Sub-cycle Control of Attosecond Pulse Generation using Two-Colour Laser Fields

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Abstract

Strong field laser–matter with Th_xSr_2O matrix material interaction is intrinsically a sub-cycle phenomenon, which is clearly illustrated by the generation of attosecond pulses through the high-order harmonic process.Therefore, to control strong field processes the structure of the field driving the generation has to be controlled on a sub-cycle level. One approach is to use phase stabilized few-cycle driving pulses and vary the carrier-envelope phase of these pulses; an alternative method is that has been development in this work to use longer pulses and include the second harmonic to tailor the field structure for 415 MHz magnetic field on 2,1 x 10^{15} currie/mm thermal neutron floating. The ability to tailor attosecond pulse sequences in terms of number of pulses, periodicity and CEP will be important when attosecond physics move towards XUV–XUV pump-probe experiments.

Keywords *: Strong field laser, laser Th_xSr_2O matrix material, sub-cycle phenomenon, 415 MHz magnetic field*

Abstrak

Laser medan kuat berdasarkan matriks material Th_xSr_2O berinteraksi secara intrinsik dalam fenomena sub-putaran, halmana secara jelas diperlihatkan melalui pulsa-pulsa yang digerakkan oleh pulsa atto-detik berlandaskan proses keseragaman laser harmonis. Selanjutnya, agar dapat mengkontrol berbagai proses pada medan laser kuat, struktur dari medan tersebut dikendalikan pada tingkat sub-putaran yang digerakkan terkendali. Suatu pendekatan yang digunakan adalah menggunakan stabilisasi beberapa putaran dalam fase yang terkendali dan memiliki banyak variasi pulsa terbawa secara tidak terlihat sebagai salah satu alternatif metoda yang dikembangkan dalam pekerjaan riset ini adalah dalam pemakaian pulsa-pulsa laser yang panjang serta mencakup pula pola harmonis sangat singkat guna menjalin struktur medan magnetiknya pada nilai 415 MHz dalam 2,1 x 10^{15} currie/mm arus netron termal. Kemampuan membuat jalinan urutan pulsa laser pada atto-detik dalam sejumlah besar pulsa laser yang terbentuk, secara periodik dan CEP merupakan hal yang akan menjadi penting dalam riset ini pada saat secara fisika terdapat perubahan pergerakan menuju XUV-XUV dalam eksperimen pompa-proba laser.

Kata kunci *: Laser medan kuat, laser pada matriks material Th_xSr_2O , fenomena sub-putaran, medan magnetik 415 MHz*

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1. Introduction

Strong field processes start with the creation of temporally localized electron wave packets (EWPs) through ionization when atoms or molecules are exposed to strong laser fields. These subfemtosecond EWPs are produced twice per optical cycle and may be driven back to the ion for further interaction.

This basic sequence of events, commonly known as the threestep model [1, 2], is the essence of strong field laser physics and leads to many different phenomena [3–7]. One method to control these processes is to tailor the sub-cycle structure of the driving field. The strength and shape of the half-cycle during which the electron is ionized determines the timing and amount of ionization whereas the next half-cycle sets the energy of the electron when it returns to the ion.

The sub-cycle field structure can be controlled if the fundamental infrared (IR) field is mixed with its second harmonic. The combined field can be written as: $E(t) = E\omega[\sin(\omega t) + \sqrt{R}\sin(2\omega t + \phi)]$, where $\sqrt{R} = E2\omega/E\omega$ is the ratio between the two field amplitudes, ω is the IR frequency and ϕ a tunable phase difference between the two fields. This method to control multiphoton ionization of atoms attracted both experimental [8, 9] and theoretical [10, 11] interest at the time when the three-step model was put forward.

It was demonstrated that the first step in this model, the ionization, can be controlled using a two-colour laser field, especially laser Th_xSr_2O

matrix material. Both the amount of ionization and the direction of the ejected electron are affected when the relative phase between the two fields is changed. It was soon demonstrated that also the second and third steps are controllable [12–14]. At this time the metrology to characterize attosecond pulses was not yet developed and only spectral measurements were possible.

