

INVITED ARTICLE

Resolving intra-atomic electron dynamics with attosecond transient absorption spectroscopy

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Attosecond transient absorption spectroscopy is a recent addition to the experimental tool set of attosecond science. This alloptical method measures different observables than the previously existing techniques based on electron and ion detection and overcomes several of their limitations. We review the present state-of-the-art of attosecond transient absorption experiments and theory. Applications cover the exploration of ultrafast electron dynamics in atoms as well as in solid-state systems. As an example we discuss the observation of transiently bound electron wavepacket dynamics in helium in more detail. This example illustrates how transient absorption spectroscopy can provide information that is fundamentally inaccessible to the techniques based on ionisation, namely dynamics occurring well below the ionisation threshold. Furthermore, we show that a model based on wavepacket interference and originally developed to explain modulations in the ion yield is not sufficient to explain the observed optical response of the system. The optical response on extremely short timescales and in systems exposed to strong laser fields is still not fully understood. This makes the method also attractive for fundamental studies. Furthermore, it is expected that the technique will also find future applications for studying molecular systems in gas phase, in solution, or as solids and will greatly benefit from the advances of ultrafast lasers with multi-100-W average power improving signal-to-noise ratio by many orders of magnitude in the near future.

Keywords: attosecond; electron dynamics; transient absorption

1. Introduction

The attosecond timescale (1 as $= 10^{-18}$ s) constitutes the current frontier in ultrafast time-resolved experiments. Attoseconds represent the natural timescale of fast electron dynamics in atoms, molecules and even solid-state systems. At the same time, these extremely short timescales force us to rethink old problems and put into question the concept of instantaneousness in many physical processes. Prominent examples are the currently intensively debated issues of tunnelling and ionisation times [1–5].

While attosecond science has already shed new light on several very fundamental processes in physics, the field itself can still be considered to be in its infancy – despite its more than 10-year-old history [6]. Reasons for this are the complexity and limitations of the existing experimental tools. For example, they all require the presence of nonperturbatively strong laser fields [7,8]. This renders it intrinsically impossible to study a system in a more natural, field-free environment and in addition often fundamentally limits the attainable time resolution.

Strong-field laser matter interactions are therefore an ingredient of every present attosecond measurement technique. 'Strong field' refers in this context to laser peak intensities on the order of 10^{13} W/cm² to 10^{15} W/cm². At such intensities, the electric field in the laser pulse reaches field strengths comparable to inner-atomic fields. As a

result, the laser field becomes capable of ionising atoms even if the photon energy amounts to only a small fraction of the ionisation potential and therefore cannot be considered a perturbation anymore.

Most attosecond spectroscopy techniques do not only expose the system to be studied to strong laser fields but also detect charged particles as the main signal, i.e. they require ionisation of the system being studied. Methods that found the most widespread use are for example the attosecond streak camera [7], the attoclock [3] and the RABBITT (resolution of attosecond beating by interference of twophoton transitions) technique [8,9]. These come at the disadvantage, particularly for more complex targets under test, that the intrinsically required strong laser fields considerably distort the system under test, very often in an unknown dynamic complexity. Detection of charged particles limits count rates (due to space charge effects) and is susceptible to stray fields. The strong near-infrared (NIR) laser field can additionally generate an unwanted signal background due to above-threshold ionisation. For easy interpretation of the measured data, the attosecond streak camera requires the system to be studied to have a relatively simple energetic structure. It should be noted that field strengths required for the RABBITT technique can be one-to-two orders of magnitude lower than for the other methods, which relaxes some of the constraints listed above.

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The all-optical technique of attosecond transient absorption spectroscopy overcomes all of these limitations. Transient absorption has already been a well-established and very successful technique in the femtosecond regime. It detects the changes in the optical response of a system with a (usually weak) probe pulse as a function of delay with respect to a pump pulse that serves to initiate the dynamics to be studied. Three different groups independently introduced this method in the attosecond domain in 2010 and 2011 [10-12]. In our laboratory, we used transient absorption spectroscopy to track the interference of transiently bound electron wavepackets close to the ionisation threshold of helium [12]. However, while attosecond transient absorption spectroscopy does not intrinsically require a strong IR laser field, all experiments performed so far were two-colour pump-probe schemes involving such a field.

