Spectral shaping of attosecond pulses using two-colour laser fields

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Abstract. We use a strong two-colour laser field composed of the fundamental (800 nm) and the second harmonic (400 nm) of an infrared (IR) laser field to generate attosecond pulses with controlled spectral and temporal properties. With a second-harmonic intensity equal to 15\% of the IR intensity the second-harmonic field is strong enough to significantly alter and control the electron trajectories in the generation process. This enables us to tune the central photon energy of the attosecond pulses by changing the phase difference between the IR and the second-harmonic fields. In the time domain the radiation is emitted as a sequence of pulses separated by a full IR cycle. We also perform calculations showing that the effect of even stronger second-harmonic fields leads to an extended tunable range under conditions that are experimentally feasible.

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1. Introduction

Attosecond pulses represent the sharpest instruments available to study ultrafast dynamics [1]. These pulses are formed when a strong laser field interacts with a gaseous medium and the underlying physics is well described by the three step model. In this model the electron tunnels through the atomic potential, which is distorted by the strong laser field (I); gets accelerated by the field in the continuum (II); and may recombine with its parent ion (III) emitting the accumulated excess energy as a burst of extreme ultra-violet (XUV) light [2, 3]. For multi-cycle pulses, the repetition of the process, combined with the inversion symmetry of the atomic potential, leads in the time domain to a train of attosecond pulses separated by half a laser cycle and in the spectral domain to odd harmonics of the laser frequency. Spectral and temporal control of the attosecond pulse generation requires shaping of the generating laser field on a sub-cycle level. One example of such sub-cycle control is the use of phase stabilized few-cycle laser pulses [4], where the short pulse duration leads to a change in the field amplitude between consecutive half-cycles. Another, more elaborate, method is to use pulses with a time-varying polarization [5]–[7]. These methods allow control to be exerted by changing the carrier envelope phase of the laser pulse, in particular leading to the generation of single attosecond pulses [7, 8]. The technique with time-varying polarization also gives control of the photon energy of the pulses [9].

An alternative approach to control the sub-cycle field structure and thereby the electron dynamics is to mix the fundamental infrared (IR) field with its second harmonic [10]–[12]. The combined field can be written as: \( E(t) = E_\omega [\sin(\omega t) + \sqrt{\mathcal{R}} \sin(2\omega t + \varphi)] \), where \( \sqrt{\mathcal{R}} = E_{2\omega}/E_\omega \) is the ratio between the two field amplitudes \( E_{2\omega} \) and \( E_\omega \), \( \omega \) is the IR frequency, and \( \varphi \) a controllable phase difference between the fields, see figure 1(a). The shape and strength of the combined field in consecutive IR half-cycles differ beyond the simple sign change that occurs in a one colour field. The electron trajectories are also altered and instead of the normal two trajectories with excursion times (\( \tau \)) shorter than one IR period (\( T \)), we now get four [12], as illustrated in figure 1(b). Two of these trajectories (a short and a long) come from ionization during the half-cycle when the electric field is strong and recombination during the weaker half-cycle, we will refer to these as high \( \rightarrow \) low trajectories. The other two trajectories start when the field is weak and return during the stronger half-cycle (low \( \rightarrow \) high) with the result that the harmonic emission from these trajectories reaches high photon energies but the yield can be orders of magnitude weaker than the emission from the high \( \rightarrow \) low trajectories due to the difference in ionization probability. Schemes to use the low \( \rightarrow \) high trajectories for isolated attosecond pulse generation have been proposed [13, 14]. We instead use the high \( \rightarrow \) low trajectories which give much higher conversion efficiencies. In previous work, we
Figure 1. (a) IR field (dashed black) ($I = 7.5 \times 10^{13}\text{ W cm}^{-2}$) and second-harmonic fields with $R = 15\%$. The phase differences for the three second-harmonic fields are $\varphi = 0\text{ rad (ice-blue)}$, $\varphi = 0.6\text{ rad (green)}$ and $\varphi = 1.2\text{ rad (red)}$. The resulting E field is the sum of the IR and one of the $2\omega$ fields. The timescale units are IR periods ($T$). (b) Return energy for the high $\rightarrow$ low and low $\rightarrow$ high trajectories calculated using classical mechanics for $\varphi = 0.6$ in solid and dotted green, respectively. Long trajectories are in the shaded regions, $\tau > 0.65T$. (c) High $\rightarrow$ low return for the three phases shown in (a). The dashed black curves in (b) and (c) show the return energy for a one-colour IR field. The kinetic energy of the electrons is given in units of the ponderomotive energy $U_p$ for the IR field. $\tau$ is the time the electron spends in the continuum from ionization to recombination and $\Delta E$ is the tunable energy range.

have demonstrated that adding a second harmonic with a relative intensity of $R = 10\%$ will induce a sufficiently strong variation between consecutive IR-half-cycles to efficiently cancel the generation every second half-cycle, which results in the generation of an attosecond pulse train (APT) with only one pulse per IR cycle [12]. In combination with the polarization gating technique mentioned above this can lead to the generation of isolated attosecond pulses [15].

