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EDITORIAL

## Attosecond technology(ies) and science

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## Editorial

# Attosecond technology(ies) and science

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## Abstract

Since 2001 and the first demonstrations of the feasibility of generating and measuring attosecond light pulses, attosecond science has developed into a very active and quickly evolving research field. Its ultimate goal is the real-time tracking of electron dynamics in all forms of matter, ranging from atoms and large molecules to the condensed phase and plasmas. The accomplishment of this goal has required and still calls for developments in ultrafast laser technology, ultrafast metrology, extreme ultra-violet (XUV) optics, pump–probe measurement schemes and non-linear laser-matter interaction. Moreover, the interpretation of the experimental results in attosecond experiments has stimulated and guided major developments in theoretical descriptions of ultrafast electronic processes in matter. Motivated by these two decades of development, several large-scale facilities, including extreme light infrastructure—attosecond light pulse source (ELI-ALPS) and several free electron laser facilities (the linac coherent light source (LCLS) at Stanford and the European XFEL in Hamburg) are now pushing the development of a new generation of attosecond sources. This considerable technological effort opens new and important perspectives in the field of ultrafast science with potential applications in photochemistry, photobiology and advanced electronics. In this context, the joint focus issue on Attosecond technology(ies) and science of *J. Phys. Photon.* and *J. Phys. B: At. Mol. Opt. Phys.* aims to provide an overview of the state-of-the-art in attosecond science, from the basic science involved in the generation and in applications of attosecond pulses to the technologies that are required.

Keywords: attosecond, technology, pump–probe spectroscopy

## 1. Historical development of attosecond science

While work on the development of attosecond sources continues to the present day, and includes novel approaches such as plasma mirrors and the generation of attosecond pulses at free electron lasers, the attosecond pulses that are used nowadays

in an ever-increasing number of laser laboratories around the world are produced by means of the process of high-harmonic generation (HHG) in atomic and molecular gases [1]. In HHG, strong field ionization (SFI) of an atomic or molecular gas launches free electrons that are accelerated under the influence of the oscillatory laser electric field. Upon re-collision with their parent ion, extreme ultra-violet (XUV) or soft x-ray

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photons are formed by electron–ion recombination [2, 3]. Given that the initial ionization occurs during a short time interval around a maximum of the laser electric field, the duration of the XUV emission is a small fraction of the duration of the optical period of the driving laser, i.e. in the attosecond domain.

The first attosecond pulses were demonstrated in 2001 [4, 5]. The main advance in these papers was that they reported a temporal characterization of the XUV radiation formed in the HHG process, demonstrating that it could lead to the formation of attosecond pulse trains (APTs) or isolated attosecond pulses (IAPs). Ever since, pulse characterization has been a persistent challenge in attosecond science. Attosecond pulse characterization serves as the initial step in the application of these pulses as the pump or probe of a fast-evolving process. Moreover, the temporal shape of attosecond pulses encodes valuable information regarding the generation mechanism itself, reflecting the sub-cycle evolution of electronic wave functions during their interaction with the strong laser field.

Well-established approaches for the temporal characterization of femtosecond pulses, applied in the visible and infrared (IR) spectral ranges, exploit nonlinear light–matter interactions in techniques such as frequency-resolved optical gating (FROG) [6], and spectral phase interferometry for direct electric-field reconstruction (SPIDER) [7]. However, their direct application to the attosecond XUV regime is challenging due to the large spectral bandwidth of these pulses, the high photon energies and the low photon flux. Entirely new approaches to diagnose bursts of XUV to soft x-ray light therefore had to be devised. Successful schemes typically involve a nonlinear interaction of a target with both the attosecond pulse and a co-propagating IR fundamental field. These schemes have been applied both in the weak IR field regime, in APT reconstruction by RABBITT (‘reconstruction of attosecond harmonic beating by interference of two-photon transitions’ [8]), and in the strong field regime, in streaking [9] schemes. In a streaking measurement, changes to the momentum distribution of photoelectrons are measured as a function of the delay between the XUV pulse and a co-propagating IR field [10, 11], while in a RABBITT measurement, interferences between two-color ionization pathways involving different harmonics are used to determine the relative phase of these harmonics [8]. Both schemes can be considered as extreme cases of a more general measurement approach called FROG-CRAB [12] (frequency-resolved optical gating for complete reconstruction of attosecond bursts). In the strong field regime, the attosecond pulse retrieval relies on a proper description of the strong field light–matter interaction. The commonly used strong-field approximation (SFA) [13, 14] neglects the Coulomb force and may lead to inaccuracies in the retrieved pulses.

Most current methods for pulse retrieval involve long integration times, and therefore require pulse-to-pulse repeatability. However, in many experimental scenarios pulse-to-pulse fluctuations and carrier envelope phase (CEP) instabilities are a major challenge. Reference [15] addresses this challenge and introduces a new retrieval algorithm, based on time-domain

ptychography. In the spirit of the work presented in [16], this algorithm allows the reconstruction of distributions of attosecond pulses and fundamental laser fields, enabling the application of the streaking scheme in the presence of pulse-to-pulse fluctuations. Finally, reference [17] addresses a method to overcome the low signal limitation in streaking experiments, by integrating the streaked electrons over a large angular range. The paper presents a theoretical scheme that allows the reconstruction of IAPs via the streaking approach from angular integrated spectra.

