light Infrastructure Attosecond Light Pulse Source (ELI ALPS) facility. Such a robust, versatile, and stackable gas cell arrangement can easily be adapted to different experimental geometries in both table-top laboratory systems and user-oriented facilities, such as ELI ALPS.

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Two-XUV-photon double ionization of neon studied at the Extreme Light Infrastructure (ELI ALPS)

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Phys. Rev. A 106 (2022) 043117

http://dx.doi.org/10.1103/PhysRevA.106.043117

Two-XUV-photon double ionization of Ne, induced by an intense few-pulse attosecond train with a ⍰4 fs envelope duration is investigated experimentally and theoretically. The experiment is performed at ELI-ALPS (Extreme Light Infrastructure Attosecond Light Pulse Source) utilizing the recently constructed 10 Hz gas phase high-order harmonic generation SYLOS GHHG-COMPACT beamline. A total pulse energy up to ~1 μJ generated in argon in conjunction with high-reflectivity optics in the XUV region allowed the observation of the doubly charged state of Ne induced by 40 eV central XUV-photon energies. The interaction of the intense attosecond pulse train with Ne is also theoretically studied via second-order time-dependent perturbation theory equations of motion. The results of this work, combined with the feasibility of conducting XUV-pump–XUV-probe experiments, constitute a powerful tool for many potential applications. Those include attosecond pulse metrology as well as time-resolved investigations of the dynamics underlying direct and sequential double ionization and their electron correlation effects.

eli attosecond

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Strong laser fields and their power to generate controllable high-photon-number coherent-state superpositions

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Phys. Rev. A 105 (2022) 033714

https://doi.org/10.1103/PhysRevA.105.033714

Recently, intensely driven laser-matter interactions have been used to connect the fields of strong laser field physics with quantum optics by generating nonclassical states of light. Here, we take a further key step and show the potential of strong laser fields for generating controllable high-photon-number coherent-state superpositions. This has been achieved by using two of the most prominent strong-laser induced processes: high-harmonic generation and above-threshold ionization. We show how the obtained coherent-state superpositions change from an optical Schrödinger “cat” state to a “kitten” state by changing the atomic density in the laser-atom interaction region, and we demonstrate the generation of a nine-photon shifted optical “cat” state, which, to our knowledge, is the highest photon number optical “cat” state experimentally reported. Our findings anticipate the development of new methods that naturally lead to the creation of high-photon-number controllable coherent-state superpositions, advancing investigations in quantum technology.

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High photon number entangled states and coherent state superposition from the extreme ultraviolet to the far infrared

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Phys. Rev. Lett. 128 (2022) 123603

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We present a theoretical demonstration on the generation of entangled coherent states and of coherent state superpositions, with photon numbers and frequencies orders of magnitude higher than those provided by the current technology. This is achieved by utilizing a quantum mechanical multimode description of the single- and two-color intense laser field driven process of high harmonic generation in atoms. It is found that all field modes involved in the high harmonic generation process are entangled, and upon performing a quantum operation, lead to the generation of high photon number optical cat states spanning from the far infrared to the extreme ultraviolet spectral region. This provides direct insights into the quantum mechanical properties of the optical field in the intense laser matter interaction. Finally, these states can be considered as a new resource for fundamental tests of quantum theory, quantum information processing, or sensing with nonclassical states of light.

eli attosecond

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Light-matter entanglement after above-threshold ionization processes in atoms

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Physical Review A
106 (2022) 063705

https://doi.org/10.1103/PhysRevA.106.063705

Light-matter entanglement plays a fundamental role in many applications of quantum information science. Thus, finding processes where it can be observed is an important task. Here, using a one-dimensional model, we address this matter by investigating theoretically the entanglement between light and electrons generated in the above-threshold ionization (ATI) process. The study is based on the backaction of the ATI process on the quantum optical state of the system, and its dependence on the kinetic energy and direction of the emitted photoelectrons. Taking into account the dynamics of the process, we demonstrate the creation of hybrid entangled states. The amount of entanglement has been studied in terms of the entropy of entanglement. Additionally, we use the Wigner function of the driving field mode to motivate the entanglement characterization when considering electrons propagating in opposite directions.

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Intense isolated attosecond pulses from two-color few-cycle laser driven relativistic surface plasma

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Scientific Reports 12 (2022) 13668

https://doi.org/10.1038/s41598-022-17762-3

Ultrafast plasma dynamics play a pivotal role in the relativistic high harmonic generation, a phenomenon that can give rise to intense light fields of attosecond duration. Controlling such plasma dynamics holds key to optimize the relevant sub-cycle processes in the high-intensity regime. Here, we demonstrate that the optimal coherent combination of two intense ultrashort pulses centered at two-colors (fundamental frequency, ω and second harmonic, 2ω) can lead to an optimal shape in relativistic intensity driver field that yields such an extraordinarily sensitive control. Conducting a series of two-dimensional (2D) relativistic particle-in-cell (PIC) simulations carried out for currently achievable laser parameters and realistic experimental conditions, we demonstrate that an appropriate combination of ω−2ω along with a precise delay control can lead to more than three times enhancement in the resulting high harmonic flux. Finally, the two-color multi-cycle field synthesized with appropriate delay and polarization can all-optically suppress several attosecond bursts while favourably allowing one burst to occur, leading to the generation of intense isolated attosecond pulses without the need of any sophisticated gating techniques.