2. Attosecond transient absorption spectroscopy

Transient absorption spectroscopy is nothing new in ultrafast science. It is in widespread use in the femtosecond domain since decades. A first step towards its attosecond incarnation was the extension into the extreme ultraviolet (XUV) spectral domain using high-order harmonic generation (HHG) sources [13,14]. A few years later the method was first demonstrated with attosecond resolution [10–12]. A typical experimental configuration of this technique is shown in Figure 1.

What makes transient absorption an attractive approach in attosecond science? It is the first universal all-optical time-resolved spectroscopy technique with attosecond resolution. The previously introduced all-optical technique of probing attosecond dynamics with chirp (PACER, [15]) made use of the electron recollision process underlying

Pulsed He target

Delay line

Toroidal mirror

Figure 1. Example of an attosecond transient absorption set-up. The incoming NIR beam is split into two parts. One part generates the attosecond pulse, while the other part passes through an optical delay line. The delayed NIR pulse is recombined with the XUV attosecond pulse and focused into the experimental target (here, a pulsed helium target). The transmitted XUV photons are detected by an XUV spectrometer.

Generation

target

HHG. In PACER, the dynamics occurring during a recollision cycle is mapped onto the XUV spectral domain through the intrinsic chirp in HHG (i.e. in a classical picture, the photon energy-dependent release and recollision times of the electron participating in the harmonic emission). However, this limits the observation window to below one optical cycle of the driving infrared laser field. Furthermore, the method relies on strong-field ionisation of the target system. Attosecond transient absorption spectroscopy does not possess such limitations as it decouples the XUV generation from the time-resolved measurement process.

The all-optical detection brings with it a number of benefits. In contrast to the charged particles detected by the earlier attosecond techniques, photons do not influence each other. Count rates are thus not limited by the onset of space– charge effects. XUV photons can be detected directly with good quantum efficiencies of the order of 40% (for photon energies of tens of electronvolt) using back-illuminated charge-coupled device (CCD) sensors. With good cooling, such detectors have a very low noise floor and a large dynamic range. XUV spectrometers are a mature technology and provide good collection efficiency, high spectral resolution and even imaging capabilities without the significant trade-offs of charged-particle detectors.

Another important point is that as opposed to all other attosecond measurement techniques, transient absorption spectroscopy does not rely on photoionisation of the system being studied. As a consequence, the method can directly probe specific transitions, including the important bound– bound variety. Since it measures the time-dependent population of the energy levels of interest rather than the appearance of photofragments at a detector it yields a different and complementary perspective on attosecond dynamics as compared to the previously existing techniques.

3. Review of recent experiments

Since transient absorption spectroscopy is a rather recent addition to the attosecond tool set, only a limited number of experiments have been performed so far. In the following, we briefly review a representative selection of these. Most of the experiments so far concentrated on infrared induced strong-field effects in rare gas atoms. Rare gases are easy to handle and simple benchmark systems for these early experiments.

The first publication using attosecond transient absorption spectroscopy reported the time-resolved observation of valence electron motion in krypton ions [10]. The authors used isolated attosecond pulses with a centre photon energy of about 80 eV to probe the inner-atomic electron dynamics induced by ionisation through a strong sub-4-fs pulse centred at ~750 nm wavelength. This NIR pulse created an oscillating electron wavepacket by the coherent superposition of the $4p_{3/2}^{-1}$ and $4p_{1/2}^{-1}$ spin-orbit split levels in Kr⁺. This resulted in a periodic modulation of the absorbance at



XUV

spectrometer



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the probe energy, which corresponded to a resonant transition from the $4p^{-1}$ to the $3d^{-1}$ excited states of the Kr⁺ ion.

Shortly after that, Wang *et al.* [11] used transient absorption spectroscopy to resolve autoionisation dynamics in argon. This time an isolated attosecond pulse spanning a photon energy band between 20 and 40 eV was used as the probe and the autoionisation process was initiated and modified by few-cycle NIR pulses. This experiment represented the first time-resolved study of autoionisation. Not surprisingly, the measured lifetimes of the autoionising states corresponded to the values deduced from the linewidth obtained with static spectroscopy. However, this experiment clearly showed that such fast ionisation processes could be resolved in the time domain with a technique that is transferable to more complex systems where the dynamics might not be unambiguously derivable from static spectroscopic data.