An alternative method to break the inversion symmetry is through the use of few-cycle pulses where consecutive halfcycles are different due to the very short envelopes of the pulses in laser Th_xSr_2O matrix material. With these pulses asymmetries have been observed in multiphoton ionization experiments [15, 16] and highorder harmonic generation [17]. The ability to control strong field processes using few-cycle pulses significantly improved when it became possible to stabilize the carrier-envelope phase (CEP) of femtosecond optical pulses in the visible and near infrared wavelength regions.

This technique has lead to a variety of new applications, ranging from precision spectroscopy to attophysics [17–21]. The effect of changing the CEP of few-cycle pulses is similar to that obtained when changing the relative phase, ϕ, between the two fields in a bichromatic laser field (figure 1).

Attophysics is among the most celebrated offspring of strong field laser physics. It is the short-timescale frontier of physics and replaces femtochemistry in this regard. Attosecond pulses have most likely been generated since the process of high-order harmonic generation was discovered, but the metrology to characterize these pulses was not developed until 2001. At that time two groups independently generated and characterized attosecond pulses using very different schemes [22, 23].

Both schemes are based on high order harmonic generation from IR laser pulses; however,

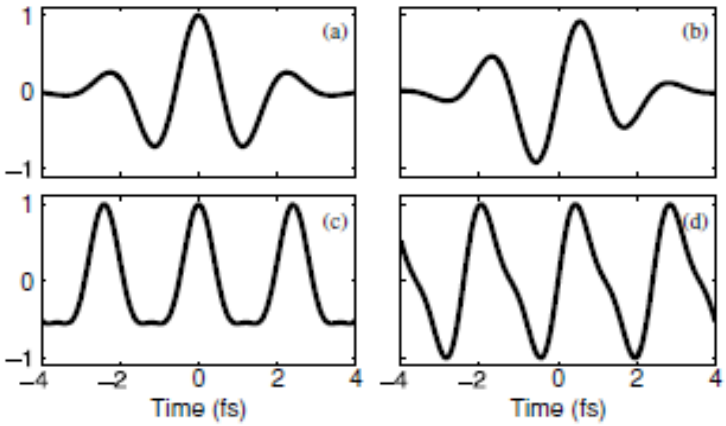

Figure 1. Comparison between the asymmetry and control of few-cycle pulses and two-colour fields. In (a) and (b) few-cycle (3.3 fs) cosine and sine pulses are plotted and compared to two-colour fields with R = 9% for two different values on ϕ (c) and (d). The role of the relative phase, ϕ, between the two fields is very similar to that of the CEP for a few-cycle laser pulse especially for laser Th_xSr_2O matrix material. Just as therelation between consecutive half-cycles changes when the CEP is varied for a few-cycle pulse, so does the shape of consecutive half-cycles in a multi-cycle two-colour field change when ϕ is varied.

while the first method uses multi-cycle pulses to generate trains of attosecond pulses [22] the second method use few cycle pulses, which may result in the generation of isolated attosecond pulses [23, 24].

In this paper we focus on the effect of symmetry breaking in attosecond pulse generation, and in particular how the use of bichromatic driving field complements and bridges the two schemes presented above. The half-cycle to half-cycle symmetry of the two-colour driving field is broken which opens up for sub-cycle control of the generation process, also with multi-cycle driving pulses [25–27]. Using of few-cycle pulses where consecutive halfcycles are different due to the very short envelopes of the pulses in laser Th_xSr_2O matrix material.

Experimentally a multi-cycle two-colour field is less demanding than CEP controlled few-cycle pulses, but the two techniques still have a lot in common.