With the first attosecond pulses having been demonstrated in 2001, the first pump–probe experiments where attosecond pulses were applied to investigate time-dependent electron dynamics followed soon thereafter in 2002, with an investigation of the lifetime towards Auger decay of core-ionized Kr atoms [18]. These early experiments confirmed the ability of attosecond pump–probe spectroscopy to determine atomic properties such as excited state lifetimes by means of measurements in the time domain, and in agreement with results that had previously been obtained in the frequency domain. Subsequently, attosecond pump–probe spectroscopy was applied to explore questions that cannot be answered in the frequency domain, such as the nature of ionization processes in the strong field regime [19], and the question whether or not single-photon ionization from different atomic or molecular orbitals occurs simultaneously or with a small relative delay [20, 21]. Among other things, these experiments have led to a better understanding of what it means to talk about ‘ionization time’ in quantum mechanics [22].

In these, and many other early applications of attosecond pump–probe spectroscopy, the experimental approaches that were used were essentially the same as those first applied to prove the existence of attosecond pulses and to characterize their pulse duration. The outcome of RABBITT and streaking experiments depends on (i) the properties of the attosecond pulse(s), (ii) the properties of the co-propagating IR field and (iii) the properties of the sample. Hence, if two of these three properties are known, one can learn about the third one. Following this logic, the attosecond streak camera technique [9] was used in order to characterize the IR field [23], and both streaking and RABBITT were used to measure ionization time delays [20, 21]. Among other things, the RABBITT technique and related two-color ionization techniques have furthermore been used to measure both the real and the imaginary part of the ionization time of photoelectron wavepackets that are formed via multi-photon, strong-field tunnel-ionization [24], and to fully characterize photoelectron wavepackets that are formed when atoms are ionized by single photons from an attosecond pulse [25]. In the current special issue, Loriot *et al* present an extension of the RABBITT method to measurements of photoionization time delays in the N<sub>2</sub> molecule, interpreting an observed delay between ionization into the X and A molecular cationic states in terms of a shape resonance in the former channel [26].

Over time, the RABBITT and streaking methods have been complemented by additional measurement schemes, such as attosecond chronoscopy [19], attosecond transient absorption (ATAS) [27, 28] and attosecond quantum state holography [29,

30]. Among these, ATAS has proven to be particularly powerful, since it combines the exquisite time-resolution that is inherent in the use of attosecond pulses with an opportunity to perform experiments with element-specificity, exploiting the fact that attosecond XUV and soft x-ray pulses are preferably absorbed by core-electrons. This enables experiments where the dynamics under investigation is observed from the point-of-view of specific spectator atoms within the sample, and has been used to great effect in studies on both atoms [31, 32], molecules [33–35] and condensed phase samples [36–38]. Current efforts to extend ATAS aim particularly at the water window (282–533 eV) [39, 40], where transient absorption around the C-, N- and O-edge can be exploited in experiments on organic molecules [41, 42] and organic semiconductors [38].

The development of attosecond science as a pump–probe spectroscopy has been complemented by the development of high harmonic spectroscopy (HHS), which aims at extracting information about a target from the analysis of its high harmonic spectrum. The appeal of HHS stems from the simplicity of its implementation, independent of whether the target is an atom, molecule, liquid [43, 44] or solid [45–47]. HHS carries the potential of combining sub-Ångstrom spatial and attosecond temporal resolution of electronic structures and dynamics. The spatial resolution originates from the Ångstrom scale of the de Broglie wavelength of the recombining electron [48], while the attosecond temporal resolution is provided by the sub-cycle nature of the re-collision process itself. HHS can be applied as a pump–probe measurement scheme: field-induced tunneling can serve as a ‘pump’ preparing a hole wave function in the sample, while the re-collision serves as a ‘probe’, mapping the evolved wave function into the HHG spectrum [49]. This idea underlies a large number of experiments in both atomic and molecular systems that resolve the structure of the contributing orbitals [50–52], hole wave function dynamics [53–56], nuclear dynamics [57–60], quantum path interferences in atomic media [61], mapping of electronic coherences [62], charge migration in molecules [60, 63], scaling laws in liquids and solids [64, 65], and recently the identification of singular points in the band structure of solids [66]. A complicating factor for the interpretation of the HHS measurement is the strong field interaction during the HHG. The understanding of the interaction has improved dramatically during the last decade and advanced theoretical and measurement methods [66–69] are nowadays able to extract a wealth of information from HHS experiments [70–74].

In addition to recombination with the molecular ion, the re-colliding electron can also scatter off its parent ion [75–77]. This scattering process encodes the molecular structure in the angular distribution of the re-scattered electron. The technique has become known under the name laser-induced electron diffraction (LIED) [78–83] and has been used to determine static molecular structures [84, 85] and the deformation of C<sub>60</sub> [86] with few-picometer spatial resolution. The method was developed to image isolated molecules with a single electron [87], and it has provided accurate imaging of aligned molecules [83], first snapshots of bond-breakage and depro-

tonation [88], and stretching and bending of a molecule [89]. Moreover, the method was recently used to reveal dissociation of a diatomic molecule [90].