The two first examples measured dynamics evolving on a few-femtosecond timescale. The first transient absorption experiment to reveal true subcycle dynamics was performed in helium exposed to a moderately strong infrared pulse and probed by a train of attosecond pulses [12]. This was also the first experiment to exploit the capability of the alloptical detection to monitor dynamics below the ionisation threshold. More on this experiment and some follow-up studies can be found in Section 5 below.

Two more recent experiments investigated the complex interaction of isolated attosecond pulses with laser-dressed helium below (and above) the ionisation threshold [16,17]. The attractiveness of helium as a target system lies in the fact that this simple atom is still accessible to exact quantum theoretical calculations. Chen et al. [16] report the manifestation of the so-called light-induced states in energetic regions that are off-resonant to the XUV excitation alone. The light-induced states are a consequence of the infrared-laser induced coupling to states that are dipoleforbidden for single-photon XUV transitions. That means that the XUV absorption spectrum will exhibit resonances that correspond to two-colour, two-photon transitions from the $1s^2$ ground state to 1sns and 1snd excited states. Due to the broad bandwidth of the involved isolated attosecond pulse, the resonance condition for such coupling can always be fulfilled irrespective of the exact XUV or NIR carrier frequencies. The population of the dipole-forbidden states can be controlled through the time delay between XUV and NIR pulses and through NIR intensity. The accurate theoretical modelling possible for this simple model experiment paves the way to also study strong-field coupling of excited states in more complex systems. Similar aspects have been investigated by Chini et al. [17]. In addition, they report three-photon coupling to excited states involving two NIR photons. The interference of direct (XUV alone) and indirect (two-colour multi-photon) contributions to the polarisation at a particular XUV frequency leads to the manifestation of half-NIR-cycle and quarter-NIR-cycle modulations of the absorbance as a function of relative XUV-NIR-pulse delay. This allows for optical control of XUV absorption and transmission on subcycle timescales.

With photon energies well above the ionisation threshold of helium both of its electrons can be excited and the resulting two-electron dynamics can be probed. Ott *et al.* [18] report the observation of two-electron wavepackets in helium with isolated-attosecond-pulse transient absorption spectroscopy. Above the first ionisation potential, the twoexcited-electron states interfere with the underlying continuum from the singly ionised helium and manifest themselves in the form of Fano resonances. The relative phase of the interfering pathways can be manipulated with a moderately strong NIR pulse, which enables control over the exact Fano line shape. A theoretical analysis revealed that the inversion of the Fano profile observed at a particular NIR intensity is expected to periodically repeat itself as a function of further increasing intensity [19].

While these previously discussed experiments studied exclusively fundamental light-matter interactions in simple rare gas atoms, a recent experiment demonstrated the application of attosecond transient absorption spectroscopy to a more complex system. Schultze et al. [20] use a few-cycle NIR pulse focused into a dielectric (fused silica) to manipulate its electronic structure in a reversible manner. The oscillating laser field periodically transfers and removes electron population to/from the conduction band of the dielectric, rendering it conductible for brief periods of time. These induced changes exhibit themselves in corresponding modulations of the absorbance at the 109 eV photon energy of the probing isolated attosecond pulse. It is worthwhile to mention that in contrast to the older ionisationbased attosecond electron streaking technique [21] which is only surface sensitive, attosecond transient absorption spectroscopy allows probing bulk dynamics in solid-state samples.

Attosecond transient absorption in molecular systems is yet to be demonstrated. However, its extension to molecular targets in gas phase or even in thin liquid jets should not pose any fundamental difficulties.

4. Overview of theoretical work

Until recently, the optical response of matter in strong laser fields and/or on attosecond timescales has received little attention from theoreticians. The reason for this lies in the fact that attosecond and strong-field science historically concentrated on analysing the ionisation fragments created in a particular interaction rather than its optical response. Only the recent advent of attosecond transient absorption spectroscopy as a highly promising new experimental technique was able to spark the interest of theoreticians in this aspect of attosecond dynamics. In this section we will review the latest developments in theory related to attosecond transient absorption spectroscopy. The present available work concentrates on dynamics in rare gases and most prominently on helium, which is understandable given the difficulty in accurately modelling more complex systems.