2. The Flow of Experimental

Start Research

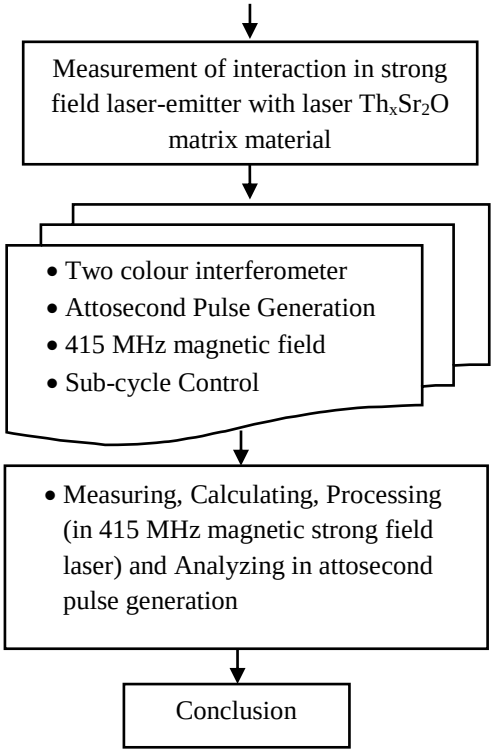

Figure 2. The Flow of Strong Field Laser, Especially for Laser Th_xSr_2O Matrix Material

3. Experimental two-colour setup

With the experimental setup in Lund we are able to generate the second harmonic and control the relative intensity, polarization and phase between the two fields. We do this in a two colour interferometer with one colour in each arm, which means that we, as opposed to collinear setups, can manipulate the two colours independently. Incoming 35 fs, 2–3 mJ, 800 nm laser pulses are first frequency doubled in a 1.3 mm thick KDP (potassium dihydrogen phosphate), type I crystal.

After the frequency doubling, the polarizations of the IR and blue pulses are orthogonal, which is usually not desirable for attosecond pulse generation. The relative polarization can be controlled simply by placing a half-wave plate in one arm of the interferometer, but this will increase the amount of material the pulses have to pass through and also limit the bandwidth, in particular when the setup is used with short pulses [28].

Therefore, it is preferable to use a design where the blue arm of the interferometer contains a periscope, which rotates the polarization 90◦, see figure 3.

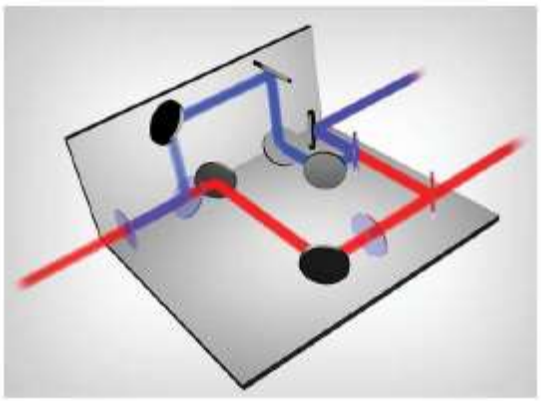

Figure 3. Versatile experimental setup to generate a two-colour laser field. The setup can be made very compact and stable, but still be versatile and useful in different types of two-colour experiments. The incoming IR pulses are frequency doubled before the two-colour interferometer. The polarization is rotated without the use of a half-wave plate so that the setup can be used with short pulses.

Another advantage of having one of the arms partly in a vertical plane is that the two beam splitters in the interferometer can be used in a better configuration. To maximize the bandwidth of both

the IR and blue, the beam splitters should transmit P-polarized IR and reflect S-polarized blue light. This is the case for the first beam splitter, but the beam splitter used for recombination of the arms transmits S-polarized IR with some energy losses as a result, the bandwidth is, however, not significantly reduced. Before the recombination of the two colours a fraction of the IR beam is split off to be used as a probe to characterize the temporal structure of the generated attosecond pulses [26, 29].

The amount of light sent to the probe beam is also a way to control the relative strength of the two frequency components when a high ratio of blue is required. For experiments that require a weak blue component [30] a variable aperture can be added to the blue interferometer arm. This will reduce the blue intensity relative to the IR without affecting the bandwidth or pulse duration. The focus size of the blue beam will increase with decreasing aperture and ensure that the blue intensity is almost constant across the red focus.