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A laser wakefield acceleration facility using SG-II petawatt laser system

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Rev. Sci. Instruments 93 (2022) 033504

https://doi.org/10.1063/5.0071761

Laser wakefield acceleration (LWFA) using PW-class laser pulses generally requires cm-scale laser–plasma interaction Rayleigh length, which can be realized by focusing such pulses inside a long underdense plasma with a large f-number focusing optic. Here, we present a new PW-based LWFA instrument at the SG-II 5 PW laser facility, which employs f/23 focusing. The setup also adapted an online probing of the plasma density via Nomarski interferometry using a probe laser beam having 30 fs pulse duration. By focusing 1-PW, 30-fs laser pulses down to a focal spot of 230 µm, the peak laser intensity reached a mild-relativistic level of 2.6×10^{18} W/cm^2, a level modest for standard LWFA experiments. Despite the large aspect ratio of >25:1 (transverse to longitudinal dimensions) of the laser pulse, electron beams were observed in our experiment only when the laser pulse experienced relativistic self-focusing at high gas-pressure thresholds, corresponding to plasma densities higher than 3×10^{-18} cm.

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Laser wakefield photoneutron generation with few-cycle high-repetition-rate laser systems

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Photonics
9 (2022) 11, 826

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We measured the exciton dynamics in van der Waals heterojunctions of transition metal dichalcogenides (TMDCs) and organic semiconductors (OSs). TMDCs and OSs are semiconducting materials with rich and highly diverse optical and electronic properties. Their heterostructures, exhibiting van der Waals bonding at their interfaces, can be utilized in the field of optoelectronics and photovoltaics. Two types of heterojunctions, MoS_2-pentacene and WSe_2-pentacene, were prepared by layer transfer of 20 nm pentacene thin films as well as MoS_2 and WSe_2 monolayer crystals onto Au surfaces. The samples were studied by means of transient absorption spectroscopy in the reflectance mode. We found that A-exciton decay by hole transfer from MoS_2 to pentacene occurs with a characteristic time of 21 ± 3 ps. This is slow compared to previously reported hole transfer times of 6.7 ps in MoS_2-pentacene junctions formed by vapor deposition of pentacene molecules onto MoS_2 on SiO_2. The B-exciton decay in WSe_2 shows faster hole transfer rates for WSe_2-pentacene heterojunctions, with a characteristic time of 7 ± 1 ps. The A-exciton in WSe_2 also decays faster due to the presence of a pentacene overlayer; however, fitting the decay traces did not allow for the unambiguous assignment of the associated decay time. Our work provides important insights into excitonic dynamics in the growing field of TMDC-OS heterojunctions.

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Low divergent MeV-class proton beam with micrometer source size driven by a few-cycle laser pulse

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Scientific Reports 12 (2022) 8100

https://doi.org/10.1038/s41598-022-12240-2

Spatial characterization of 0.5 MeV proton beam, driven by 12 fs, 35 mJ, 1019 W/cm^2 intense laser-foil interaction is presented. The accelerated proton beam has been applied to obtain a high-resolution, point-projection static radiograph of a fine mesh using a CR-39 plate. The reconstruction of mesh edge blurring and particle ray tracing suggests that these protons have an effective source size (FWHM) of just 3.3 ± 0.3 µm. Furthermore, the spatial distribution of the proton beam recorded on the CR-39 showed that the divergence of these particles is less than 5-degree (FWHM). The low divergence and small source size of the proton beam resulted in an ultralow transverse emittance of 0.00032 π-mm-mrad, which is several orders of magnitude smaller than that of a conventional accelerator beam.

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Threshold target thickness in high-contrast laser-driven ion acceleration

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Scientific Reports
12 (2022) 8100

https://doi.org/10.1063/5.0116088

The experiments have revealed a “threshold” target thickness for proton acceleration by a femtosecond petawatt class laser above which the forward accelerated proton energy weakly depends on the target thickness, while for thinner targets, it gradually decreases. For targets thicker than the threshold, the pre-pulse-induced preplasma at the target front can boost ion acceleration by increasing laser absorption, while this is ineffective for thinner targets due to prepulse-induced plasma formation at the target rear. This dual nature of the preplasma is described analytically, and particle-in-cell simulations confirm this concept. A linear dependence of threshold target thickness on the prepulse intensity is predicted.

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Design of semiconductor contact grating terahertz source with enhanced diffraction efficiency

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Crystals
12 (2022) 8, 1173

https://doi.org/10.3390/cryst12081173

We report a semiconductor contact grating terahertz source design based on a rectangular profile for phase-matched terahertz generation in the long infrared pump wavelength range. The calculations show that the best diffraction efficiency can be achieved by a filling factor significantly smaller than 50%. Furthermore, the possibility of diffraction efficiency enhancement was investigated by applying three different antireflective coating structures. Numerical simulations have indicated that at 2.06 µm and 3.0 µm pump wavelength, diffraction efficiencies greater than 91% and 89% can be achieved by adding an appropriate antireflective coating to the GaP and GaAs contact grating structure, respectively. In addition, numerical simulations were performed to investigate the influence of wall angles on diffraction efficiency. The results reveal that the wall angle does not significantly affect the diffraction efficiency: while keeping the wall angle deviation from the vertical below 25 degrees, the efficiency drop remains below 5% for otherwise optimal grating parameters.