## 2. Laser sources

The development of advanced laser sources was an essential prerequisite for the emergence of attosecond science, and continues to be a decisive factor for the advance of the field and the emergence of entirely new classes of experiments. This is easy to understand when considering that electron re-collision in a strong laser field is at the core of most attosecond investigations and technologies. Thus, the degree of control over the electric field waveform from a laser, its wavelength, polarization, peak amplitude and spatio-temporal shape directly impacts the range of possible investigations and their outcomes [91].

The early development of attosecond science was entirely based on chirped-pulse amplification-based Ti:Sapphire technology and received a major impetus with the ability to generate few-cycle pulses with carrier-to-envelope phase (CEP) stability [92, 93] at high peak intensity. It is interesting to recall that while the ultrafast optics community pursued the goal of producing ultrashort pulse waveforms, the metrology community aimed at producing optical frequency combs—both are just two sides of the same coin. The ability to generate such CEP-stable and intense few-cycle pulses [94] immediately led to the advent of IAPs in the XUV region [11, 95]. The ability to control the number of laser cycles and the CEP moreover led to an improved understanding of the electron’s continuum and re-collision dynamics [96] and the underlying quantum trajectories [97]. CEP-controlled few-cycles pulses were also used in first experiments controlling molecular electron dynamics on attosecond timescales [98].

A second breakthrough came with the ability to generate CEP-stable few-cycle pulses at mid-infrared wavelengths [99, 100]. Such long wavelengths enable a ponderomotive scaling of the strong field interaction due to the much longer excursion of the tunnel-ionized electron [76, 101] and correspondingly higher kinetic energies upon re-collision, directly impacting the range of photon energies produced in HHG. For long wavelengths, the variation of the laser cycle is slow compared to the bound wave packet dynamics. This places the strong-field interaction in the quasi-static regime [87, 102] for which theories provide accurate predictions [103–105], thus allowing to unambiguously probe fundamental tunneling physics [106–108]. HHG results in a spectrum with a cut-off that scales with  $\lambda^2$  and hence HHG with longer wavelength lasers provides a route towards tabletop coherent x-rays in the keV range [38, 109, 110]. More advanced schemes based on the synthesis of fields in the IR have also been developed for the optimization of the attosecond pulse generation process [111]. Nowadays, IAPs in the soft x-ray water-window [112–114] can be generated, and LIED [82, 83] constitutes a powerful probe to study molecular structure with attosecond temporal and Angstrom-scale spatial resolution. Moreover, the availability to generate low photon energies in the

mid-IR has opened up the field of HHG in solids, allowing to probe complex dynamics in the solid state, as will be discussed below [64].

A third breakthrough came with the development of solid-state-, fiber- and OPCPA-based systems that combine high peak power with high average power [115–118]. These systems nowadays permit to conduct experiments at 1–3 orders of magnitude higher repetition rates than the first generation, 1 kHz Ti:Sapphire systems. The high repetition rate creates the ability to achieve unprecedented signal-to-noise ratios in a plethora of investigations, as well as the implementation of attosecond experiments with coincident detection of all electrons and ions [15, 119]. In addition, OPCPA enables the generation of attosecond pulses and pulse trains that are sufficiently intense to permit XUV pump–XUV probe experiments [120, 121]. Moreover, the increased reliability of these systems enables scientists from other communities to benefit from attosecond technologies without themselves needing to be specialists of laser technology.

### 3. Strong-field ionization (SFI)

One of the most fundamental nonlinear phenomena encountered in attosecond science is strong field tunnel ionization. Field-induced tunneling is the starting point of attosecond science, initiating a broad range of strong field phenomena—from HHG to electron holography or LIED. In the presence of a strong laser field, the Coulomb barrier of a bound electron is suppressed, leading to tunnel ionization within a fraction of the optical cycle [122]. Tunneling dynamics as determined by the rapid sub-cycle modification of the electromagnetic field has a natural time scale in the attosecond regime. We can classify this dynamics by the well-known Keldysh parameter,  $\gamma = \omega \sqrt{2IP}/E$ , (with the laser frequency  $\omega$ , the ionization potential IP, and the laser field amplitude  $E$ ). The quasi-static regime corresponds to  $\gamma \ll 1$ , where the electron tunnels through a static barrier, having a temporal shape dictated by the instantaneous amplitude of the electric field. As  $\gamma$  approaches 1, the rapid oscillations of the laser field excite the electronic wave function as it propagates under the tunneling barrier via non-adiabatic dynamics [123].