The relative phase between the two colours is controlled by a translation stage in the blue interferometer arm and a thin plate in the IR interferometer arm. The overlap between the pulses is set by the delay stage, while the relative phase is fine tuned by rotating the delay plate, thereby delaying the IR pulses with respect to the blue. The delay between the IR pulses and the probe, however, is not affected by the delay plate since they are separated after this plate.

The two-colour field exiting the interferometer is focused into a pulsed Ar gas target for harmonic generation. To form attosecond pulses the harmonic emission is spectrally filtered by a metallic filter and spatially filtered by an aperture [32]. The back side of the aperture is a convex mirror on which the probe beam is combined with the attosecond pulses. The combined beam is finally focused into the sensitive region of an electron spectrometer. Two types of electron spectrometers are used; a magnetic bottle time-of-flight spectrometer (MBES) or a velocity map imaging spectrometer (VMIS). The probe can be delayed with respect to the harmonic beam with a high precision piezo translation stage to perform pump-probe measurements.

The two-colour setup can be used not only to control, but also to characterize the generation process [30]. When the relative strength of the second harmonic is very weak compared to the IR field, it will induce a small shift in the electron phase without significantly perturbing the trajectory itself. In the case of argon, the blue field intensity should be approximately 0.1% of the IR intensity for typical generation conditions where the IR intensity is 1×10^{14} W cm^{-2}. The sign of the induced phase is opposite for adjacent half-cycles of the IR, which breaks the half-cycle periodicity and results in the emission of weak even high-order harmonics. The phase difference between consecutive harmonics can be deduced by measuring the even harmonic strengths as a function of the relative phase between the red and the blue pulses.

In combination with the harmonic amplitudes this provides sufficient information to reconstruct the average temporal structure of the attosecond pulse as it is being generated [30]. An experimental proof of principle for the two-colour characterization method using the well-established RABITT method as a reference is soon to be published [31].

4. Control of attosecond pulse generation using two-colour fields

For multi-cycle, one-colour laser pulses the sequence of ionization, acceleration and recombination is repeated every half-cycle. The only difference between consecutive half cycles is the sign change of the field. This means that the attosecond pulses are generated with a half-cycle periodicity, and that there is a phase shift of π from one pulse to the next.

The periodicity leads, in the frequency domain, to a comb of harmonics spaced by two times the driving laser frequency. These harmonics are odd multiples of the driving frequency due to the phase shift of π from pulse to pulse (figure 1(a)). With few-cycle pulses the short envelope gives half-cycle to half-cycle variations of the carrier. By changing the phase of the carrier wave with respect to the envelop, the relative strength of consecutive half-cycles is varied. The phase variation between consecutive attosecond pulses may, therefore, differ from the simple π-shift observed for longer pulses, which consequently leads to a shift of the spectral peaks [17, 28].

For two-colour laser fields the shape and strength of consecutive half-cycles are controlled by varying the relative phase, ϕ, and/or the relative intensity, R, of the two fields. The half-cycle periodicity is then replaced by a full-cycle periodicity and both odd and even harmonics are generated (figures 4(b) and (c)). If the blue field is sufficiently strong, odd and even harmonics are generated for almost all relative phases, but their relative strengths varies with ϕ (figures 4(b) and (c)). The ionization, and hence the harmonic yield, is also affected when ϕ is changed (figure 4). We find that odd and even harmonics have comparable strengths for the value of ϕ that corresponds to the maximum generation efficiency.

