Ideal laser waveform construction for the generation of super-bright attosecond pulses

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Keywords: high harmonic generation, attosecond pulse, nonlinear optics

Abstract

A new scheme is proposed to synthesize an ideal laser waveform via the superposition of few-cycle and half-cycle pulses with an optimized delay time or laser wavelengths and intensity matching. The numerical analysis confirms that this scheme is much more efficient for supporting the production of an isolated super-bright attosecond pulse, which provides a way in the final realization for the attosecond pump and attosecond probe techniques.

With the remarkable advances achieved in the field of ultrafast optics, the investigations on high-order harmonic generation (HHG) have already become routine in ordinary laboratories with a focus on strong laser physics or photonics, due to its potential applications in molecular tomography, production of ultrafast extreme ultraviolet (XUV) light sources, and so on [1–5]. Continuous efforts in HHG projects have eventually led to the birth of attosecond pulse trains (APT) or isolated attosecond pulse (IAP), whose successful usage in probing electron wavepacket dynamics with attosecond-temporal and angstrom-spatial resolutions has been confirmed [6–9].

In contrast to the attosecond pulse train, an isolated attosecond pulse is more beneficial for usage, especially when measuring and controlling the ultra-fast electronic motion inside an atom or molecule. As for the production of an isolated bright attosecond pulse, the essential prerequisite condition is an HHG plateau with simultaneous ultra-broad, ultra-smooth, and ultra-intense characteristics [10–20]. In practice, if the driving pulse for creating the HHG has a suitable laser waveform, such as that shown in figure 1(a), the above prerequisite condition would be easily satisfied. Based on the semi-classical ‘three-step’ model [21, 22] for HHG, if the target atoms are ionized mainly around the field peak within a narrower time domain (Zone 1 in figure 1(a)), electrons would be born with a higher ionization rate. In addition, if these electrons are subsequently accelerated by the laser field within a broader time domain with relatively weak amplitude (Zone 2 in figure 1(a)), it means the traveling time is extended for acceleration. The travel time extension and simultaneous ionization yield’s enhancement would push more electrons to gain more laser energy before recombination and the harmonics photon’s emission. If such a specific waveform is possible, an isolated bright attosecond pulse would be produced. Although researchers have made versatile tries to approach this final aim, the harvest is still not satisfactory.

In this work, a more practical new scheme to synthesize the above ideal laser waveform is proposed via a three-color field with superposing two half-cycle pulses (HCP) to a few-cycle pulse (FCP) (such as that in figure 1(b)). Here, when superimposed by one or two unipolar HCPs at different positions (i.e., with suitable time delays), the FCP’s laser waveform is significantly modified [23], which inversely and naturally would induces new nonlinear effects [24–29]. As for such a superposition mechanism for a specific laser waveform, we try it in the HHG process and check whether it is possible for supporting the generation of an isolated ultra-bright attosecond pulse. As shown below, the answer is yes.

The ideal laser waveform first is synthesized via the above mechanism, then it is used to interact with argon (Ar) atoms to investigate the HHG process numerically, based on the Lewenstein model [21] merging with the ADK (Ammosov–Delone–Krainov) theory [30]. The basic ingredients are one FCP and two HCPs. The synthesized three-color field can be expressed as
would be enhanced. The key is to enhance the instantaneous laser intensity at Zone 1 ionization rate at Zone 1 much. However, because of the weaker ionizing energy via laser acceleration through the cutoff energy extends so 150 eV, although its spectral intensity is significantly reduced. Electrons, ionized in Zone 2, gain much acceleration when passing through a weaker field, where the compensation between HCPs and FCPs plays a role. More interesting is the occurrence of the second plateau, which is much longer, with a cutoff energy around 50 eV. Except for this, a large spectral modulation is clearly shown in the plateau of the HHG spectrum, which is the result of the interference of long and short quantum paths. If investigating the driving field carefully, shown in figure 2(b), the dominant ionization only occurs at Zones 2 and 3 around the laser peak (ellipses indicated), due to the limitation of the few-cycle oscillations. Electrons, ionized in Zone 2, gain much acceleration when they pass through Zone 3 with a high enough laser amplitude, which contributes to the formation of a longer plateau with high transformation efficiency and a simultaneous larger cut-off energy (solid line in figure 2(a)).

The harmonics intensity is determined mainly by the ionization probability at Zone 2, where the electrons are born. Therefore, if we want to increase the harmonics intensity in the plateau, to add an additional HCP with a peak exactly at Zone 2 to an FCP would be necessary (figure 2(c)), and the corresponding HHG spectrum (dashed line in figure 2(a)) clearly shows the occurrence of two plateaus. The first plateau (low-order part of the HHG) is narrower, but with the comparable spectral intensity to that with only one FCP as a driver. With a careful investigation of the synthesized laser field (figure 2(c)), it is not difficult to find that the narrowing of the first plateau is because those electrons born at Zone 2' then gain weaker acceleration when passing through a weaker field, where the compensation between HCPs and FCPs plays a role.