The gas density in transient absorption experiments is usually chosen high enough to introduce a significant optical density at the XUV probe photon energy. As a result reshaping and propagation effects for the attosecond pulses may not be negligible anymore. This requires the treatment of these phenomena in theories for a wide range of physical systems and can give rise to new effects that do not occur on a single-atom level. Gaarde et al. [22] discuss absorption and reshaping in helium exposed to a moderately strong infrared laser field. The dressing field considerably modifies the optical properties at the XUV photon energies by modulating the atomic energy levels and possibly coupling different atomic resonances either directly or through twocolour processes involving the XUV. For example, a dressing field at 1600 nm wavelength may induce transparency in the $1s \rightarrow 2p$ XUV transition through the mechanism of Autler-Townes splitting (sometimes also referred to as electromagnetically induced transparency) [22]. Any pulse in resonance with this transition will undergo dramatic reshaping depending on the exact parameters of the infrared dressing field. In a similar system, the dynamical control of XUV absorption and emission through the laser-induced coupling of the 2p and 2s was treated theoretically [23]. Laser-induced transparency at XUV photon energies in helium was also found for 800 nm dressing of the $2s2p(^{1}P^{o})$, $2p^2({}^1S^e)$ and $2s^2({}^1S)$ autoionising states [24,25].

Chen *et al.* [26] treated the special case of a train of attosecond pulses interacting with laser-dressed helium. The calculations show periodic modulations of the absorption probability at the different harmonics forming the pulse train and as a function of XUV–NIR delay. The theoretical results are in agreement with an experiment that interpreted these modulations in terms of the interference of transiently bound electron wavepackets [12]. While the observed periodicity of the modulation corresponds to twice the NIR carrier frequency, the theory also predicts twice as fast modulations for a specific choice of parameters. However, Chen *et al.* show that these faster contributions can be lost due to propagation effects [26]. This underlines further the importance of macroscopic effects in attosecond transient absorption spectroscopy.

Motivated by the experiments of Goulielmakis *et al.* [10], recent theoretical work discussed attosecond transient absorption spectroscopy of Krypton [27]. This study concentrates on the aspect of (infrared) laser-generated ions that are typically found in a superposition of many ionic states. The direction of the laser polarisation favours the excitation of certain excited states over others. This spatial selectivity is called 'alignment' of the Kr⁺ ions. The experimentally observed degree of alignment is lower than predicted by existing theories and pulse propagation effects

cannot explain this discrepancy [27]. More work is needed for a better understanding of these observations.

Krypton is also the system being investigated by Baggesen *et al.* [28]. This work, however, specifically models a true XUV pump – XUV probe experiment with attosecond resolution. This experiment would monitor inner-shell hole creation and its decay through Auger electron emission. The realisation of such measurements is out of reach of present experimental capabilities due to the low cross sections for XUV two-photon non-linear processes.

5. Electron wavepacket interference

In the following we discuss the first attosecond transient absorption experiment that used attosecond pulse trains rather than isolated attosecond pulses to probe the intra-atomic dynamics [12]. This was also the first transient absorption experiment to report subcycle (with respect to the NIR pulse) dynamics and to investigate processes below the ionisation threshold. We highlight the main experimental findings and show recent extensions of the experiment aiming at the verification of the underlying theoretical model.

Attosecond pulse trains (APTs) have proven to be particularly useful for initiating and probing the interference of electron wavepackets (EWPs) [29-31]. The interference allows us to directly access phase information of the system and/or the process being studied. At the same time, the interference enables control over the dynamics by modifying parameters that influence the phase of the interfering wavepackets. It was shown by Johnsson et al. that an APT located below and around the ionisation threshold of helium in the presence of an NIR field could periodically modulate the ionisation probability of the helium atoms as a function of time delay between the NIR pulses and the APT [30]. In this experiment, the NIR intensity was chosen of the order of 10^{13} W/cm² such that it was low enough not to induce strong-field processes in ground-state helium. The periodicity of the ion yield modulations with delay was found to be twice the frequency of the NIR-driving laser. This observation was explained by the interference of transiently bound EWPs. The underlying mechanism is schematically depicted in Figure 2a). The oscillating IR field deforms the atomic potential. This modifies the absorption probability for the APT photons as a function of relative delay between the two fields. As a result, the modulation of ionisation yield observed by Johnsson et al. should also manifest itself in the delay-dependent absorbance at the APT photon energies. This idea forms the basis of our transient absorption experiment.