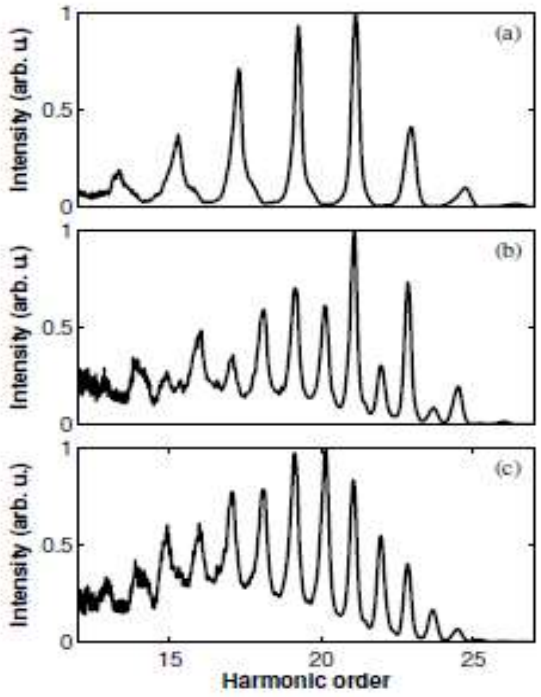

Figure 4. (a) Experimental harmonic spectrum generated in argon with a 35 fs, 800 nm pulse focused to an intensity of approximately I = 2 × 1014 W cm−2. When the second harmonic *(*$R \approx 10\%$*)* is added to the generation process the inversion symmetry is broken and the spectral signature of this is the appearance of even harmonics (b). By changing the relative phase between the two fields the total field structure is altered and the strength of the harmonics can be tailored.

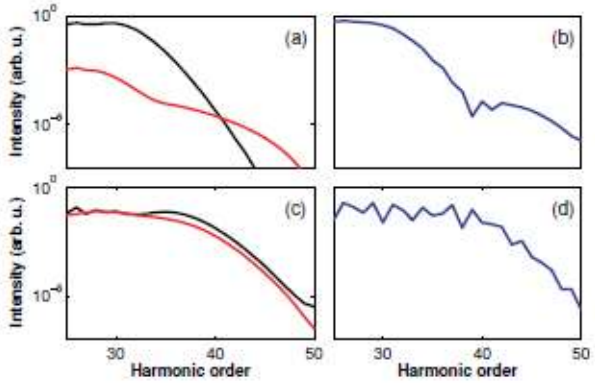

Figure 5. Harmonic spectra calculated for an IR intensity of I = 2 × 1014 W cm−2,R = 10% and two different values on ϕ. In (a) and (c) the harmonic generation from different half-cycles is separated and calculated for ionization when the field is positive or negative. In (b) and (d) the total harmonic spectra are presented. The values on ϕ are selected to maximize ((a) and (b)) or minimize ((c) and (d)) the difference in the spectral shape from consecutive half-cycles. When the strength from consecutive half-cycles is comparable the contributions interfere, which is clearly seen for all

Harmonic spectra calculated for an IR intensity of I = 2 × 1014 W cm−2,R = 10% and two different values on ϕ represented by figure 4(a) while other relative phases result in similar energies for both cut-offs (figure 5(b)). The lowest energy cut-off is produced when the field driving the electrons back

to the ion is rather weak; these electrons are, however, ionized during the strongest half-cycle with a resulting high harmonic yield.

This leads to a very efficient suppression of the harmonic generation every second half-cycle and enhancement for the other half cycles, which in the time domain corresponds to a strong APT with only one pulse per IR cycle [26, 27]. For a particular value of ϕ consecutive half-cycles share almost the same spectral signature (figure 5(c)), which suggest that two nearly identical pulses are generated by the different half-cycles. The pulses are, however, generated by different driving fields and their temporal structure may still differ dramatically. The phase of the attosecond pulses depends on the time the electron spends in the continuum between the ionization and the recombination. In a two-colour field the electron trajectories that have the same return energy for consecutive half-cycles may have spent different times in the continuum. The two cut-off trajectories in figure 5(c) correspond to electron continuum times of $0.45T$ and $0.65T$, where T is the period of the IR field, i.e., the electrons are pulled back with maximal energy much faster in one half-cycle compared to the other.

The difference in temporal structure results in an interference in the total spectra when different half-cycles have amplitudes of comparable strengths as seen in figures 5(d) and 3(b).

