A case study for terahertz-assisted single attosecond pulse generation

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Abstract
We numerically investigate the use of strong THz radiation in assisting single attosecond pulse generation by few-cycle, 800 nm laser pulses. We optimize focusing conditions to generate short and powerful single attosecond pulses of high-energy photons by keeping the parameters of the THz field within the limits achieved experimentally. We show that using optimal focusing geometry isolated attosecond pulses shorter than 100 as can be obtained even in the absence of further gating or XUV compression techniques, using an 8 fs generating pulse. Furthermore, quantum path control of short- and long-trajectory components is demonstrated by varying the delay between the THz and IR pulses.

1. Introduction and motivation
Recent developments in THz field generation resulted in the production of extreme high electric fields exceeding 100 MV cm\textsuperscript{-1} with a carrier frequency up to 72 THz and a stable carrier-to-envelope phase [1]. This field strength is already comparable with the peak electric field of laser pulses used for high-order harmonic generation (HHG) in noble gases (usually between 300 and 1000 MV cm\textsuperscript{-1}). Since THz fields with the highest intensities are produced by different frequency generation from amplified laser pulses, the resulting THz pulses are naturally synchronized in time with the laser pulses [1].

Single-atom calculations have already revealed several aspects of HHG assisted by THz or static electric fields. The production of even harmonics and the increased emission rate in the lower plateau region has been demonstrated when multi-cycle infrared (IR) laser pulses are used as the driving field [2]. With higher intensity THz or static electric fields, the extension of the cutoff has been observed producing a double-plateau-structured spectrum [3–5]. Using few-cycle laser pulses, the addition of the THz field can create a supercontinuum in the spectra enabling the production of single attosecond pulses (SAP) [6]. With chirped IR driving pulses the created supercontinuum, theoretically, can support 10 attosecond short SAPs [7]. In calculations predicting the production of SAPs however the used amplitude of the THz or static electric field has been higher than what is achievable experimentally, and the used laser pulses were at most 6 fs long [6, 7]. The experimentally obtained 100 MV cm\textsuperscript{-1} electric field requires very tight focusing of the THz beam.

Here, we present the effects of THz field on HHG in gases beyond the single-atom level, by modelling the process in a realistic focusing geometry, and using parameters of the THz pulse obtained experimentally by Sell \textit{et al} [1]. In a recent study [8], it was shown that in the presence of a THz field macroscopic effects can help the selection of a SAP from the attosecond pulse train when using 8 fs driving pulses. In this study, we discuss the importance of focusing geometry on phase matching and harmonic yield, and show that despite the limited THz pulse energy the most powerful SAP can be produced by relatively loose focusing and lower THz electric-field strength. We also show that the selection of the short- or long-trajectory components can be achieved by varying the delay between the THz and IR pulses.

Our goal is to model experimental conditions as close as possible; therefore, a full three-dimensional model has been used which allows the optimization of parameters that can be measured and controlled experimentally [9]. For calculating the single-atom response, the established theory of strong-field
approximation is used, known to well reproduce the important aspects of HHG in gases [10]. The propagation equations for the laser, THz, and harmonic fields are solved by using a Fourier transform method in paraxial approximation [11], by taking into account the effects of absorption, dispersion on atoms and on electrons, and the optical Kerr effect. The three fields propagate independently in a medium with refractive indices mainly influenced by the plasma created by the total electric field.

2. Results

2.1. Basic configuration

We have modelled harmonic generation in neon gas by 8 fs, 800 nm IR laser pulse combined with a THz pulse having 19 μJ energy and 72 THz carrier frequency (4.17 μm wavelength) focused at 31 μm spot size producing 108 MV cm⁻¹ peak field strength as generated and measured by Sell et al [1]. The pulses have been assumed to be Gaussian both in space and time, the THz pulse having 76 fs duration to be consistent with the above mentioned parameters. The beam diameters are assumed to be 5 mm and 26 mm for the IR and THz beams, respectively, so that using an optical element with f = 0.6 m focal distance yields the same w₀ = 31 μm beam waist for both. The two fields have collinear polarization and propagate in the same direction, while the gas medium containing neon at 33 mbar pressure is placed right after the focus. The wavelength, energy and duration of the THz pulse were consistent with the experimentally demonstrated values through all the calculations, while the energy of the IR pulse (usually not a limiting factor in experiments) has been chosen to produce 6 × 10¹⁴ W cm⁻² intensity in the focus for all cases.