The APT is generated by HHG in xenon with \sim 30-fs pulses from a Ti:sapphire laser system. The remaining NIR radiation is removed after the HHG source by a thin aluminium filter. Before the HHG target, a small fraction of the original NIR laser beam is split off and time-delayed with respect to the APT. This NIR and the APT beam are



Figure 2. (a) Schematic picture underlying the transiently bound electron wavepacket interference. A pulse in the APT (dark blue) excites an EWP (green). At the same time the NIR laser deforms the atomic potential (solid and dashed black lines). With proper timing, the EWP may escape into the continuum where it still gets accelerated by the field of the NIR laser. The EWP returns to the parent ion at the instant at which the next pulse in the APT excites the next EWP. (b) Energetic configuration of the experiment. The spectrum forming the APT consists of two harmonics below (H13 and H15) and two above (H17 and H19) the ionisation threshold of helium (IP). Some energy levels accessible through single-photon excitation from the ground state are also shown.

recombined collinearly and focused into the helium target (particle density $\approx 5 \times 10^{17} \text{ cm}^{-3}$). As in the case of Johnsson *et al.*, we choose our NIR intensity of the order of 10^{13} W/cm². We collect the transmitted harmonic photons after the helium target and detect them with a CCDequipped XUV spectrometer. The experimental set-up corresponds to the one schematically depicted in Figure 1. Our APT consists mainly of harmonic orders 13 and 15 located below the field-free ionisation potential (IP) of helium and harmonic 17, which is above the IP. The energetic configuration of our experiment is shown in Figure 2b). It follows that in the absence of the NIR field harmonics 13 and 15 are not absorbed unless they overlap with the internal resonances of helium.

In a time domain picture (Figure 2a), each pulse in the APT excites an EWP. The EWPs are only transiently bound because the potential barrier may be sufficiently lowered for proper delay between the NIR and the APT such that the wavepackets can escape into the continuum. The EWPs feel the NIR field even after their escape and get accelerated. The oscillating NIR field will direct the EWPs (or part of them) back into the vicinity of the ion. Since the same NIR field generated the APT, it has the same periodicity. A returning EWP therefore arrives at its parent ion at the instant when a subsequent attosecond pulse excites the next wavepacket. The interference of the wavepackets excited by subsequent attosecond pulses significantly alters the ionisation probabilities with delay. Johnsson *et al.* found that this interference effect dominates the modulation of the absorption through the deformation of the atomic potential by an order of magnitude [30].

The experimental transient absorption data is shown in Figure 3. One immediately sees that transient absorption spectroscopy yields more information than the traditional methods based on photo-electron or photo-ion detection. Attosecond transient absorption not only provides the timeresolved ionisation probability, but also measures the spectrally resolved absorption probabilities. When comparing the transmitted photon yield modulations at the different harmonics forming the APT, one observes that they are not



Figure 3. (a) Spectra of the harmonics forming the APT after transmission through the helium target and as a function of NIR–APT delay. The rapid modulations are clearly visible for harmonics 15 and 17, but less so for harmonics 13 and 19 on this scale. (b) Line cuts off the signal at harmonic 13 for different experimental parameters. The thickness d_{AI} of the aluminium filter used to remove the remaining NIR light after APT generation and the NIR intensity I_{IR} in the helium target was varied. The aluminium filter thickness modifies the spectral phase of the APT, whereas the NIR intensity changes the phase accumulated by the recolliding EWP. It can be seen that the phase of the modulations is insensitive to the spectral phase of the APT but rather sensitive to NIR intensity.

necessarily in phase with respect to each other. Our experiments have shown that they react very sensitively to a change of the NIR intensity but that they are insensitive to the spectral phase of the APT. This observation qualitatively confirms the simple model depicted in Figure 2a). A change in the NIR intensity modifies the phase accumulated by a transiently bound EWP during its excursion into the continuum [32,33]. When the returning EWP interferes with the EWP just excited by the subsequent pulse in the APT, this contribution governs the relative phase. It thus decides between constructive or destructive interference. The spectral phase of the APT is imprinted on all excited wavepackets in an identical way and therefore does not have an influence on the interference condition. The NIR intensity and the relative NIR-APT delay can therefore be regarded as knobs controlling the spectrally dependent absorption probability.