Quantum tunneling raises fundamental questions such as the ability to measure, or simply define, the tunneling time. These questions have been the subject of numerous discussions since the early days of quantum mechanics [124–127]. Attosecond metrology enables to re-address these concepts, probing the temporal evolution of field-induced tunneling on its natural time scale [19, 128]. One of the most successful schemes is the attoclock technique [129–134], in which an elliptically polarized laser pulse acts as the rotating hand of a clock, which maps the instant of birth of the electron onto its momentum distribution. Within this special issue, references [135, 136] present a broad review focused on the different definitions of the tunneling times, the historical debate concerning their measurements as well as the experimental observations performed by attoclock experiments. High harmonic spectroscopy (HHS), initiated by the tunneling mechanism,

serves as an alternative accurate probe of its basic properties. HHG two-color metrology has been applied to resolve the time at which the electron exits the tunneling barrier [137] and the evolution of the instantaneous tunneling probability within the optical cycle [24].

While attosecond metrology re-opened the study of quantum tunneling, most of the measurements were focused on the simple case of a single active electron that tunnels through an atomic barrier. Many fundamental questions have remained open: what is the role of multi-electron effects? How does the structure of the original orbital influence the spatio-temporal properties of the tunneled electron? Can we resolve relativistic effects? When tunneling evolves from a molecular system, the structure of the molecular orbital can play an important role, dictating the angular distribution of the ionized electronic wave function [138–140]. Furthermore, the tunneling process may occur from several molecular orbitals simultaneously via multi-channel ionization [141]. These ionization pathways can be strongly correlated as they interact with the strong laser field [52, 54, 56].

Within the current special issue, reference [142] shows that the simultaneous measurement of field-induced tunneling and HHS isolates both the structural dependence of the tunneling process and its multi-channel nature. Reference [143] presents a theoretical study of anisotropic tunnel ionization in polar molecules and analyzes the multi-electron dynamics induced by the laser field. Field-induced tunneling in molecular systems can be strongly influenced by nuclear dynamics. For example, in small molecular systems the internuclear distance may change substantially on a sub-cycle time scale [144]. Reference [145] studies the role of nuclear movement in the tunneling ionization of hydrogen molecular ions and its influence on the ionized electron's momentum distribution. In addition, competition between several strong field channels can be observed. Reference [146] shows that when tunneling is induced by a two-color field a control over two channels, field-induced tunneling and dissociation, is achieved.

Upon increase of the intensity of the laser field, relativistic effects play an important role, and the description of the underlying dynamics needs to go beyond the dipole approximation [147–149]. Reference [150] presents a review of the combined influence of non-dipole and Coulomb focusing effects observed via experimental and theoretical investigations. Finally, strong field light–matter interactions may lead to multiple ionization, commonly via sequential tunnel ionization of charge states with increasing ionization potential. While in most atomic and molecular systems this process is assumed to evolve by means of a sequence of one-electron processes, in complex systems such as clusters and nano-tips, multi-electron phenomena can play an important role. Reference [151] describes a theoretical analysis that reveals the basic mechanism in such systems. The paper shows that when ionization potentials of subsequent charge states are close to each other, multiple tunneling events can evolve on a sub-cycle timescale, so that the tunneling of one electron can block the next tunneling electron.

## 4. Atomic photoionization

Attosecond science reveals dynamics over a broad range of laser intensities—from strong field phenomena, such as field-induced tunneling, to weak field processes. Single-photon ionization is one of the most fundamental light–matter interactions in nature, projecting bound wave functions onto ionized electronic wave functions, which can then be experimentally resolved. By probing the emitted electron, one can decode the full dynamics of the photoionization process. Attosecond science revolutionized our ability to measure photoionization dynamics in a large range of systems, providing direct insight into the basic properties of the mechanism. One fundamental question, extensively studied over the past decade is: how long does it take for an electron to photoionize? The photoionization delay—also known as the Wigner delay [152], is determined by the phase shift of the wave function associated with the photoelectron emerging from the binding potential, compared with the wave function of a freely propagating electron. First experiments resolved the relative photoemission time delay between photoelectrons originating from two distinct energy levels by applying the streaking [20] and the RABBITT [21] techniques. Performing an angle-resolved RABBITT measurement [153] reveals diverse fingerprints of the attosecond photoionization dynamics [154, 155]. Within this special issue, reference [156] reports angle-resolved studies of XUV-IR ionization in a RABBITT scheme using a COLTRIMS apparatus and resolves the ionization dynamics of valence orbitals in atomic systems. Extending this measurement from atoms to molecules encodes rich information associated with the anisotropic nature of the molecular potential. Such measurements have revealed delays associated with different vibrational states of a molecular ion [157] or delays in chiral photoionization [158].

When photoionization evolves in the presence of a low frequency laser field, its basic properties can be altered, leading to the appearance of new interference features. This laser-assisted photoelectric effect encodes rich dynamical and structural information. Early on, Johnsson *et al* demonstrated that a two-color XUV-IR field can be used to manipulate the ionization yield of helium on a sub-cycle timescale [159]. In a frequency domain picture, these oscillations may be understood as the result of an interference between ionization pathways via different intermediate states dressed by the IR field [160]. Reference [161] describes a combined experimental and theoretical study of XUV ionization of atomic argon in the presence of an IR laser field, leading to the appearance of photoelectron sidebands that originate from intra-cycle interference. The paper demonstrates the ability to recover these interferences, which cannot be commonly resolved due to focal volume averaging, via a velocity map imaging measurement. Reference [162] performs a systematic study of IR-assisted XUV photoionization of atomic helium and resolves population transfer to high angular momentum states. Changing the IR intensity and fundamental wavelength, while performing energy and angle-resolved photoelectron measurements, enabled a determination of residual populations of Rydberg states created by the two-color field.