In this paper we show that the field structure within each half-cycle can be controlled by using an even stronger blue field. The generated APT still consists of one pulse per IR cycle and the signature of the additional control is that central energy of the attosecond pulses can be tuned by changing $\varphi$. We demonstrate that already $R = 15\%$ is sufficient to tune the peak of the harmonic spectrum between 23 and 29 eV. We also perform calculations to investigate the influence of higher field strengths. We find that the tunable range can be extended to $2.5U_p$, $U_p$ being the ponderomotive energy of the IR, for $R = 50\%$ and by carefully choosing the conditions a narrow frequency range can be selected and further enhanced.

In figure 1(c), we plot the return energy as a function of return time for the high $\rightarrow$ low trajectories for three different values of $\varphi$ together with the return energy for a one colour driving field. For a one colour driving field the highest return energy (cut-off) corresponds to an excursion time ($\tau$) of $0.65T$ where $T$ is the period of the IR field [16]. Trajectories with longer excursion times result in more divergent harmonic emission and can hence be strongly reduced after the generation using an aperture [17]. For the two-colour field we will continue to use $\tau = 0.65T$ to distinguish between short and long trajectories since the divergence of
the emission mainly depends on the time spent in the continuum. The highest energy reached during the short (high → low) trajectory depends strongly on the relative delay between the two fields, \( \varphi \), figure 1(c). We find both experimentally and theoretically that we get most of the emission where the slope of the return energy curve decreases. In some cases the curve goes through a maximum at this point (figure 1(c) blue curve). By selecting only the emission from the short, high → low trajectories we thus get a pulse train with only one pulse per cycle and a central energy that can be tuned by changing the delay between the two fields. The central energies of spectra calculated within the strong field approximation (SFA) for the same parameters are marked with + in figure 1(c) and the energy difference between the two markers i.e. the tunable range is denoted \( \Delta E \).

2. Experimental method

The setup used to generate the attosecond pulses and characterize their spectral and temporal properties is schematized in figure 2. Incoming 35 fs, 800 nm, 2 mJ laser pulses are sent through a 0.6 mm KDP (potassium dihydrogen phosphate), type I crystal for second-harmonic generation. The IR and the generated second-harmonic radiation (blue) are separated in a dichroic interferometer where the phase difference between the two colours, \( \varphi \), can be adjusted. Before the IR and the blue are combined at the exit of the interferometer, the polarization of the blue is made parallel with the IR and a fraction of the IR is split off as a probe. After the interferometer the two-colour field is focused into a 1 kHz pulsed Ar gas-target to generate harmonics, which are filtered spectrally and spatially. The spectral filtering is done using a 200 nm thick aluminium (Al) transmission filter that blocks the two-colour driving field and the low-order harmonics and improves the synchronization of the transmitted harmonics [18]. The filter does not significantly affect the shape of the high harmonic spectrum as the transmission.
of Al is rather constant in the studied spectral region (20–40 eV)\(^4\). The emission is spatially filtered using a 1.5 mm aperture, located 400 mm from the laser focus, which efficiently blocks the more divergent contributions from the long trajectories [19]. The spatial filter serves the dual purpose of being a recombination mirror. The back side is a convex mirror off which the probe is reflected and made collinear with the XUV light.