Based on the qualitative picture in Figure 2a, one would predict that replacing the APT with a single attosecond pulse would lead to a significant reduction of the absorbance modulation contrast. Some residual modulations with delay are still expected because of the periodic deformation of the atomic potential by the NIR field. The theoretical model presented by Johnsson *et al.* also predicts a sizable reduction in modulation strength when reducing the number of pulses in the APT to ultimately one [30]. Similarly, changing the polarisation of the NIR field from linear to elliptical should prevent the recollision of the outgoing EWP with its parent ion for high enough ellipticity. In HHG, recollisions are already well suppressed for ellipticities $\varepsilon > 0.13$ [34]. Without these recollisions, EWP interference should not occur anymore.

We tested both of these predictions experimentally. Figure 4 shows the corresponding raw data. As can be seen, modulations along the delay coordinate are still present rather prominently. When changing the polarisation state of the IR field from linear to an ellipticity $\varepsilon \approx 0.66$, the modulation contrast still amounts to more than 60% of its initial value. The dependence of the modulation contrast on ellipticity is much weaker than anticipated from the qualitative picture involving a recollision process. Furthermore, pronounced modulations are present even when only a single attosecond pulse participates in the interaction. The isolated attosecond pulse in our experiment was generated with polarisation gating [35,36] and measured to have a duration of \sim 290 as. Its spectrum peaked at 35 eV with a full-width at half-maximum of $\sim 9 \text{ eV}$ and a long spectral tail extending down to the 15 eV cut-on of the aluminium filter that was used to reject the remaining driving laser light. Note that the data in Figure 4b reproduces the structures recently reported in experimental and theoretical work by Chen et al. and Chini et al. [16,17]. While the observations made in [12] and Figure 3 support the wavepacket interference model, this newer work suggests that this picture is at least not complete. For the case of excitation with isolated attosecond pulses, the multi-pathway interference of multi-photon



Figure 4. Transmitted XUV spectra as a function of XUV–NIR delay. (a) APT with elliptically polarised NIR. The modulations on the three harmonics (H13, H15, H17) are still clearly visible despite the high NIR ellipticity of $\varepsilon \approx 0.66$. (b) Single attosecond pulse with linearly polarised NIR. Also in this case strong modulations of the transmitted signal can be seen in various spectral regions.

excitation of the so-called light-induced states is found to be the origin of the observed modulations [16,17]. The inconsistency of some of our own experimental observations discussed above with the wavepacket interference picture indicates that the light-induced-state mechanism may also explain at least part of the time-dependent structure seen in the case of excitation with an APT. This discussion underlines the fact that a better understanding of the optical response of strong-field interactions is needed. The transfer of models devised for the interpretation of ionisation physics to the description of the optical properties is not straightforward.

6. Outlook and conclusion

Attosecond transient absorption spectroscopy is a rather young addition to the attosecond toolset. It has already been applied to time-resolve a variety of ultrafast processes in systems as simple as rare gas atoms or as complex as solidstate materials. While there are presently no attosecond transient absorption experiments on molecules, it should be straightforward to extend the technique to fill this complexity gap. At suitable photon energies, the method may also be applied to thin liquid jets to probe molecules in solution. Rather than monitoring the optical response of a system only in transmission, transient reflection geometries may be favourable under certain circumstances. We have provided a review of the present state-of-theart in attosecond transient absorption spectroscopy with respect to experiment as well as theory. Our more detailed discussion of transiently bound electron dynamics in helium has shown how the method resolves laser induced intraatomic processes with sub-NIR-cycle resolution. However, this example has also shown that further work is needed to improve our understanding of optical interactions on such short timescales and with target systems being exposed to strong laser field strengths. While attosecond science has focused on ionisation and the detection of the resulting fragments, the advent of the all-optical transient absorption method will broaden the scope of the field, now also enabling access to previously hidden dynamics such as bound–bound processes.