More interesting is the occurrence of the second plateau, which is much longer, with a cutoff energy around 150 eV, although its spectral intensity is significantly reduced. Electrons born at Zone 1’ gain the maximum energy via laser acceleration through the field at Zone 2’, where the two-color combined field’s amplitude is enhanced due to the positive contribution of HCP to FCP. This is the reason why the cutoff energy extends so much. However, because of the weaker ionizing field amplitude and corresponding obvious decrement of the ionization rate at Zone 1’, based on ADK ionization theory, the spectral intensity is reduced. Naturally, if the second plateau (dashed line in figure 1(a)) is enhanced, the accompanying attosecond pulse would be enhanced. The key is to enhance the instantaneous laser intensity at Zone 1’, which subsequently induces more electrons to contribute to the second plateau. In order to achieve the aim, now we add another HCP to form a three-color field, as shown in figure 2(d). As for the corresponding HHG spectrum in figure 2(a) (dotted line), as expected, the spectral intensity of the second plateau enhances by several orders in magnitude greater than that for the two-color case, and simultaneously the cut-off energy is almost fixed.

Moreover, most importantly, this second plateau becomes much smoother, indicating the multi-path interference effects nearly disappear in favor of the production of an isolated ultra-short attosecond pulse.
As for a clear insight into the generation of such an additional longer and smoother plateau in the HHG spectrum, the time-frequency analysis to the dipole moment is implemented, corresponding to the above three cases, respectively. The results are shown in figure 3.

When only one FCP interacts with the atoms, the multi-path interferences can be found, as shown in figure 3(a), which is consistent with those in figure 2(a). If when only one HCP is added at Zone 2', as shown in figure 3(b), the contribution from both the short and long quantum paths is comparable, such as in the photon energy range of 100–180 eV, which finally results in the clear spectral modulation occurring in the second plateau of the HHG spectrum (red dashed line in figure 2(a)). Generally, if one part of the continuum spectrum with a constant spectral phase is selected, for example here with photon energy from 70–120 eV, via performing the inverse fast Fourier transformation, an attosecond pulse or pulse train can be obtained. As shown in figure 3(e), a dominant attosecond pulse (from the short path’s contribution) with around 128 attosecond pulse duration is directly obtained, which is accompanied by a much weaker satellite pulse (from the long path’s contribution). In contrast, if two HCPs are added at Zone 2" and Zone 1", respectively, with a suitable time delay (see figure 2(d)), the contribution from the long quantum path is completely suppressed, as shown in figure 3(c). The dominant contribution from the short path is the reason why the second plateau is so smooth. Simultaneously, the satellite pulse disappears (see figure 3(f)). Importantly, the laser intensities of the attosecond pulse are higher than that of a single FCP of duration, which contains only one optical cycle, as shown in figure 3(d).

Moreover, the short path’s contribution is enhanced by one order for the two-HCP case compared to that for the one-HCP case (figures 3(b) and (c)). The corresponding isolated attosecond pulse is enhanced by around 170 times, as indicated in figures 3(e) and (f). Therefore, not only the intensity but also the isolation property of the attosecond pulse is improved, and the above demonstration confirms the significant advantage of this ideal waveform reconstruction scheme.

The above demonstration has confirmed the effectiveness of the three-color laser synthesizing scheme for supporting the generation of an isolated intense and short attosecond pulse. As for practical application, a further optimization procedure to the laser parameters is necessary. The first step is to determine the wavelength range of the two HCPs so that the strongest isolated attosecond pulse is obtained. As for an isolated bright attosecond pulse, it is anticipated that the accompanied satellite pulse is suppressed, but simultaneously the main attosecond pulse is not affected. It is natural to define the intensity ratio between $I_{\text{short}}$ and $I_{\text{long}}$ from the
short and long quantum paths, respectively, as an indicator for the characterization to the attosecond pulses. Their dependence on the central wavelengths of both HCPs is shown in figure 4. Figures 4(a)–(c) clearly show that if the wavelengths of HCPs are set at around $\lambda_1 = 1700$ nm and $\lambda_2 = 350$ nm, they will be the optimal values for the production of an isolated attosecond pulse with enough energy flux, with the satellite pulses suppressed.

Figure 3. Time-frequency analysis of the HHG driven by (a) one FCP; (b) one FCP and one HCP combination; (c) one FCP and two HCPs combination. (d)–(f) are three different electric fields for the attosecond pulses synthesized within the photon energy range of 70–120 eV.