The harmonic emission and the co-propagating probe are focused into an atomic Ar beam inside a velocity map imaging spectrometer (VMIS) [20]. The VMIS projects the 3D momentum distribution of the ejected photoelectrons onto a micro-channel plate detector coupled to a phosphor screen, which is read out by a CCD camera. From the measured 2D projection, the full 3D momentum distribution of the photoelectrons can be unambiguously retrieved using an iterative inversion algorithm [21], and from this the photoelectron and corresponding photon spectra shown below can be easily extracted. With the probe present at the time of ionization the energy distribution of the electrons can be shifted as a function of XUV–probe delay [22]–[24]. The shift in momentum is proportional to the vector potential of the probe field, \( A(t) \), at the time of ionization, \( \Delta p = -e A(t) \). The streaking of the photoelectrons allows us to recover the duration of the attosecond pulses [25] since the pulse periodicity is matched exactly to a full IR cycle and consecutive ionization events are identical for a multi-cycle laser [26]. The strength of the probe field has to be chosen below that necessary to turn the photoelectrons around and thereby induce coherent electron scattering [26] in order not to perturb the temporal characterization. Coherent electron scattering occurs when the dimensionless parameter \( \tilde{\gamma} = \sqrt{\frac{\hbar \omega_c - I_p^2}{2U_p}} \) is less than 1, where \( \hbar \omega_c \) is the central energy of the XUV light and \( I_p \) is the ionization potential of the target atom. In the experiment \( \tilde{\gamma} = 3.7 \) and no electron–ion interaction is expected.

3. Results

Experimental results showing spectral shaping are presented in figures 3(a) and (c). In (a) harmonic spectra are plotted as a function of \( \varphi \), for an IR intensity estimated to be \( 7.5 \times 10^{13} \text{ W cm}^{-2} \) and \( \mathcal{R} = 15\% \). The odd and even harmonics have comparable strengths and oscillate with \( \varphi \) with a period of \( \pi \). The positions of the oscillation maxima depend strongly and almost linearly on the harmonic order. As a consequence, the central frequency can easily be varied simply by changing \( \varphi \). The measured tunability extends over 6 eV, limited mainly by the target gas (Ar) and the intensities used. The tunable range, \( \Delta E \), can be increased by increasing the IR intensity while keeping \( \mathcal{R} \) fixed. Higher intensity might demand a target gas with higher ionization potential. For a fixed \( \mathcal{R} \), \( \Delta E \) is proportional to \( U_p \) and thus the intensity. For the intensity we use in the calculations and experiment presented in figures 1 and 3, \( \Delta E \approx U_p \).

We also perform a temporal measurement at a phase \( \varphi \) marked with green in figure 3(a). The phase was chosen to be within the range where the APT consists of one pulse per IR cycle, i.e. between the ice-blue and red lines. In figure 4, the presence of the probe shifts the photoelectron distribution. The on-axis photoelectron spectra are plotted as a function of the XUV–probe delay in (a) together with an angular distribution in (b), taken at the XUV–probe delay indicated by the white line in (a), where streaking is maximized. The fact that the energy transfer in (a) is either upward or downward proves that we only have one attosecond pulse per IR cycle. The phase of the attosecond pulses can be retrieved either by considering the

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Figure 3. (a) and (b) photon energies as a function of $\varphi$, (a) an experimental scan and (b) a calculated scan where only short trajectories are included. (c) and (d) spectra for minimum and maximum energy taken at the relative phases marked with ice-blue and red lines in the scans, (c) experimental and (d) calculated. The green line in (a) indicates the relative phase for which the pulse duration was measured.

Figure 4. (a) Electron energy distribution along the polarization axis as a function of XUV–probe delay. (b) Electron momentum distribution at the XUV–probe delay when streaking is maximized. The probe-field is polarized along the $y$-axis. (c) Momentum asymmetry as a function of GDD, red curve and left $y$-axis, and pulse duration as a function of GDD, green curve and right $y$-axis. The ice-blue arrows point out the path starting from the measured $\Delta p_1/\Delta p_2$ to the retrieved GDD where it meets the red curve and the pulse duration where it meets the green curve.
full scan [27] (figure 4(a)) or the up/down asymmetry, $\Delta p_1/\Delta p_2$, of the angular distribution at XUV-probe delays corresponding to maximum streaking i.e. when $A(t) = 0$ at the time of ionization (figure 4(b)). We choose the latter technique which in principle can be used to measure the pulse-duration of the attosecond pulses with a single shot. Electrons leaving the atom upward and downward to the probe polarization direction are affected differently by the probe field [22]. In figure 4(b) the upward electrons get a more narrow distribution than the downward electrons ($\Delta p_1 < \Delta p_2$). For each combination of harmonic spectrum and probe intensity the ratio $\Delta p_1/\Delta p_2$ depends on the group delay dispersion (GDD) of the attosecond pulses in a unique way. The probe intensity can be extracted from the maximum momentum shift in the scan. For our measured harmonic spectrum and probe intensity we calculate, using the SFA, how the electron distributions change with GDD. This allows us to deduce $\Delta p_1/\Delta p_2$ as a function of GDD (figure 4(c) red line). The variation of the pulse duration with GDD is indicated by the green line. From our experimentally measured ratio, $\Delta p_1/\Delta p_2 = 0.85$, we obtain a GDD of 0.015 fs² and a pulse duration of 300 as. The negative GDD of the Al-filter compresses the pulse from 450 as towards the transform limit of 250 as.