The all-optical nature of transient absorption spectroscopy may allow the implementation of detection schemes that have been very successful in conventional femtosecond spectroscopy in the visible and infrared spectral region. With the ongoing rapid progress of high-power, high-repetition-rate laser sources [37–39], lock-in detection should eventually become possible and increase the sensitivity of attosecond spectroscopy by orders of magnitude [40]. These developments should ultimately make attosecond tools accessible to more research groups and scientific fields. This is a prerequisite for attosecond science to have similar impact on our understanding of ultrafast phenomena in nature as femtosecond science had in previous decades.

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References

- M. Schultze, M. Fiess, N. Karpowicz, J. Gagnon, M. Korbman, M. Hofstetter, S. Neppl, A.L. Cavalieri, Y. Komninos, T. Mercouris, C.A. Nicolaides, R. Pazourek, S. Nagele, J. Feist, J. Burgdorfer, A.M. Azzeer, R. Ernstorfer, R. Kienberger, U. Kleineberg, E. Goulielmakis, F. Krausz, and V.S. Yakovlev, Science **328**, 1658 (2010).
- [2] K. Klünder, J.M. Dahlström, M. Gisselbrecht, T. Fordell, M. Swoboda, D. Guenot, P. Johnsson, J. Caillat, J. Mauritsson, A. Maquet, R. Taïeb, and A. L'Huillier, Phys. Rev. Lett. 106, 143002 (2011).
- [3] P. Eckle, A.N. Pfeiffer, C. Cirelli, A. Staudte, R. Dörner, H.G. Muller, M. Büttiker, and U. Keller, Science 322, 1525 (2008).
- [4] A.N. Pfeiffer, C. Cirelli, M. Smolarski, D. Dimitrovski, M. Abu-samha, L.B. Madsen, and U. Keller, Nature Phys. 8, 76 (2012).
- [5] A. Landsman, M. Weger, J. Maurer, C. Cirelli, A. Ludwig, L. Gallmann, and U. Keller, preprint, arXiv:1301.2766 (2013).
- [6] L. Gallmann, C. Cirelli, and U. Keller, Annu. Rev. Phys. Chem. 63, 447 (2012).

