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Special Issue: Dynamics in Tailored Ultrashort Light Fields

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FOREWORD

Special Issue: Dynamics in Tailored Ultrashort Light Fields

This special issue on Dynamics in Tailored Ultrashort Light Fields reports on the progress in a research field at the intersection of femtosecond science, attosecond science and coherent control. Since light is one of our main tools to observe and manipulate processes on microscopic scales, the accessible timescales have always depended on our capabilities of temporal light-pulse shaping. Today, sophisticated pulse-shaping techniques allow for tailoring the time-dependent electric fields rather than only the envelope of ultrashort pulses. Examples of control by tailored fields include ionization and fragmentation dynamics of atoms, molecules or nanoobjects, generation of radiation or rearrangement of bound charges. The ability to exert control over the electric field along with the use of polarization-controlled or two-colour laser fields are themes common to many of the articles within this special issue. The motivation to devote an issue of Journal of Modern Optics to this topic arose out of the research network *Quantum Dynamics in Tailored Intense Fields* which is based in Germany and funded by the German Research Foundation DFG in the frame of a Priority Programme.

For many years, one of the well-known applications of polarization-controlled fields has been the production of isolated attosecond pulses by polarization-gating, which employs two time-delayed counter-rotating circularly polarized pulses such that high-harmonic generation can take place only during the short period of significant overlap between the pulses. Chen et al. report simulations on a promising variation of this method, where the two pulses are of unequal strength, suppressing ionization in the early stage of the field and supporting an increase in the high-harmonic cut-off. Jia et al. discuss another central problem in attoscience, namely the preparation of attosecond charge migration in molecules. They discuss specifically the electron flux connected to a laser-prepared electronic superposition state in the iodo-acetylenic cation. It is shown that the onset of electron flux occurs already during the action of the applied pulse, which is tailored such that the charge-migration amplitude is maximized.

Odžak et al. present a theoretical overview of the laser-induced physics with strong bicircular fields, including above-threshold detachment, laser-assisted radiative recombination and the generation of circularly polarized harmonics. Currently, bicircular fields, in particular those with two counter-rotating colours, constitute an intensively pursued area in strong-field science as they offer the possibility to generate bright circular harmonics. Another configuration of two-colour fields is studied in the article by Eicke and Lein, where it is shown theoretically that the relative-phase-dependence of strong-field ionization by orthogonally polarized ω-2ω fields can serve as a measurement of ionization times on the attosecond timescale. Furthermore, interference structures such as intracycle interference and photoelectron holography are observed in two-colour ionization.

Liu and Barth report on advances of their work on the generation of spin-polarized photoelectrons by ionization of nitric oxide molecules with circularly polarized pulses. In the present paper, they show that an effective non-zero degree of spin polarization is found even when the molecules are randomly oriented. These results demonstrate convincingly that the earlier ideas by Barth and Smirnova can be generalized from atoms to molecules.

In the work by Rupp *et al.* on electron emission from few-cycle-pulse driven isolated nanoparticles, experiment and theory are combined to show that the many-particle charge interaction dominates over material dependence at sufficiently high near-field intensities. This leads to a clear physical picture for the generation of electron bunches from nanoparticles.

While the majority of articles of the special issue focus on multiphoton processes in low-frequency fields, Kumar Giri et al. discuss single-photon ionization in high-frequency fields, concentrating specifically on the effect of fluctuations. Deviations from a linear perturbative increase in the ionization probability with pulse energy and strong dependences on the pulse shape are observed at high-pulse energies. It is worth mentioning that the high-frequency response of matter will soon receive substantial attention when the first user experiments at the European X-Ray Free-Electron Laser will take place during the year 2017.

As noted briefly above, ultrashort bichromatic fields with specifically designed polarization profiles have emerged as a new twist for coherent manipulation of ultrafast electron dynamics in new applications including the generation of circularly polarized high-harmonic pulses and coherent control of electron directionality by multipath interferences. Kerbstadt et al. introduce a pulse-shaper based concept for the generation and measurement of bichromatic polarization-tailored femtosecond laser pulses. To this end, they extend established polarization-shaping techniques in order to sculpture orthogonal linearly polarized and counter-rotating circularly polarized bichromatic fields from the input spectrum. This approach allows to independently control the spectral amplitude, phase and polarization state of both colours and delivers bichromatic waveforms ideally suited for high-harmonic control.

Majety and Scrinzi report new findings on the ionization of helium atoms by a strong near-circular few-cycle field ('attoclock setting'). With their simulation, they show that electron–electron correlations cannot be responsible for the much-debated discrepancy between the theoretical and measured emission angle of photoelectrons. This result will fuel the ongoing debate about the correct interpretation of the attoclock data. It may also stimulate new experiments on this topic or alternative approaches to access time information in strong-field ionization.