Uniformly scalable lithium niobate THz pulse source in transmission geometry

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Opt. Express 30 (2022) 4434

https://doi.org/10.1364/OE.440883

A novel THz source, based on optical rectification in LiNbO3 using the tilted-pulse-front technique, is proposed and experimentally demonstrated. The pulse-front tilt is introduced by a volume phase holographic grating, efficiently used at perpendicular incidence in transmission, and the THz pulses are produced in a $LiNbO_3$ plane-parallel nonlinear echelon slab, arranged parallel to the grating. As a unique feature, the entire setup has a plane-parallel, transmission-type configuration, which straightforwardly enables distortion-free scaling to large sizes, high pulse energies and high THz field strengths. The possibility of operating the setup at cryogenic temperature for increased THz generation efficiency is also investigated. Calculations predict efficiencies of 95% for diffraction and 2% for THz generation at room temperature with a refractive-index-matching liquid between the grating and the echelon slab.

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The role of asymmetry in few-cycle, mid-IR pulses during THz pulse generation

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Journal of Optics
24 (2022) 045502

https://doi.org/10.1088/2040-8986/ac5289

The efficiency of terahertz (THz) pulse generation improves at longer driving wavelengths. For this reason, the use of mid-infrared (MIR) sources is more advantageous compared to visible or near-infrared systems. In this work, we investigate how single-color and two-color schemes of MIRpulses with few-cycle pulse durations compare in producing THz pulses. The results reveal that as the duration of the driving pulses decreases, the second harmonic generation crystal can be omitted from the system. Our numerical study pinpointed three regions where the optimal pulse parameters are fundamentally different for the most efficient THz pulse generation. The f irst is the two-color approach, where the two-color scheme is dominant at 3.2 optical cycles and over. The second is the single-color approach, where the single-color scheme becomes dominant at 1.7 optical cycles and below. Therefore, it simplifies the traditional two-color scheme for THz pulse generation. There is also a third transitional region where the two-color scheme still prevails, but the sign of the relative phase between the input pulses becomes important. Considering the effect of the relative phase and the carrier to envelope phase (CEP) effect on the THz pulse generation, the results have shown that as the pulse duration become shorter, the role of the CEP becomes important for efficient THz generation. By measuring the efficiency of the THz generation in this optical arrangement, quantifying the CEP becomes possible, which could become an important experimental tool for few-cycle, MIR laser technology.

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Control of plasmonic field enhancement by mode-mixing

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Applied Physics Letters 120 (2022) 5, 053103

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We demonstrate experimentally that nanoscale control of plasmonic field enhancement becomes available by changing the polarization state of light. This is revealed by photoelectron emission from plasmonic nanorods illuminated with linearly and circularly polarized femtosecond laser pulses. Simulations show that the tunability of the field enhancement originates from the mode-mixing property of circularly polarized illumination, meaning simultaneous excitation of multiple plasmon modes of the nanostructures. Performing trajectory calculations of the photoemitted electrons, we prove that the kinetic energy scaling law remains the same irrespective to the polarization state.

eli attosecond

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

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Phys. Chem. Chem. Phys. 2022, Advance Article

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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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A quantum-chemical perspective on the laser-induced alignment and orientation dynamics of the CH_3X (X = F, Cl, Br, I) molecules

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Computational Chemistry 43 (2022) 8, 519-538

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Motivated by recent experiments, the laser-induced alignment-and--orientation (A&O) dynamics of the prolate symmetric top CH_3X (X = F, Cl, Br, I) molecules is investigated, with particular emphasis on the effect of halogen substitution on the rotational constants, dipole moments, and polarizabilities of these species, as these quantities determine the A&O dynamics. Insight into possible control schemes for preferred A&O dynamics of halogenated molecules and best practices for A&O simulations are provided, as well. It is shown that for accurate A&O -dynamics simulations it is necessary to employ large basis sets and high levels of electron correlation when computing the rotational constants, dipole moments, and polarizabilities. The benchmark-quality values of these molecular parameters, corresponding to the equilibrium, as well as the vibrationally averaged structures are obtained with the help of the focal-point analysis (FPA) technique and explicit electronic-structure computations utilizing the gold-standard CCSD(T) approach, basis sets up to quintuple-zeta quality, core-correlation contributions and, in particular, relativistic effects for CH_3Br and CH_3I. It is shown that the different A&O behavior of the CH_3X molecules in the optical regime is mostly caused by the differences in their polarizability anisotropy, in other terms, the size of the halogen atom. In contrast, the A&O dynamics of the CH_3X series induced by an intense few-cycle THz pulse is mostly governed by changes in the rotational constants, due to the similar dipole moments of the CH_3X molecules. The A&O dynamics is most sensitive to the B rotational constant: even the difference between its equilibrium and vibrationally-averaged values results in noticeably different A&O dynamics. The contribution of rotational states having different symmetry, weighted by nuclear-spin statistics, to the A&O dynamics is also studied.

eli attosecond

Achieving high molecular alignment and orientation for CH_3F through manipulation of rotational states with varying optical and THz laser pulse parameters

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Increasing interest in the fields of high-harmonics generation, laser-induced chemical reactions, and molecular imaging of gaseous targets demands high molecular "alignment" and "orientation" (A&O). In this work, we examine the critical role of different pulse parameters on the field-free A&O dynamics of the CH_3F molecule, and identify experimentally feasible optical and THz range laser parameters that ensure maximal A&O for such molecules. Herein, apart from rotational temperature, we investigate effects of varying pulse parameters such as, pulse duration, intensity, frequency, and carrier envelop phase (CEP). By analyzing the interplay between laser pulse parameters and the resulting rotational population distribution, the origin of specific A&O dynamics was addressed. We could identify two qualitatively different A&O behaviors and revealed their connection with the pulse parameters and the population of excited rotational states. We report here the highest alignment of $\langle\cos^2\theta\rangle=0.843$ and orientation of $\langle\cos(\theta)\rangle=0.886$ for CH_3F molecule at 2 K using a single pulse. Our study should be useful to understand different aspects of laser-induced unidirectional rotation in heteronuclear molecules, and in understanding routes to tune/enhance A&O in laboratory conditions for advanced applications.