As mentioned, the instantaneous response of a system dressed by the IR field can be resolved via attosecond transient absorption spectroscopy (ATAS). This scheme has been applied to resolve fundamental attosecond timescale phenomena in a wide range of systems, such as the dynamics of valence shells [27], core shells [33], excited states [163, 164], autoionizing resonances [32] and light-induced states [165] in both atoms, molecules and solids [37, 166].

Within this special issue, reference [167] applies ATAS to investigate the laser field-modified dipole response of the continuum threshold of helium. The paper resolves light-induced time-dependent structures in the absorption spectrum, below and above the ionization threshold, and isolates the contributions of the unbound electron to these structures. As this experimental scheme evolves beyond atomic systems, new aspects appear, reflecting the anisotropic nature of more complex structures such as molecules. [168] studies ATAS in systems without inversion symmetry. Performing a numerical and analytical analysis, the paper shows that the lack of parity conservation leads to new spectral absorption features, which reveal the symmetry properties of both the system under investigation and the laser field itself, as dictated by the carrier envelope phase.

## 5. Attosecond molecular physics

Compared to atoms, the application of attosecond techniques to molecular systems significantly increases the number of degrees of freedom and the range of timescales that need to be considered. Whereas in atomic experiments with attosecond pulses, the relevant timescales are either optical (the duration of the laser pulses used, or the duration of the optical cycle of these pulses) or electronic (i.e. related to electronic energy level spacings in the atom), molecules have vibration and rotation as additional degrees of freedom, with associated timescales. Although these timescales are generally longer than the attosecond timescale, interesting situations nevertheless arise when molecules containing light atoms are investigated, where electronic and nuclear motion can become strongly coupled. The same is true, on longer timescales, for molecular dynamics involving conical intersections.

The development of attosecond science has prompted novel ideas towards realization of the longstanding dream of laser-controlled chemistry [169]. The duration of attosecond pulses is so short that during an attosecond pulse the position of even the lightest atoms may be considered ‘frozen’. The initial response of molecules to attosecond pulses is therefore a purely electronic charge migration process [170, 171], potentially leading to so-called ‘charge-directed reactivity’ [172]. While ideas on attosecond charge migration and attosecond to few-femtosecond coupled electronic and nuclear dynamics were formulated in theoretical papers soon after the first laboratory demonstration of attosecond pulses [173], the first experimental investigations of charge migration and ultrafast coupled electronic and nuclear dynamics in molecules have been made in recent years. Selected highlights have been the observation of electron localization in dissociating  $\text{H}_2^+$  molecules [174] and the observation of coherent electronic

dynamics in photo-ionized phenylalanine [175] and tryptophane [176].

In the current special issue, in an example of attosecond XUV pump–XUV probe spectroscopy, Fukahori *et al* [120] report experiments on electron-nuclear wave packet dynamics in O<sub>2</sub>, using two time-delayed APTs. They measure the O<sup>+</sup> fragment kinetic energy distribution as a function of the delay between the two pulses, and observe out-of-phase  $2\omega$ -oscillations at high and low fragment kinetic energy, which are interpreted in terms of probing (by the XUV probe laser) of an electron-nuclear wave packet consisting of the coherent superposition of two cationic electronic states (both produced by the XUV pump laser). Among several theoretical studies in this special issue, Komarova *et al* [177] report on the coupled electron-nuclear dynamics of LiH, LiD and LiT under the influence of an intense, CEP-stable, few-cycle near-IR (720 nm wavelength) laser field. The CEP controls the initial population of excited states, which in turn controls the ionization and the effect that two regions where non-adiabatic coupling is important have on the final excited state populations. Arnold *et al* consider the role of the photoelectron that is ejected in the course of ionization by an attosecond pulse on the coherence properties of superpositions of cationic electronic states [178]. They include the photoelectron as a point charge in a quantum–classical simulation of the attosecond ionization process and observe that the photoelectron causes a partial decoherence within the first 50 attoseconds, with this timescale being independent of the spectral range of the ionizing pulse. Finally, while most studies have so far focused on the single electron response, correlated electron–electron dynamics can play an important role. Reference [179] performs a theoretical study of electron–electron correlation as well as non-adiabatic effects in post-ionization dynamics of molecular systems. Applying a charge-transfer molecular model system, the paper distinguishes different pathways leading to ionization, both direct ionization and ionization involving elastic and inelastic electron scattering processes.