The article by Golubev et al. comes back to the goal of controlled charge migration in molecules. They attack the problem using various types of quantum control, such as chirp or pulse sequences. They show that suitable two-pulse sequences can stop charge oscillations in propioloc acid and thus may be useful to trigger specific chemical reactions of the molecule. This work is one step in the process of understanding whether attochemistry will come true in the sense that the manipulation of initial ultrafast electron dynamics provides access to the control of subsequent reactions.

Braun et al. present a route towards a complete picture of strong-field coherent control experiments. In a series of two-colour pump-probe experiments they simultaneously study the transient and final state dynamics of potassium atoms driven strongly by intense chirped laser pulses. They observe population inversion and the creation of superposition states as well as the build up of adiabaticity and the transient dressing of the system. Their results highlight the importance of analysing non-perturbative light–matter interactions from different perspectives in order to establish a comprehensive understanding of physical mechanisms governing strong-field coherent control.

Besides isolated atoms and molecules, various types of condensed matter systems find increasing interest for application of tailored light fields. The above-mentioned isolated nanoparticles can be viewed as an extreme example of condensed matter. Another type of nanoobject is scrutinized in the work by Paschen et al. They investigate the two-colour-induced electron emission from tungsten nanotips as a function of the two-colour delay and an additional DC bias field. Nearly perfect modulation of the electron emission of 97.5% is found under optimized conditions, providing a perspective for controlled generation of femtosecond electron bunches from solid-state systems. Heidenreich et al. investigate light-induced charging dynamics of helium nanodroplets doped with different atomic species. The dynamics is governed by two entwined processes, namely ignition of a helium nanoplasma by dopant ionization and charging of the dopant atoms by the helium nanoplasma. In the experiment, they observe most efficient nanoplasma ignition and charging when helium droplets are doped with xenon atoms whereas only low charge states are measured for potassium and calcium atoms as dopants. This experimental study is complemented by classical molecular dynamics simulations to analyse the charging dynamics.

The generation of coherent radiation via high-harmonic generation is a well-known field of laser science. Recently, also the process of low-order or zero-order harmonic generation, including the terahertz range, has moved into the spotlight. Such emission, known as Brunel radiation, results from the charge current implied by the appearance of photoelectrons. This radiation is investigated theoretically by Babushkin et al. The authors demonstrate the possibility to manipulate the radiation spectrum by employing two-colour irradiation such as an ω-2ω field or a superposition of two incommensurate frequencies. The authors suggest that by variation of the frequency difference, one can recover aspects of either tunnel regime or multiphoton regime without changing the intensity.

An entirely different sort of tailored field is applied in the work by Wätzel et al. Their calculations demonstrate how optical vortices, i.e. light fields with orbital angular momentum, can generate unidirectional currents in different types of quantum systems, namely nanostructures and atoms, via Raman-type processes. The authors argue that in practice nanostructures may be more suitable than atoms for such a control scenario due to an easier alignment between target system and vortex field.

Seiffert et al. develop a model to describe adequately the emission of backscattered electrons from laser-ionized dielectric nanospheres. They generalize the well-known three-step model for small isolated systems such as atoms to the case of surfaces by adding a trapping potential that affects the maximal recollision energy crucially. The maximum energy of the backscattered photoelectrons is then found to be substantially above the well-known cut-off at 10 times the ponderomotive potential for isolated atoms. This model may become an important guideline for future studies of nanostructures in strong laser fields.

Paul et al. study the interaction of tailored intense light fields with asymmetric molecules motivated by the observation that photoionization of randomly oriented chiral molecules with circularly polarized light can generate forward/backward asymmetries in the electron distribution. In order to study the underlying physical mechanism of the asymmetries, they study photoionization from an asymmetric two-dimensional model system with intense, few-cycle, circularly polarized infrared pulses. By numerically solving the time-dependent Schrödinger equation, they find polarization-dependent asymmetries within the polarization plane. Although these asymmetries are essentially determined by the initial geometry of the nuclear charge configuration, it is demonstrated that they can be enhanced or weakened by ionization with few-cycle laser pulses.

At last, Pati et al. describe an intriguing phenomenon arising when two laser beams interfere in a close-to-collinear geometry. If the laser-induced grating consists of only a few grooves, the subcycle delay between the pulses can be used to control the shift between the grooves and their envelope, termed by the authors as 'groove-envelope phase'. The measured data on self-diffraction from such a grating in borosilicate glass is found to be in good agreement with calculations. These results may well become relevant for future attosecond studies in transparent solids.

Overall, this special issue demonstrates the diversity of systems and types of tailored fields that are used to manipulate matter on ultrashort timescales. In particular, the ability to shape light fields on subcycle timescales has turned out to be a powerful tool in quantum control. Quite unlike 20 years ago, refined polarization settings and intense phase-controlled two-colour fields are about to become established tools in ultrafast science. Presumably, we are just at the beginning of measuring and modelling a wealth of new exciting phenomena.

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