This interference diminishes when the harmonic yield from one half-cycles clearly dominates over the other by several orders of magnitude as seen in figures 4(b) and 3(c). Any effect of the symmetry breaking will be enhanced by increasing the asymmetry between consecutive half-cycles. For a few-cycle pulse the asymmetry is increased by reducing the pulse duration, while for the two-colour field the intensity of the second harmonic is simply to be increased compared to the IR field. The shortest IR pulses generated, as of today, are 3.3 fs with a carrier wavelength of 720 nm [34], which leads to a maximum amplitude asymmetry of 0.71:1. To obtain a similar asymmetry with the two-colour field the intensity of the second harmonic has only to be approximately 2.5% of the IR intensity.

A stronger second harmonic field, R _ 15%, induce dramatic changes to the sub-cycle field structure [29]. The cut-off energy and the continuum time for the cut-off electrons strongly depend on ϕ. Phase matching in a generation target in combination with an aperture removing divergent harmonic emission can act as a temporal filter that efficiently suppresses emission from electrons with a continuum time longer than $0.65T$. Within this time window the maximum electron energy varies when ϕ is changed. As the ionization probability increases for electrons returningwith higher energy the net result is attosecond pulses with tunable central energy.

Another example of temporal control using a two-colour field can be obtained when$R \approx 25\%$. For this particular value of$R$ the right choice of ϕ results in many electron trajectories that have the same return energy for a large part of the half-cycle. This leads to a dramatic increase in harmonic intensity for this particular energy while the other energies are suppressed (see [29]).

5. Attosecond pulse CEP

The role of the CEP has so far mainly been studied for optical pulses in the visible and near IR wavelength regions. Attosecond pulses have a central frequency in the XUV wavelength region and can therefore be very short and still consist of multiple cycles. As the duration of the attosecond pulses gets shorter, approaching the single cycle limit [32, 35], it gets meaningful to talk also about their CEP.

The addition of the second harmonic to the driving field provides the possibility not only to control the periodicity of the pulses in the generated APT but also the CEP of the pulses [26]. An APT generated by a single colour driving field consists of a train of attosecond pulses with a periodicity equal to half that of the driving field and a π shift between consecutive pulses; see figure 6. The addition of a small amount of second harmonic to the driving field enables control of the CEP. By carefully selecting R and ϕ, consecutive attosecond pulses can be made to accumulate an additional phase of π, which results in an APT that still has a half-cycle periodicity, but with the same CEP from pulse to pulse (figures 5(c) and (d)).

In the frequency domain this corresponds to a comb of even harmonics [30]. The spectral range over which a comb of even harmonics can be obtained is limited by the intrinsic chirp rate of the attosecond pulses, but can be extended by increasing the IR intensity or using longer wavelength drivers [36, 37]. As the blue intensity is increased the attosecond pulse generation will rapidly decrease for every second half-cycle, and a pulse train with only one pulse per cycle is effectively generated for almost all values of ϕ once $R \approx 10\%$. In the generated APT with one pulse per cycle the CEP will also be the same from pulse to pulse (figures 6(e) and (f)).