Our theoretical calculations consist of solving the time-dependent Schrödinger equation within the SFA [28]. The atomic dipole $\langle x(\omega) \rangle$ from the calculations is converted to an intensity spectrum by taking the product $\omega^4 |\langle x(\omega) \rangle|^2$. Experimentally, contributions from the short trajectories can be selected through phase matching and spatial filtering [29]. To mimic this effect in the single atom calculations we restrict the SFA integration to include only the excursion times with: $0 < \tau < 0.65T$. Comparing calculations and experiment, in figure 3, we find a good agreement between the experimental (a) and the calculated (b) spectra. Note that the calculations, based on a continuous driving field, give information about the intensity but not the shape of individual harmonics. The experimental results together with the calculations confirm the idea of sub-cycle control induced by the second-harmonic field. The probability for ionization and return of the electron wave packet is increased over a certain energy range close to the highest detected energy. This energy range can be tuned by changing the phase difference $\phi$.

We investigate theoretically how the sub-cycle control can be improved by increasing $R$ even further. The situation where $R = 25\%$ and $\phi = 0.6$ (see figure 5) stands out in particular since the electron is pulled back with an almost constant energy over an extended period of time, $\sim T/4$, i.e. there are many sub-cycle tunnelling times that lead to the same final energy (figure 5(b)). This results in a significant enhancement of the harmonic emission at that particular energy. The intensity within the enhanced energy region is well above the already strong harmonics for $\phi = 0$ and 1.2 rad in figure 5(c).

For $R > 25\%$, the electron is pulled back to the atom in a more complex way and the return energies are now highly dependent on $\phi$. In figure 6(b), the calculated return energy is plotted as a function of $\tau$ for three phase differences. We note that the energy curve can peak twice; once for $0 < \tau < 0.65T$ and another time for $0.65T < \tau < T$. The highest obtainable energy within the short and long trajectory integration windows vary in opposite ways with $\phi$. This results in a central photon energy that decreases with $\phi$ for the short trajectories and increases for the long trajectories, see figures 6(a) and (c). Under these conditions, $2 \times 10^{14}$ W cm⁻², $R = 50\%$, the tunable range is increased and we find that it is feasible to tune the energy over $2.5U_p$. 

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Figure 5. (a) A calculated scan of $\varphi$ including the phases presented in (b) and (c). The calculations are done for an IR intensity of $2 \times 10^{14}$ W cm$^{-2}$ and $\mathcal{R} = 25\%$. (b) Kinetic energy of the returning electrons for the three $\varphi$-values shown in figure 1, $\varphi = 0$ rad (ice-blue), $\varphi = 0.6$ rad (green) and $\varphi = 1.2$ rad (red). (c) Harmonic spectra showing the enhanced emission at 42 eV corresponding to the flat region in the return energy curve (b).

Figure 6. Calculations for an IR intensity of $2 \times 10^{14}$ and $\mathcal{R} = 50\%$. (a) SFA calculations including short trajectories, $0 < \tau < 0.65 T$. (b) Electron return energies for $\varphi = -0.4$ rad (ice-blue), $\varphi = 0.6$ rad (green) and $\varphi = 1.6$ rad (red). (c) SFA calculations for long trajectories, $0.65 T < \tau < T$. The $\varphi$-values used to calculate the return energies are indicated in the scans by the vertical dashed lines. The horizontal dashed lines mark the peak of the spectra and connect the peak to the return energy curves.

4. Conclusion

We have shown that by altering the driving laser field on a sub-cycle level using a two-colour laser field, we can control the spectral properties of the generated APT by varying the phase difference $\varphi$ between the IR and its second harmonic. With this method we have extended the spectral control beyond that of transmission filters. For applications this will be very useful as the photon energy of the emitted attosecond pulses can be tuned close to or just below the
ionization potential of a target gas to study below threshold ionization [30]. For almost all values of \( \varphi \) one pulse per IR cycle is generated. For a sequence of pulses the central frequency can be tuned by varying \( \varphi \) except for a narrow transition \( \varphi \)-region where two pulses per cycle are generated. In the same manner, isolated attosecond pulses generated by a two-colour field can be tuned in frequency by varying \( \varphi \) within the range where only one pulse is generated. By increasing the second-harmonic intensity compared to the experiment presented here, we predict that the tunable frequency range can be extended further and provide a flexible attosecond source to meet future needs.

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