In order to optimize the generating conditions, the effects of several parameters were studied. The focusing geometry was optimized, limited by the constraints of using maximum spatial overlap between the IR and THz pulses (i.e. same beam waist). Besides the focusing conditions the delay between the two pulses, the length and position of the gas cell were optimized in order to generate short and powerful SAPs of high-energy photons.

Our calculations show that indeed the cutoff of the harmonic radiation can be extended from 110 to 200 eV in the near field (see figure 1) when an 8 fs, 78 μJ IR pulse is used in the above mentioned focusing geometry in a 2 mm long target cell. However, the tight focusing used to obtain the extreme high electric field of the THz pulse results in a Rayleigh range of just 0.7 mm (3.7 mm for the IR beam), which is not beneficial to phase match the generated harmonics as illustrated in figure 1(b) showing decreasing signal after 2 mm propagation. By increasing the beam waist, the peak intensity of the THz field drops, limiting the achievable cutoff, but helping to phase match the higher spectral components.

2.2. Single attosecond pulses

Using f = 1.7 m focusing and 0.585 mJ IR pulse energy, the corresponding beam waist is 85 μm; the addition of the THz field with 38 MV cm⁻¹ peak amplitude and same 85 μm waist extends the cutoff by 30 eV (see figure 2(a)), reaching 154 eV compared to the cutoff at 124 eV obtained using only the IR pulse. The plateau region is phase matched during propagation through a 5 mm gas cell. This configuration supports the production of 145 as SAP obtained by selecting harmonics above 116 eV (see figure 3(e)). The larger interaction volume resulting from the increased spot size and favourable phase matching conditions through a 5 mm cell results in an increase of more than two orders of magnitude in the peak power of the
Table 1. Summary of the THz field and SAP parameters obtained for different focusing geometries using two different spectral filters, and cell lengths optimized for SAP peak power. Contrast ratio labelled $\infty$ means that the contrast is higher than the precision of our calculations ($\approx 10^{7}$).

| $f$ (m) | $w_0$ ($\mu$m) | $E_{\text{peak}}$ (MV cm$^{-1}$) | $L_{\text{optimal}}$ (mm) | cutoff (eV) | SAP duration (as) | SAP peak power (arb. units) | contrast (dB) |
|--------|---------------|-------------------------------|-------------------------|-------------|------------------|---------------------|--------------|
| 0.61   | 31            | 108                           | 2.1/0.6                 | 205         | 97/136           | 14.2/0.62           | 22.3/144     |
| 1.1    | 56            | 60                            | 5.1/1.6                 | 163         | 110/184          | 384/24.1            | 22.1/138     |
| 1.7    | 86            | 38                            | 5.2/2.3                 | 155         | 150/230          | 1947/68.2           | 21.6/138     |
| 2.45   | 125           | 27                            | 4.3/2.1                 | 148         | 174/260          | 3085/29.8           | 12.2/138     |

$^{a}$ 116/150 eV stands for distinguishing the two spectral filters.

Figure 3. Attosecond pulses obtained by selecting harmonic radiation above 116 eV (harmonic 75) using different focusing geometries with the IR only (top row), and with the combined field (bottom row). The transform limit of the pulses is also shown in cases when SAP is obtained.

Figure 4. Attosecond pulses obtained by selecting harmonic radiation above 150 eV (harmonic 97) using different focusing geometries with the combined field. The transform limit of the pulses is also shown.

Selecting only harmonics above 150 eV (harmonic 97), SAP can be obtained in all the focusing geometries used so far (see figure 4 and table 1). However, the loose focusing ($f = 2.5$ m) also shifts the cutoff below 150 eV, resulting in a reduced pulse power compared to the case with $f = 1.7$ m focusing. We note that the SAP obtained this way is nearly transform limited (figure 4(c)), because cutoff harmonics possess no chirp [12] and the bandwidth is very narrow.

The 50 as transform limit of the SAP presented in figure 3(d) corresponds to an effective bandwidth of 36 eV (with a time-bandwidth product of 0.44, characteristic of Gaussian pulses), which can be explained by the strong drop of the harmonic yield at $\approx 145$ eV seen in figure 1(b). By selecting only harmonics above 150 eV, the spectrum is flatter which explains the shorter transform limit of the synthesized SAP shown in figure 4(a).