- [7] M. Hentschel, R. Kienberger, C. Spielmann, G.A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, Nature 414, 509 (2001).
- [8] P.M. Paul, E.S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H.G. Muller, and P. Agostini, Science 292, 1689 (2001).
- [9] H.G. Muller, Appl. Phys. B 74, S17 (2002).
- [10] E. Goulielmakis, Z.-H. Loh, A. Wirth, R. Santra, N. Rohringer, V.S. Yakovlev, S. Zherebtsov, T. Pfeifer, A.M. Azzeer, M.F. Kling, S.R. Leone, and F. Krausz, Nature 466, 739 (2010).
- [11] H. Wang, M. Chini, S. Chen, C.-H. Zhang, F. He, Y. Cheng, Y. Wu, U. Thumm, and Z. Chang, Phys. Rev. Lett. 105, 143002 (2010).
- [12] M. Holler, F. Schapper, L. Gallmann, and U. Keller, Phys. Rev. Lett. 106, 123601 (2011).
- [13] Z.-H. Loh, M. Khalil, R.E. Correa, R. Santra, C. Buth, and S.R. Leone, Phys. Rev. Lett. 98, 143601 (2007).
- [14] Z.-H. Loh, C.H. Greene, and S.R. Leone, Chem. Phys. 350, 7 (2008).
- [15] S. Baker, J.S. Robinson, C.A. Haworth, H. Teng, R.A. Smith, C.C. Chirilă, M. Lein, J.W.G. Tisch, and J.P. Marangos, Science **312**, 424 (2006).
- [16] S. Chen, M.J. Bell, A.R. Beck, H. Mashiko, M. Wu, A.N. Peiffer, M.B. Gaarde, D.M. Neumark, S.R. Leone, and K.J. Schafer, Phys. Rev. A 86, 063408 (2012).
- [17] M. Chini, X. Wang, Y. Cheng, Y. Wu, D. Zhao, D.A. Telnov, S.-I. Chu, and Z. Chang, Sci. Rep. 3, 1105 (2013).
- [18] C. Ott, A. Kaldun, P. Raith, K. Meyer, M. Laux, Y. Zhang, S. Hagstotz, T. Ding, R. Heck, and T. Pfeifer, preprint, arXiv:1205.0519v1 [physics.atom-ph] (2012).
- [19] L. Argenti, C. Ott, T. Pfeifer, and F. Martin, preprint, arXiv:1211.2566v1 [physics.atom-ph] (2012).
- [20] M. Schultze, E.M. Bothschafter, A. Sommer, S. Holzner, W. Schweinberger, M. Fiess, M. Hofstetter, R. Kienberger, V. Apalkov, V.S. Yakovlev, M.I. Stockman, and F. Krausz, Nature (2012). doi:10.1038/nature11720
- [21] A.L. Cavalieri, N. Müller, T. Uphues, V.S. Yakovlev, A. Baltuska, B. Horvath, B. Schmidt, L. Blümel, R. Holzwarth, S. Hendel, M. Drescher, U. Kleineberg, P.M. Echenique, R. Kienberger, F. Krausz, and U. Heinzmann, Nature 449, 1029 (2007).
- [22] M.B. Gaarde, C. Buth, J.L. Tate, and K.J. Schafer, Phys. Rev. A 83, 013419 (2011).
- [23] M. Wu, S. Chen, K.J. Schafer, and M.B. Gaarde, Phys. Rev. A 87, 013828 (2013).
- [24] M. Tarana and C.H. Greene, Phys. Rev. A 85, 013411 (2012).
- [25] W.-C. Chu and C.D. Lin, Phys. Rev. A 85, 013409 (2012).
- [26] S. Chen, K.J. Schafer, and M.B. Gaarde, Opt. Lett. 37, 2211 (2012).
- [27] R. Santra, V.S. Yakovlev, T. Pfeifer, and Z.-H. Loh, Phys. Rev. A 83, 033405 (2011).
- [28] J.C. Baggesen, E. Lindroth, and L.B. Madsen, Phys. Rev. A 85, 013415 (2012).
- [29] T. Remetter, P. Johnsson, J. Mauritsson, K. Varju, Y. Ni, F. Lépine, E. Gustafsson, M. Kling, J. Khan, R. Lopez-Martens, K.J. Schafer, M.J.J. Vrakking, and A. L'Huillier, Nat. Phys. 2, 323 (2006).
- [30] P. Johnsson, J. Mauritsson, T. Remetter, A. L'Huillier, and K.J. Schafer, Phys. Rev. Lett. 99, 233001 (2007).
- [31] J. Mauritsson, P. Johnsson, E. Mansten, M. Swoboda, T. Ruchon, A. L'Huillier, and K.J. Schafer, Phys. Rev. Lett. 100, 073003 (2008).
- [32] M. Lewenstein, P. Salières, and A. L'Huillier, Phys. Rev. A 52, 4747 (1995).

- [33] P. Salières, B. Carré, L.L. Déroff, F. Grasborn, G.G. Paulus, H. Walther, R. Kopold, W. Becker, D.B. Milosevic, A. Sanpera, and M. Lewenstein, Science **292** (5), 902 (2001).
- [34] P. Antoine, A. L'Huillier, M. Lewenstein, P. Salières, and B. Carré, Phys. Rev. A 53, 1725 (1996).
- [35] I.J. Sola, E. Mével, L. Elouga, E. Constant, V. Strelkov, L. Poletto, P. Villoresi, E. Benedetti, J.-P. Caumes, S. Stagira, C. Vozzi, G. Sansone, and M. Nisoli, Nat. Phys. 2, 319 (2006).
- [36] G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Velotta,

S. Stagira, S. De Silvestri, and M. Nisoli, Science **314**, 443 (2006).

- [37] T. Südmeyer, S.V. Marchese, S. Hashimoto, C.R.E. Baer, G. Gingras, B. Witzel, and U. Keller, Nat. Photonics 2, 599 (2008).
- [38] C.J. Saraceno, F. Emaury, O.H. Heckl, C.R.E. Baer, M. Hoffmann, C. Schriber, M. Golling, T. Suedmeyer, and U. Keller, Opt. Express 20, 23535 (2012).
- [39] C. Heese, C.R. Phillips, B.W. Mayer, L. Gallmann, M.M. Fejer, and U. Keller, Opt. Express 20, 26888 (2012).
- [40] U. Keller, IEEE Photon. J. 2, 225 (2010).