US

Photoexcited intramolecular charge transfer in dye sensitizers: predictive in silico screening for dye-sensitized solar cell devices

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ACS Omega
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Efficient photoinduced intramolecular charge transfer (ICT) from donor to acceptor in dye molecules is the functional basis and key property in the working of a dye-sensitized solar cell (DSSC). To understand the ICT process in photoexcited dye molecules, we analyze the electronic properties and structural parameters of a chosen set of experimentally synthesized donor–acceptor (D–A) and donor−π-spacer−acceptor (D−π–A) type dye molecules in their ground, excited, and cationic states. The correlation between structural modification and charge redistribution in different parts of the molecule helps to identify the extent of π-conjugation and spatial rearrangement of electron density localization along the molecular skeleton. We find that prominent twisting of several groups and the resulting molecular bond rearrangements in larger parts of the molecule promote efficient donor to acceptor ICT, such as in D–A type ADEKA$_1$ and C275 dyes. Thus, based on the modest computation of structural and electronic properties of dye molecules in their respective ground, excited, and cationic states, we identify the desired structural changes that facilitate tunable intramolecular charge transfer to highlight a simple and direct prescription to screen out probable efficient dye molecules among many samples. Our approach complements recent experimental evidence of capturing the structural view of the excited-state charge transfer in molecules.

US

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Distinctive onset of electron correlation in molecular tautomers

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Phys. Rev. A
105 (2022) 062808

https://
doi.org/10.1103/
PhysRevA.105.062808

We investigate the attosecond response of the electronic cloud of a molecular system to an outer-valence ionization. The time needed for the remaining electrons to respond to a sudden perturbation in the electronic structure of the molecule is a measure of the degree of electron correlation. Using the ab initio multielectron wave-packet propagation method, we analyze the ultrafast many-body dynamics following the removal of different outer-valence electrons of two tautomers of the uracil molecule and show that this response time can be sensitive to the molecular structure and the symmetry of the ionized molecular orbital.

eli attosecond

US

Rational amphiphilic ligand engineering enables enhanced stability and efficiency of $CsPbBr_3$ nanocrystals based light emitting diodes

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Adv. Opt. Materials 2022, 2201176

https://doi.org/10.1002/adom.202201176

Metal halide perovskites have shown great potential for lighting. However, their low stability under irradiation/thermal stress and/or ambient storage conditions are critical for light-emitting diodes (LEDs). Among the stabilization strategies, ligand surface modification is effective toward stable perovskites, but the dynamic ligand adsorption/desorption process on the surface is a limiting factor. Herein, a new family of biogenic and amphiphilic capping agents, phosphatidyl-L-serine (Ptd-L-Ser), combining stronger multibinding motifs compared to conventional capping agents has led to superior $CsPbBr_3$ ($CsPbBr_3$-Ptd-L-Ser) with significantly enhanced stability upon storage/heating/water, keeping excellent photoluminescence quantum yields of ≈80% over half year. Spectroscopic/theoretical studies reveal that the origin of this behavior is the increased exciton binding energy associated to the versatility of multiple bindings. This results in $CsPbBr_3$-Ptd-L-Ser nanocrystals-based green-LEDs featuring excellent stabilities of >700 h (20 mA) and >200 h (100 mA) that strongly contrast with the reference devices with pristine $CsPbBr_3$ nanocrystals (120 h (20 mA) and 27 h (100 mA)). White LEDs (WLEDs) with chromaticity coordinates of (0.34, 0.33) and high luminous efficiency of 76 lm W^{-1}, keeping stable over weeks, are further demonstrated under continuous operational conditions, thereby suggesting $CsPbBr_3$-Ptd-L-Ser nanocrystals can be a potential candidate for commercial WLED technology.

eli attosecond

US

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Self-aggregation, H-bonding, and photoresponse in film and solution states of azobenzene containing polyurea

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Phys. Chem. Chem. Phys. 24 (2022) 38, 23447-23459

https://doi.org/10.1039/D2CP01200F

We critically understand the hydrogen bonding interactions and electronic transitions occurring in a thin film as well as in solution of a photo-responsive polymer, azo-polyurea (azo-PU). We synthesize azo-PU by covalent attachment of the azobenzene chromophore to the main chain of polyurea. Azo-PU shows reversible photoisomerization between trans and cis states upon light exposure, the occurrence of which is typically analysed using the π–π* and n–π* electronic transition peaks in the UV-visible absorption spectrum. We find that the π–π* and n–π* bands undergo a redshift and blueshift respectively on dissolving azo-PU in DMF solvent, resulting in a single overlapped peak in the spectrum. However, upon UV irradiation, these bands split into two independent transitions that are characteristic of azo-PU solid films. These observations are explained based on the changes in polymer–polymer and polymer–solvent interactions through hydrogen bonding and self-aggregation tendency. The experimental findings are corroborated using DFT simulations which provide useful insights into electronic orbital transitions, electron distribution, and hydrogen bonding interaction through IR vibrational modes.