The differences in the degree of cutoff extension by the THz field at different focusing geometries can be attributed not only to the different amplitude of the THz field, but also to macroscopic effects as well [8]. Using saddle point analysis, we have found that at single-atom level a long wavelength THz (compared to the 800 nm IR) or static electric field increases the harmonic radiation’s cutoff by 0.4–1 eV for each MV cm$^{-1}$ field strength. Although this number also depends on the intensity and wavelength of the IR (it is $\approx 0.65$ eV/(MV cm$^{-1}$) for 800 nm IR having $6 \times 10^{14}$ W cm$^{-2}$ peak intensity), and the scaling with the THz or dc field amplitude is not exactly linear. For a more detailed analysis of the mechanism of cutoff extension see for example the paper by Taranukhin et al [5].

A summary of the results is presented in table 1. The values for SAP duration, peak power and contrast presented in
focusing is used producing 56 $\mu$m (see figure 5(a)), where macroscopic effects do not start to play, the radiation produced in a very short cell (0.2 mm, see $r$, $t$ plotted in (a)) maps (see figure 5). First we calculate the table are calculated for the optimal cell lengths in the actual focusing geometry, whereas the values presented in figures 3 and 4 are calculated for cell lengths more commonly used in experiments. The contrast ratio is defined between the peak powers of the two most powerful attosecond pulses.

For the position of the gas cell, we have found that the most reliable solution is to place it with the input pinhole in the focus. With the looser focusing geometries, slightly better results were obtained by moving the cell 1 mm before the focus; however, the increase is not significant. With the $f = 1.7$ m case for example 10% increase can be obtained in the SAP peak power when an extended, 6 mm long cell is used, having the input pinhole 1 mm before the focus.

2.3. Quantum path control

So far, the two pulses were assumed to be synchronized, i.e. to have a field maximum at $t = 0$. In this case, the short-trajectory components are phase matched and the corresponding long ones are eliminated due to phase mismatch. To demonstrate the effect, the intensity of the propagated XUV pulse is plotted in ($r$, $t$) maps (see figure 5). First we calculate the radiation produced in a very short cell (0.2 mm, see figure 5(a)), where macroscopic effects do not start to play, and thus illustrating the single atom results. On axis ($r = 0$), we observe almost half a cycle of modulated radiation consisting of both short- and long-trajectory components [8, 10] generating earlier (short) and later (long) emissions. The short- and long-trajectory components merge into the cutoff with increasing radial coordinate corresponding to decreasing IR and THz field strength. Our analysis (not illustrated here) shows that both short- and long-trajectory components are generated at any $z$ coordinate in the cell; however, the long ones gradually disappear from the propagated field due to phase mismatch (see figure 5(b)).

We find that the short trajectories dominate the harmonic radiation after 1 mm propagation (not shown), with the long ones completely eliminated after 2 mm. Using the same $f = 1.1$ m focusing mirror (56 $\mu$m beam waist), and delaying the THz pulse by 1 fs compared to the IR, we observe an almost identical field intensity map at the beginning of the cell (see figure 5(c)). However, during propagation only long trajectories are phase matched in the first mm of the gas cell, with the corresponding short ones eliminated (see figure 5(b)). We conclude that by using well-chosen focusing and varying the delay between the THz and IR pulses, the selection of long trajectories can be achieved allowing one to select the sign of the chirp of the resulting SAP.

Another effect reported in single-atom calculations is the increase of the emission rate in the lower part of the plateau. We have also observed this effect; however, in the best of our cases the harmonic yield increased by only several times compared to the IR only case, not by orders of magnitude as reported in [3].

3. Conclusion

In conclusion, we have analysed the effects of experimentally obtained THz pulses on HHG by modelling the process using a complete three-dimensional model. The importance of focusing geometry on phase matching and harmonic yield has been discussed and shown that, despite the limited THz pulse energy, the most powerful SAP can be produced by relatively loose focusing. Short, isolated attosecond pulse having a duration of 97 as is predicted using tight focusing and 2 mm long gas cell. We have also shown that the selection of the short- or long-trajectory components (defining the sign of the resulting SAP’s chirp) can be achieved by varying the delay between the THz and IR pulses.

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