While these research works targeting ‘atto-chemistry’ are all examples of attosecond pump–probe spectroscopy, with attosecond pulses initiating (pump) and/or observing (probe) attosecond time-scale dynamics, molecular electronic and/or nuclear dynamics have also been extracted by means of HHS [50] or from the angular distributions of re-scattered photoelectrons in SFI and LIED experiments [80]. For example, measurements of high-harmonic spectra as a function of the orientation of a molecular sample with respect to the polarization axis of the driving laser have demonstrated that ionization of molecules by a strong laser field can lead to the formation of a coherent superposition of electronic states, which is probed by the re-colliding electron within the HHG process [55]. And, as mentioned, the re-colliding electron can scatter off the molecular ion, providing the basis for LIED as a method to determine static molecular structures and molecular dynamics in the short time interval between ionization and re-collision [82, 86, 88, 89]. Another recent topic in attosecond science is the investigation of chiral molecules. Whereas photoionization of chiral molecules by circularly polarized light is

characterized by different probabilities for forward and backward emission along the laser propagation axis, it was determined using an approach similar to RABBITT that delays of a few tens of attoseconds exist between these two emission directions [158].

Presently, molecular HHS and SFI are active fields of research. Correspondingly, several representative examples are found within this special issue. In a topical review, Zhang *et al* give an overview of comprehensive research that has been carried on strong-field dissociative ionization and Coulomb explosion of the hydrogen molecule, using coincident electron–ion detection to obtain detailed and photon-number resolved insight into energy-sharing between the electrons and the nuclei, directional bond-breaking and the formation of Rydberg states [180].

## 6. HHG control

Within recent years, a superb control over the fundamental properties of high harmonics has been demonstrated. One of the main challenges that has recently drawn significant attention is the ability to manipulate the polarization state of the harmonics, which are naturally produced with linear polarization. An additional challenge is high-resolution spectral manipulation over the harmonic spectrum, which spans over a large spectral range. In addition, the lack of optical components in the XUV or x-ray regime [181], calls for control schemes to manipulate the spatial profile of the HHG beam. High harmonic generation with two-color bi-circular fields [182] opened a range of opportunities from the first demonstration of three-foil symmetric XUV generation [183] to today’s intricate control over the spin orbital momentum (polarization) and orbital angular momentum of the XUV waveform [69]. More elaborate schemes such as the combination of two non-collinearly interacting beams with counter-rotating elliptical fields are shown to provide a spatially varying polarization grating which may permit to interrogate spin properties of materials in space and time, all at once [184].

A persistent challenge in attosecond science is the efficiency of the high harmonic generation process, which can be manipulated on the microscopic single-atom-level [61] and through macroscopic phase matching [185]. To this end, a plethora of schemes have been implemented to control phase matching (see for example [186–188]) with the common difficulty to engineer schemes to the short coherence length of the HHG process. Making use of tailored micro-machined chips, improved phase matching has recently resulted in a 20 times increased yield for XUV harmonics [189]. Despite these achievements, one of the challenges associated with the use of gas-phase HHG as a source of attosecond pulses remains the relatively low efficiency, with typical conversion efficiencies rarely higher than  $10^{-4}$  and often much lower.

## 7. Plasma dynamics

Because of the low efficiency of gas phase HHG, alternative strategies for the production of attosecond pulses have



targeted plasmas and bulk media. The production of a plasma upon irradiation of a surface by a laser pulse with near or beyond relativistic intensity produces harmonic radiation by coherent wake emission (CWE) [190]—a mechanism that presents some analogy with the re-collision process of HHG in gases—and the relativistic oscillating mirror (ROM) mechanism [191]. In the CWE regime, both attosecond phase-locking [192] and the production of IAPs by means of the attosecond lighthouse effect [193] have been demonstrated. Beyond the development of attosecond sources, and in analogy to HHS in atoms or molecules, the harmonic signal generated from plasmas carries rich information on the plasma properties and its dynamics in intense laser fields, down to the attosecond time scale.

While most experiments on HHG in plasmas have so far been based on the use of multi-TW Ti–Sapphire lasers, this research is now also being pursued with few-cycle OPCPA systems and/or laser pulses that are post-compressed in a hollow core fiber. In [194] Böhle *et al* report progress on the development of an intense IAP source using a CEP-stabilized, TW-level, 3.5 fs laser to drive the ROM mechanism. Near-continuous XUV spectra are reported that are indicative of IAP production for most values of the CEP, with a theoretically predicted percent-level generation efficiency. In addition to being of interest as a possible source of attosecond laser pulses, plasmas are also of great interest in attosecond science in and of themselves. In [195] Kruse *et al* report a novel computational scheme that permits the calculation of coherent diffraction images for the scattering of intense, few-femtosecond XUV pulses from sub-micron He droplets that are converted into plasmas under the influence of the incoming radiation [196]. The required XUV pulses have recently become available in the laboratory [121]. While propagating through the droplet, the XUV pulses induce Rabi-cycling on a few-femtosecond timescale, which leaves its mark in the calculated diffraction images that can be measured.

## 8. High harmonic generation in solids

Whereas HHG in plasmas that are formed on surfaces as described above has a relatively long history, a major, more recent discovery, with far-reaching implications, has been the discovery of HHG in the bulk of solid materials [64]. While in the gas phase the strong field dynamics is described in a universal manner, where electron trajectories propagate in the continuum, in solids the dynamics is strongly dominated by the structural properties of the target itself during all steps of the interaction.