US

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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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Adv. Funct. Materials 2022, 2209249

https://doi.org/10.1002/adfm.202209249

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 work 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.

eli attosecond

US

Effect of metal ad-atoms on the structural, electrical, and optical properties of boron-nitride nanostructures towards optoelectronics: a DFT based study

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Egypt. J. Chem.
65 (2022) 13, 745-752

https://doi.org/10.21608/ejchem.2022.130936.5763

In the present study, we investigate the structural, electronic, and optical properties of pure and doped boron-nitride (BN) nano-systems using density functional theory (DFT) simulations. Metal dopant C, Ni and Cu were introduced. Structural properties, such as bond length and formation energy, were observed, and the bond lengths were found to agree with experimental results previously reported in the literature. The metal atoms in the metal-doped BN nanosystem were shown to have a direct effect on the nature of the surface. The calculated formation energies show that the stability of the BN nanosystem is enhanced upon metallic doping, as clearly shown in the case of Ni-doped boron nitride (NiBN). The metal dopant is found to reduce the energy gap and enhance overall electrical conductivity. UV-visible spectrum calculations show that the metal atom causes a red-shift in the spectrum towards the red wavelengths. Open-circuit voltage (VOC) calculation shows that Ni-doping of BN enhances the VOC by 420 meV with respect to pristine BN, thereby confirming the positive impact of the dopant Ni on the two-dimensional h-BN surface and consequent possible usefulness in optoelectronics.

eli attosecond

US

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Antioxidant potential of Santowhite as synthetic and ascorbic acid as natural polymer additives

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Polymers
14 (2022) 17, 3518

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A wide variety of additives are used to improve specific characteristics of the final polymeric product. Antioxidant additives (AAs) can prevent oxidative stress and thus the damage of polymeric materials. In this work, the antioxidant potential and thus the applicability of Santowhite (SW) as synthetic and ascorbic acid (Asc) as natural AAs were explored by using computational tools. Two density functional theory (DFT) methods, M05-2X and M06-2X, have been applied in combination with the 6-311++G(2d,2p) basis set in gas phase. Three antioxidant mechanisms have been considered: hydrogen atom transfer (HAT), single electron transfer-proton transfer (SET-PT), and sequential proton loss electron transfer (SPLET). Bond dissociation enthalpy (BDE), ionization potential (IP), proton dissociation enthalpy (PDE), proton affinity (PA), and electron transfer enthalpy (ETE) have been computed for each potential hydrogen donor site. The results indicate that the antioxidant potential of Asc is higher than SW. Furthermore, some of the C-H bonds, depending on their position in the structures, are potent radical scavengers, but O-H groups are more prone to donate H-atoms to free radicals. Nonetheless, both additives can be potentially applied to safeguard common polymers and prohibit oxidative stress-induced material deterioration.

eli attosecond

US

Effect of metal ad-atoms on the structural, electrical, and optical properties of boron-nitride nanostructures towards optoelectronics: a DFT based study

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Egypt. J. Chem.
65 (2022) 13, 745-752

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In the present study, we investigate the structural, electronic, and optical properties of pure and doped boron-nitride (BN) nano-systems using density functional theory (DFT) simulations. Metal dopant C, Ni and Cu were introduced. Structural properties, such as bond length and formation energy, were observed, and the bond lengths were found to agree with experimental results previously reported in the literature. The metal atoms in the metal-doped BN nanosystem were shown to have a direct effect on the nature of the surface. The calculated formation energies show that the stability of the BN nanosystem is enhanced upon metallic doping, as clearly shown in the case of Ni-doped boron nitride (NiBN). The metal dopant is found to reduce the energy gap and enhance overall electrical conductivity. UV-Visible spectrum calculations show that the metal atom causes a red-shift in the spectrum towards the red wavelengths. Open-circuit voltage (VOC) calculation shows that Ni-doping of BN enhances the VOC by 420 meV with respect to pristine BN, thereby confirming the positive impact of the dopant Ni on the two-dimensional h-BN surface and consequent possible usefulness in optoelectronics.

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Judit BUDAI
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Nonadiabatic nano-optical tunneling of photoelectrons in plasmonic near-fields

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Nano Letters
22 (2022) 6, 2303–2308

https://doi.org/10.1021/acs.nanolett.1c04651

Nonadiabatic nano-optical electron tunneling in the transition region between multiphoton-induced emission and adiabatic tunnel emission is explored in the near-field of plasmonic nanostructures. For Keldysh γ values between ~1.3 and ~2.2, measured photoemission spectra show strong-field recollision driven by the nanoscale near-field. At the same time, the photoemission yield shows an intensity scaling with a constant nonlinearity, which is characteristic for multiphoton-induced emission. Our observations in this transition region were well reproduced with the numerical solution of Schrödinger's equation, mimicking the nanoscale geometry of the field. This way, we determined the boundaries and nature of nonadiabatic tunneling photoemission, building on a key advantage of a nanoplasmonic system, namely, that high-field-driven recollision events and their signature in the photoemission spectrum can be observed more efficiently due to significant nanoplasmonic field enhancement factors.