Figure 4. Peak intensity (a) $I_{\text{short}}$ (corresponding to the short path), (b) $I_{\text{long}}$ (corresponding to the long path), and (c) intensity ratio ($I_{\text{short}}/I_{\text{long}}$) of attosecond pulses versus the wavelengths $\lambda_1$ and $\lambda_2$ of the two HCPs. (d) Intensity ($I_{\text{short}}$) of attosecond pulse versus the laser peak intensity $I_2$ of the second HCP.
After the two suitable wavelengths are chosen, as a second step, the laser peak intensity of the HCP can be further optimized to be $1.9 \times 10^{14}$ W cm$^{-2}$, as shown in figure 4(d). After the above two steps of optimization, one ideal laser waveform can be constructed (figure 1(b)), which induces a much smoother super-continuous spectrum and keeps it intense enough simultaneously. It is the key for the production of an isolated intense attosecond pulse.

One half-cycle pulse has already generated experimentally for about 20 years since the first demonstration [23]. Subsequently, one HCP with an intensity of $10^{11} \sim 10^{13}$ W cm$^{-2}$ has been utilized extensively [24–29]. One may suspect its practical realization and think it is far from obvious and seems just a mere speculation, based on the consideration that to produce one HCP, especially with a stable carrier-envelope phase or suitable central wavelength, is still difficult.

In order to relax this limitation, now we use a several-cycle pulse instead of the above HCPs. The laser field parameters we used for three-color-field synthesizing are 1) The lasers wavelength are $\lambda_0 = 800$ nm, $\lambda_1 = 1600$ nm, and $\lambda_2 = 400$ nm; 2) The pulse durations and time delays are $\tau_0 = 5$ fs, $\tau_1 = 35$ fs, $\tau_2 = 15$ fs, $t_0 = 0$, $t_1 = 0.25$ T, and $t_2 = 0.75$ T (T is the time period of the 800 nm laser field); 3) The laser intensities are $I_0 = 2.5 \times 10^{14}$ W cm$^{-2}$ and $I_1 = I_2 = 1.0 \times 10^{14}$ W cm$^{-2}$, respectively.

Fortunately the demonstration to the three-color-field scheme for producing an intense attosecond pulse is also successful, and the results (figures 5 and 6) are quite similar to those demonstrated above. The clearly enhanced and super-continuous second plateau (dotted line in figure 5(a)) exists, and the time-frequency analysis to the dipole moments (see figure 6(a)) clearly shows that the short quantum path plays the main contribution to the second plateau of HHG. Via selecting one part of the second plateau of the HHG continuum spectrum with a constant spectral phase, the harmonic photon energy within the range of 160–240 eV, for example here, and then performing the inverse fast Fourier transformation, an isolated attosecond pulse can be directly obtained (see figure 6(b)). After a suitable phase compensation treatment, an isolated attosecond pulse with 50 as duration can be generated (see figure 6(c)).

Moreover, the sensitivity to the time delays between the three fields of the attosecond pulse’s peak intensity or duration is investigated (see figure 6(d)). With the time delay increasing from some small value, its intensity first enhances to a peak and then decreases gradually. The pulse duration first decreases rapidly, then changes slowly, and finally increases rapidly. When the time delay is 0.26 T, the attosecond pulse’s intensity is maximum, and simultaneously the pulse duration is about 48 as. The advantage is that, on the time delay from around 0.25 T–0.28 T, the attosecond pulse’s duration is non-sensitively dependent on the time delay, although its peak intensity changes between one and half of its optimal value.

Figure 5. HHG spectra under three kinds of laser combination schemes (a) and the corresponding laser waveform and ionization rate for each case: single-color (b), two-color (c), and three-color (d).
Up to now, the above three-color-laser synthesizing scheme for supporting the generation of an isolated intense attosecond pulse has been successfully demonstrated. In conclusion, because the contribution of HCPs can significantly modify the driving laser waveform, the advantages of the new proposal to synthesize an ideal laser waveform for supporting the production of an isolated super-bright attosecond pulse were demonstrated. One interesting thing is that the presence of one HCP or a short several-cycle pulse can induce a two-plateau structure in the HHG spectrum. With suitable laser intensity, wavelengths, and time delays in the three-color-field scheme, one much more intense, broader, and smoother super-continuous spectrum appears, which is in favor of creating an isolated ultra-intense attosecond pulse. Moreover, the above phenomena can also be obtained by synthesized three-color laser fields. This provides a new scheme to create an isolated intense attosecond pulse and provide an additional way for true application in the attosecond pump–probe technique.

**Acknowledgments**

The authors are grateful to Prof. Y Cheng and J Yao for inspired discussions and the research funds from NNSF (Grants No. 11134010, No. 11374318, No. 11374202, and No. 11474223) of China, the Qing Lan Project of Jiangsu Province, the 100-talents Plan of the Chinese Academy of Sciences, and the Department of Human Resources and Social Security of China.

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