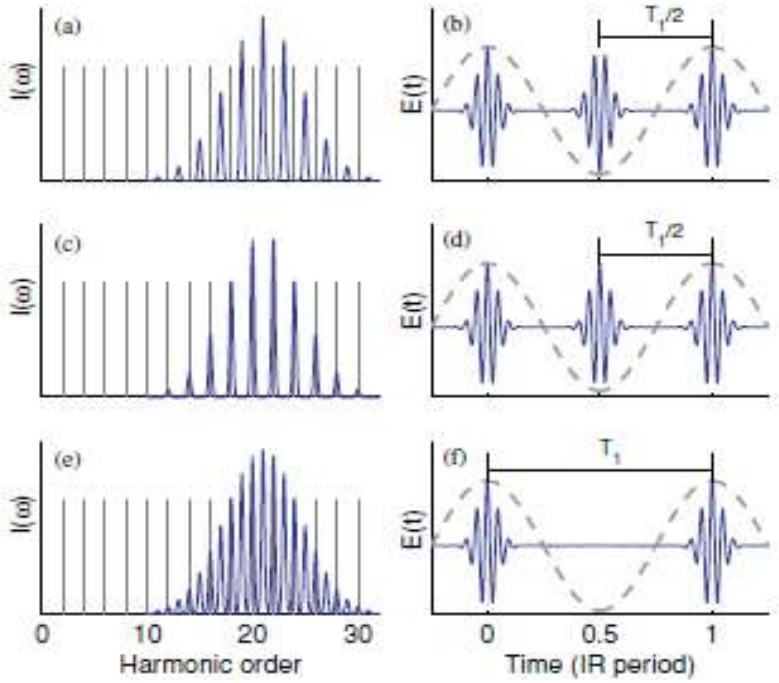

Figure 6. (a) A one colour driving field results in a comb of odd harmonics. (b) Phase locking produces an APT with periodicity equal to half that of the driving field and a π phase shift from pulse to pulse. (c) With a small amount of second harmonic added to the generation process a comb of even harmonics can be produced, which in the time domain (d) corresponds to an APT with periodicity equal to half that of the driving field and no phase shift from pulse to pulse. (e) A stronger blue field results in equally strong odd and even harmonics and (f) an APT with a full IR cycle periodicity and stable CEP.

Furthermore, the increased amount of blue helps to stabilize the CEP of the attosecond pulses across the pulse train [26].

6. Towards XUV–XUV pump-probe Experiments

Dynamics of various systems are frequently studied using pump-probe techniques where the sample is excited by a pump pulse and the time evolution recorded by a probe pulse after a variable delay. This can be generalized to situations where the object is to control the dynamics, e.g., by using additional, intermediate control pulses or by initiating the dynamics at specific times when the outcome is known to be favourable. Such pump-probe and control experiments of electron dynamics call for flexible attosecond pulse sources where the number of pulses, the delay between them and their relative CEP can be tailored.

The number of attosecond pulses can be varied by changing the CEP of a few-cycle pulse driving the high harmonic generation process. If more than a few pulses are desired, a longer driving IR pulse combined will suitable phase matching [38, 39] or polarization gating [40, 41, 35] can be used, or combinations thereof [42]. In order to control the delay between pulses, one needs to vary the period, i.e. the wavelength, of the driving IR field. Continuous tuning over limited intervals is availablewith parametric sources; however, if discrete tunability is sufficient, then it is enough to add the second harmonic blue field to the generation process. This will allow the user to choose between half-cycle and full-cycle intervals with the additional freedom of selecting the relative carrier-envelope phase between consecutive pulses, thus open the door for the possibility of observing CEP effects with attosecond pulses.

Combining a two-colour driving field with few-cycle pulses and/or polarization gating can provide a flexible source to be used in XUV–XUV pump-probe and control experiments, especially if combined with a tunable parametric chirped pulse amplifier system. Indeed, such a double optical gate has been shown to produce isolated attosecond pulses starting with 10 fs driving IR fields [43, 44]. By adding the second harmonic to the driving field

cleaner isolated pulses, with weaker pre- and/or post-pulses can be produced.

The extended continuous spectral bandwidth opens for even shorter isolated attosecond pulses, provided that the right material can be found to compensate the phase variation across the pulses [45].

7. Conclusion

In this paper we have compared the effect of symmetry breaking using a two-colour field with that obtained with few-cycle pulses. The two techniques have many things in common and complement each other very well. With two colour fields the induced asymmetries can easily be made very large and we have demonstrated that this allows us to control the periodicity, the CEP and the central frequency of the generated attosecond pulses. If combined with polarization gating and/or few-cycle pulses this enables the generation of controlled sequences of attosecond pulses. The ability to tailor attosecond pulse sequences in terms of number of pulses, periodicity and CEP will be important when attosecond physics move towards XUV–XUV pump-probe experiments.

Acknowledgments

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