Judit BUDAI
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Ultrasensitive probing of plasmonic hot electron occupancies

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Nature Communications
13 (2022) 6695

https://doi.org/10.1038/s41467-022-34554-5

Non-thermal and thermal carrier populations in plasmonic systems raised significant interest in contemporary fundamental and applied physics. Although the theoretical description predicts not only the energies but also the location of the generated carriers, the experimental justification of these theories is still lacking. Here, we demonstrate experimentally that upon the optical excitation of surface plasmon polaritons, a non-thermal electron population appears in the topmost domain of the plasmonic film directly coupled to the local fields. The applied all-optical method is based on spectroscopic ellipsometric determination of the dielectric function, allowing us to obtain in-depth information on surface plasmon induced changes of the directly related electron occupancies. The ultrahigh sensitivity of our method allows us to capture the signatures of changes induced by electron-electron scattering processes with ultrafast decay times. These experiments shed light on the build-up of plasmonic hot electron population in nanoscale media.

Ultrafast plasmonic photoemission in the single-cycle and few-cycle regimes

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Scientific Reports
12 (2022) 3932

https://doi.org/10.1038/s41598-022-07259-4

Due to the highly increased interest in the development of state-of-the-art applications of photoemission in ultrafast electron microscopy, development of photocathodes and many more applications, a correct theoretical understanding of the underlying phenomena is needed. Within the framework of the single active electron approximation the most accurate results can be obtained by the direct solution of the time-dependent Schrödinger equation (TDSE). In this work, after a brief presentation of a numerically improved version of a mixed 1D-TDSE method, we investigated the characteristics of electron spectra obtained from the surface of metal nanoparticles irradiated with ultrashort laser pulses. During our investigation different decay lengths of the plasmonic-enhanced incident field in the vicinity of the metal were considered. Using the simulated spectra we managed to identify the behavior of the cutoff energy as a function of decay length in the strong-field, multiphoton and transition regimes.

Polarization dependence of atomic high-order harmonic generation: Description using a discrete basis

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Péter FÖLDI

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Phys. Rev. A
105 (2022) 033105

https://
doi.org/10.1103/
PhysRevA.105.033105

The optical generation of high-order harmonics is known to have a strong polarization dependence: In contrast to linearly polarized excitation, circularly polarized light induces practically no harmonics. In the current paper we focus on atomic targets, the case when a well-established physical picture explains the effect: For circular polarization, the photoionized electrons never return to their parent nuclei, and the energy they gained while being accelerated by the field is not transferred into high-order harmonic radiation. This is essentially a picture that is based on real-space electron trajectories (or the dynamics of the wave functions). Here, we provide an alternative description that uses the discrete Sturmian basis, and points out how quantum mechanical interference effects and selection rules can explain the polarization dependence of the process. This emphasizes the importance of space-time symmetries during the process of high-order harmonic generation.

Péter DOMBI
Zsuzsanna PÁPA

Femtosecond LIPSS on indium-tin-oxide thin films at IR wavelengths

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Applied Optics
61 (2022) 2, 386-391

https://doi.org/10.1364/AO.444653

We investigated laser-induced periodic surface structures (LIPSS) generated on indium-tin-oxide thin films with femtosecond laser pulses in the infrared region. Using pulses between 1.6 and 2.4 μm central wavelengths, we observed robust LIPSS morphologies with a periodicity close to λ/10. Supporting finite-difference time-domain calculations suggests that the surface forms are rooted in the field localization in the surface pits leading to a periodically increased absorption of the laser pulse energy that creates the observed periodic structures.

Katalin HIDEGHÉTY

Magnetic resonance imaging - based delineation of organs at risk in the head and neck region

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Adv. in Rad. Oncology 8 (2022) 2, 101042

https://doi.org/10.1016/j.adro.2022.101042

Purpose: The aim of this article is to establish a comprehensive contouring guideline for treatment planning using only magnetic resonance images through an up-to-date set of organs at risk (OARs), recommended organ boundaries, and relevant suggestions for the magnetic resonance imaging (MRI)–based delineation of OARs in the head and neck (H&N) region.

Methods and materials: After a detailed review of the literature, MRI data were collected from the H&N region of healthy volunteers. OARs were delineated in the axial, coronal, and sagittal planes on T2-weighted sequences. Every contour defined was revised by four radiation oncologists and subsequently by two independent senior experts (H&N radiation oncologist and radiologist). After revision, the final structures were presented to the consortium partners.

Results: A definitive consensus was reached after multi-institutional review. On that basis, we provided a detailed anatomic and functional description and specific MRI characteristics of the OARs.

Conclusions: In the era of precision radiation therapy, the need for well-built, straightforward contouring guidelines is on the rise. Precise, uniform, delineation-based, automated OAR segmentation on MRI may lead to increased accuracy in terms of organ boundaries and analysis of dose-dependent sequelae for an adequate definition of normal tissue complication probability.

eli attosecond

US

Application of lacunarity for quantification of single molecule localization microscopy images

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Cells
11 (2022) 19, 3105

https://doi.org/10.3390/cells11193105

The quantitative analysis of datasets achieved by single molecule localization microscopy is vital for studying the structure of subcellular organizations. Cluster analysis has emerged as a multi-faceted tool in the structural analysis of localization datasets. However, the results it produces greatly depend on the set parameters, and the process can be computationally intensive. Here we present a new approach for structural analysis using lacunarity. Unlike cluster analysis, lacunarity can be calculated quickly while providing definitive information about the structure of the localizations. Using simulated data, we demonstrate how lacunarity results can be interpreted. We use these interpretations to compare our lacunarity analysis with our previous cluster analysis-based results in the field of DNA repair, showing the new algorithm's efficiency.

eli attosecond

Beam pulse structure and dose rate as determinants for the Flash effect observed in zebrafish embryo

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Radioth.and Onc.
170 (2022) S69-S70

https://doi.org/10.1016/j.radonc.2022.05.025

Background and purpose: Continuing recent experiments at the research electron accelerator ELBE at the Helmholtz-Zentrum Dresden -Rossendorf the influence of beam pulse structure on the Flash effect was investigated.