As discussed, the recent success of HHG in solids was made possible by the availability of mid-infrared light sources, since they permit generating harmonic emission below the correspondingly lower ionization threshold of solid-state materials (compared to gaseous media). Due to the availability of this technology, we are nowadays able to study attosecond dynamics and strong field effects in solids without the need for expensive ultrahigh vacuum setups. Since the first observation of HHG in solids [64], solid-state HHS has opened a window into the electronic structure and dynamics in crystals

[45, 46, 66, 197–204], resolving complex many-body [205] or topological phenomena [206]. Still, despite the spectacular progress, many of the underlying generation mechanisms are still hotly debated. Several possible mechanisms have been introduced, describing the process as originating from intra-band currents [64], or as stemming from inter-band currents [197]. In the intra-band picture, the nonlinear harmonic emission results from the nonlinearity of the band itself. The inter-band model can be described in close analogy to the three-step mechanism in a gas phase system. Around the peak of the laser field, an electron tunnels from the valence to the conduction band, forming an electron–hole pair. The laser field subsequently accelerates the pair, leading to their recombination and the emission of extreme ultraviolet (XUV) radiation. As demonstrated in a recent paper [207], the electron-tunneling process that transfers the electron from the valence to the conduction band *itself* may also be a dominant source of harmonic emission.

Within this special issue, reference [208] performs a theoretical study, based on semiconductor-Bloch equations, to describe ultrashort pulse propagation in a three-band solid state system. This model captures the sub-cycle inter-band polarization and intra-band current, showing the important role of virtual currents and dynamic Bloch oscillations. Reference [209] studies the underlying recombination mechanism in MgO, by applying a two-color probing scheme. This measurement reveals that harmonics emitted from the input surface can be associated with re-colliding electron–hole pair trajectories. In contrast, harmonics emitted from the exit surface of the crystal are strongly distorted due to nonlinear propagation of the strong laser field within the sample. [210] presents an alternative approach to measure the underlying HHG dynamics in solids. When the HHG process is generated by a few-cycle pulse, the narrow envelope of the pulse can serve as a gating function. Resolving the CEP dependence of the harmonics then enables the reconstruction of the attosecond pulse’s temporal shape. Applying this approach to measure harmonic emission in MgO resolves a group delay shift in a region of strong absorption, associated with the quantum interference between the continuum and bound states that participate in the generation process.

These achievements are occurring during a time in which material science has made a tremendous leap that has made 2D materials available and that has led to the existence of technologies where these materials can be assembled in a controlled fashion much alike Lego stones. It is very likely that the combined availability of an ultrafast probe of electronic and nuclear dynamics in combination with advanced materials will provide a tremendous boost to material science. At the present time, we have already seen hallmark experimental investigations such as the elucidation of quasi-particle collisions in transition metal dichalcogenides (TMDCs) [211], measurement of Dirac currents in graphene [204, 212, 213], and theoretical predictions on the nonlinear response [214], phase transitions and topological effects [206, 215–217] in 2D materials. Clearly, the carrier dynamics in the 2D material graphene [204] provides a rich ground for investigations since it combines control over the existence of a bandgap with control over

pathway interferences of Dirac carriers [218]. Carrier transport is of major importance for the development of real-world devices which are limited by heat dissipation due to scattering and ballistic transport distances [219]. To address such problems, the combination of the high temporal resolution of high harmonic-based VUV and XUV sources with photoemission spectroscopies [220] provide novel insight into the scattering and screening dynamics in solids.

## 9. Outlook

As we have tried to indicate in this introductory overview, and as may be recognized from the impressive number of papers that were submitted to this special issue on attosecond technologies, the field of attosecond science has—in the last two decades—rapidly developed from a specialized research activity that could only be pursued in a very limited number of specialized laser laboratories, into a powerful new research area capable of addressing important research questions in the four fundamental states of matter: gas, liquid, solid and plasma. It is tempting to try to foresee how attosecond technologies and attosecond science will continue to develop in the coming years.

In terms of technology, we anticipate that further development of solid state, fiber- and OPCPA-based systems will allow attosecond sources to become cheaper and more reliable, while at the same time improving the source characteristics in terms of repetition rate and available photon fluxes. At the same time, advances in the technology of free electron lasers will certainly lead to the generation of intense attosecond bursts accessible for non-specialists [221, 222]. Additionally, if the fluxes that are achieved in solid state HHG can be brought to the level that is achieved in gas phase HHG, then this would allow significant simplification of the experimental setups that are needed for attosecond science experiments, with accompanying advantages in terms of cost, reliability and accessibility. Finally, attosecond pulses generated from plasmas using the emerging generation of femtosecond PW lasers could provide the light intensities required for the experimental investigation of quantum electrodynamics in the strong field regime [223, 224].

Another key aspect of the future development of attosecond technology will certainly be the pursuit of attosecond pulse generation at higher photon energies than has thus far been the case. Attosecond-resolved core-level x-ray absorption spectroscopy is a powerful spectroscopic technique, which is based on the x-ray-initiated transition of a core-level electron to an unoccupied bound or continuum state. This method is ideally suited to fully exploit the large spectral bandwidth of the attosecond pulse, since the energy resolution of the measurement is determined by the spectrometer resolution. However, an important aspect for the interpretation of such measurements is the fact that the x-ray-initiated core hole can strongly alter the distribution of measured valence states, thus changing the absorption spectrum and complicating its interpretation. Core-level x-ray absorption spectroscopy [225–227] is well established (mainly) at synchrotron light sources in which hard x-rays access 1s (K), 2s (L1) or 2p (L2, 3)

core-transitions to interrogate a wide variety of gases, liquids or solids. X-ray absorption near-edge spectroscopy (XANES), also called near-edge x-ray absorption spectroscopy (NEXAFS), interprets the immediate energetic vicinity of the absorption edge and, thus, provides information about electronic bound states. The unambiguous interpretation of spectra necessitates using *K* or *L* transitions, which translates into x-ray photon energies in the soft and hard x-ray domain. Higher-lying states (*M*, *N*) may be used as well, but the interpretation needs advanced theory to address final state multiplet effects, which otherwise quickly obscure the analysis [228]. A challenge for attosecond technology has therefore been the ponderomotive scaling of the photon energies to the soft x-ray regime to reach *K* and *L* shell absorption edges of interest. This technology is nowadays available [38, 39, 41, 113, 114], which makes x-ray absorption spectroscopy an ideal tool to benefit from the incredible time resolution on the attosecond scale. Provided the attosecond pulse's spectrum is broadband enough, it may even probe an extended spectral region beyond the absorption edge, which is known as extended x-ray absorption spectroscopy (EXAFS) [229]. EXAFS arises from the scattering of the continuum electron on nearest-neighbor atoms, and can thus provide molecular, or lattice, distances in addition to the electronic structure from XANES. Both, XANES and EXAFS benefit from the incredible temporal resolution provided by attosecond technology. After the first demonstration of the viability for high resolution soft x-ray spectroscopy [37, 38], a range of experiments showed its capabilities with investigations in solids [165, 230–234] and molecules [41, 235], and the possibility to combine XANES with EXAFS [229].

Besides considering the future development of attosecond technologies, it is an intriguing question to consider in which research fields attosecond science may—in future—have a significant impact. Here, admittedly strongly influenced by our subjective view of the attosecond science research field, we offer the following suggestions:

In the coming years, attosecond science may develop into a platform for studies of quantum information. Though perhaps surprising at first, the reasons for this assertion are in fact rather simple. The short wavelength and short optical period of attosecond pulses place attosecond radiation in the XUV and soft x-ray regime. This means that attosecond pulses are by definition ionizing radiation. A peculiar aspect of ionization is that it splits a quantum system (e.g. an initially neutral atom or molecule) into a composite quantum system, consisting of an ion and an electron. More often than not the ion and the photoelectron will be entangled, meaning that they cannot be described by a single product wave function and meaning that measurements on the ion will influence what is measurable in the photoelectron, and vice versa [236]. Similarly, two entangled electrons ejected from the same ion can carry signatures of their entanglement in the photoemission signal. In this case attosecond technology can be used to study the interference resulting from this entanglement and the influence of the inter-electronic Coulomb interaction on the coherence of the entangled electron. The role of entanglement is well-established in quantum information, and arguably underpins

its rapid development in recent years. However, in attosecond science, the role of entanglement, and its influence on the coherences that can be observed in pump–probe experiments, has thus far hardly received any attention. We may anticipate that this is going to change substantially in the coming years.

–As mentioned in the introductory text, preliminary steps have been done in the direction of investigating processes of interest for photochemistry and photobiology. Nevertheless, the molecules studied so far only constitute small building blocks of more complex molecules of biological relevance. We anticipate that advances in attosecond technology will allow for more complex targets such as DNA and proteins to be investigated in their natural environment (i.e. solvated in water). In this context, methods such as XANES and NEXAFS (see above) will be extended to the attosecond domain to identify how mechanisms related to electronic correlations (i.e. charge migration, shake-up, Auger and interatomic Coulombic decay (ICD) [237]) may affect the final functionality of the macro-molecule. Similarly, control over the electronic time scale could be exerted to optimize photocatalytic reactions for instance using transition-metal complexes. Attosecond pulses can be used not only to map changes of the electron densities in the metal center and the surrounding atoms, but also to directly determine the rate of the intersystem crossing (ISC). ISC is an extremely fast process in which a singlet excited electronic state makes a transition to a triplet excited state due to spin–orbit interaction. There is still a lot of debate on how ISC relates to the structural change around the metal center and gaining direct access to it will allow for a better optimization of the photocatalytic process.

Based on all of the above, we conclude that the success story of attosecond science has really only just begun, and that the field has a bright future ahead, where existing and newly developed attosecond methods will be used in an ever-increasing number of laboratories. Like the individual frames of the movies that are recorded in attosecond pump–probe experiments, the current collection of articles in the joint focus issue on attosecond technology(ies) and science of *J. Phys. Photon.* and *J. Phys. B: At. Mol. Opt. Phys.* should be considered as a snapshot in time, and we anticipate with great interest how the movie continues from hereon.

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