Materials and methods: The proton beam pulse structure of an isochronous cyclotron (UHDRiso) and a synchrocyclotron (UHDRsynchro) was mimicked at ELBE by quasi-continuous electron bunches at 13 MHz delivering mean dose rates of 287 Gy/s and 177 Gy/s and bunch dose rates of 106 Gy/s and 109 Gy/s, respectively. For UHDRsynchro, 40 ms macro pulses at a frequency of 25 Hz superimposed the bunch delivery. For comparison, a maximum beam intensity (2.5 × 105 Gy/s mean and ~10^9 Gy/s bunch dose rate) and a reference irradiation (of ~8 Gy/min mean dose rate) were applied. Radiation induced changes were assessed in zebrafish embryos over four days post irradiation.

Results: Relative to the reference a significant protecting Flash effect was observed for all electron beam pulse regimes with less severe damage the higher the mean dose rate of the electron beam. Accordingly, the macro pulsing induced prolongation of treatment time at UHDRsynchro regime reduces the protecting effect compared to the maximum regime delivered at same bunch but higher mean dose rate. The Flash effect of the UHDRiso regime was confirmed at a clinical isochronous cyclotron comparing the damage induced by proton beams delivered at 300 Gy/s and ~9 Gy/min.

Conclusion: The recent findings indicate that the mean dose rate or treatment time are decisive for the normal tissue protecting Flash effect in zebrafish embryo.

US

Sacrificial agent gone rogue: electron-acceptor-induced degradation of $CsPbBr_3$ photocathodes

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ACS Energy Lett.
7 (2022) 417−424

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Lead halide perovskites (LHPs) have emerged as perspective materials for light harvesting, due to their tunable band gap and optoelectronic properties. Photocatalytic and photoelectrochemical (PEC) studies, employing LHP/liquid junctions, are evolving, where sacrificial reagents are often used. In this study, we found that a frequently applied electron scavenger (TCNQ) has dual roles: while it leads to rapid electron transfer from the electrode to TCNQ, enhancing the PEC performance, it also accelerates the decomposition of the $CsPbBr_3$ photoelectrode. The instability of the films is caused by the TCNQ-mediated halide exchange between the dichloromethane solvent and the LHP film, during PEC operation. Charge transfer and halide exchange pathways were proposed on the basis of in situ spectroelectrochemical and ex situ surface characterization methods, also providing guidance on planning PEC experiments with such systems.

eli attosecond

US

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Exciton dynamics in MoS_2-Pentacene and WSe_2-Pentacene heterojunctions

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ACS Nano 16 (2022) 10, 16668–16676

https://doi.org/10.1021/acsnano.2c06144

We measured the exciton dynamics in van der Waals heterojunctions of transition metal dichalcogenides (TMDCs) and organic semiconductors (OSs). TMDCs and OSs are semiconducting materials with rich and highly diverse optical and electronic properties. Their heterostructures, exhibiting van der Waals bonding at their interfaces, can be utilized in the field of optoelectronics and photovoltaics. Two types of heterojunctions, MoS_2-pentacene and WSe_2-pentacene, were prepared by layer transfer of 20 nm pentacene thin films as well as MoS_2 and WSe_2 monolayer crystals onto Au surfaces. The samples were studied by means of transient absorption spectroscopy in the reflectance mode. We found that A-exciton decay by hole transfer from MoS_2 to pentacene occurs with a characteristic time of 21 ± 3 ps. This is slow compared to previously reported hole transfer times of 6.7 ps in MoS_2-pentacene junctions formed by vapor deposition of pentacene molecules onto MoS_2 on SiO_2. The B-exciton decay in WSe_2 shows faster hole transfer rates for WSe_2-pentacene heterojunctions, with a characteristic time of 7 ± 1 ps. The A-exciton in WSe_2 also decays faster due to the presence of a pentacene overlayer; however, fitting the decay traces did not allow for the unambiguous assignment of the associated decay time. Our work provides important insights into excitonic dynamics in the growing field of TMDC-OS heterojunctions.

eli attosecond

US

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Probing light-induced conical intersections by monitoring multidimensional polaritonic surfaces

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J. Phys. Chem. Lett.
13 (2022) 1172–1179

https://
doi.org/10.1021/
acs.jpclett.1c03465

The interaction of a molecule with the quantized electromagnetic field of a nanocavity gives rise to light-induced conical intersections between polaritonic potential energy surfaces. We demonstrate for a realistic model of a polyatomic molecule that the time-resolved ultrafast radiative emission of the cavity enables following both nuclear wavepacket dynamics on, and nonadiabatic population transfer between, polaritonic surfaces without applying a probe pulse. The latter provides an unambiguous (and in principle experimentally accessible) dynamical fingerprint of light-induced conical intersections.

eli attosecond

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Radiative emission of polaritons controlled by light-induced geometric phase

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Chem. Communications 58 (2022) 12612

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Polaritons – hybrid light-matter states formed in cavity – strongly change the properties of the underlying matter. In optical or plasmonic nanocavities, polaritons decay by radiative emission of the cavity, which is accessible experimentally. Due to the interaction of a molecule with the quantized radiation field, polaritons exhibit light-induced conical intersections (LICIs) which dramatically influence the nuclear dynamics of molecular polaritons. We show that ultrafast radiative emission from the lower polariton is controlled by the geometric phase imposed by the LICI. This finding provides insight into the process of emission and, furthermore, allows one to compute these signals by augmenting the Born-Oppenheimer approximation for polaritons with a geometric phase term.

Competition between collective and individual conical intersection dynamics in an optical cavity

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Light-induced nonadiabatic phenomena arise when molecules or molecular ensembles are exposed to resonant external electromagnetic fields. The latter can either be classical laser or quantized cavity radiation fields, which can couple to either the electronic, nuclear or rotational degrees of freedom of the molecule. In the case of quantized radiation fields, the light–matter coupling results in the formation of two new hybrid light–matter states, namely the upper and lower 'polaritons'. Light-induced avoided crossings and light-induced conical intersections (CIs) between polaritons exist as a function of the vibrational and rotational coordinates of single molecules. For ensembles of N molecules, the N − 1 dark states between the two optically active polaritons feature, additionally, so-called collective CIs, involving the coordinates of more than one molecule to form. Here, we study the competition between intramolecular and collective light-induced nonadiabatic phenomena by comparing the escape rate from the Franck–Condon region of a single molecule and of a molecular ensemble coupled to a cavity mode. In situations where the polaritonic gap would be large and the dark-state decay channels could not be reached effectively, the presence of a seam of light-induced CI between the polaritons facilitates again the participation of the dark manifold, resulting in a cooperative effect that determines the overall non-radiative decay rate from the upper into the lower polaritonic states.

Coherent and incoherent superposition of transition matrix elements of the squeezing operator

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We discuss the general matrix elements of the squeezing operator between number eigenstates of a harmonic oscillator (which may also represent a quantized mode of the electromagnetic radiation). These matrix elements have first been used by Popov and Perelomov (1969 Zh. Eksp. Teor. Fiz. 56 1375–90) long ago, in their thorough analysis of the parametric excitation of harmonic oscillators. They expressed the matrix elements in terms of transcendental functions, the associated Legendre functions. In the present paper we will show that these matrix elements can also be derived in a different form, expressed by the classical Gegenbauer polynomials. This new expression makes it possible to determine coherent and incoherent superpositions of these matrix elements in closed analytic forms. As an application, we describe multiphoton transitions in the system 'charged particle + electromagnetic radiation', induced by a (strong) coherent field or by a black-body radiation component (with a Planck–Bose photon number distribution). The exact results are compared with the semi-classical ones. We will show that in case of interaction with a thermal field, the semi-classical result (with a Gaussian stochastic field amplitude) yields an acceptable approximation only in the Rayleigh–Jeans limit, however, in the Wien limit it completely fails.

Eng

Investigation of the adsorption properties of cyclic C6 molecules on h-BN/Rh(111) surface, efforts to cover the boron nitride nanomesh by graphene

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The 2D atomic structure of h-BN would be an excellent dielectric layer to complement graphene electronics. When grown sequentially on metal substrates to create GR/h-BN/metal sandwich structures, these nanomaterials can be used in various applications. To this end, in this project we studied the adsorption and dissociation of cyclohexene and benzene on clean and h-BN covered Rh(111) surfaces at low and at high temperatures. Although we observed that both molecules adsorb on the h-BN/Rh(111) surface at 160 K, there is only a weak interaction between these molecules and h-BN. Moreover, h-BN proved to be completely inert to the split of cyclohexene and benzene after low temperature exposures. In our high temperature experiments, we tested the stability of h-BN towards oxygen, hydrogen and we also followed the effects of high exposure adsorption of the C6 molecules on the nanomesh. We observed a different behavior following the decomposition of the two hydrocarbon species. In one case we developed a graphene-like carbon structure in parallel with BN, while in the other process the carbon layer formed on top of the surface of h-BN/Rh(111). Our results were evidenced by Auger Electron Spectroscopy (AES), High Resolution Electron Energy Loss Spectroscopy (HREELS) and Mass Spectrometry (MS).

Eng

A round dance of acetaldehyde molecular ensembles on Rh(111) surface; formation and decomposition of various paraldehyde conformers

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The adsorption, desorption and polymerization of acetaldehyde (AA) on Rh(111) surface were studied by TDS, HREELS, work function measurements and theoretical calculations. Polymerization, as a new feature after adsorption of AA on Rh(111), makes the interpretation of the surface processes more exciting. The single molecule itself has at least two possibilities for adsorption on Rh(111) surface, namely η^1–(O)–CH_3CHO_a and η^2–(O,C)–CH_3CHO_a adsorption forms are considered. Moreover, η^1–(O)–CH_3CHO_a aldehyde molecules can react with each other forming oligomers or even longer polymers, as well. Paraldehyde, the triple molecular mass species of AA, shows some surprising kinetic and thermodynamic features on clean Rh(111) surface. However, the observed surface formation of the oligomer and its thermodynamically most stable gas phase conformer seems to be in contradiction. The interpretation of this phenomenon is the goal of the present study.