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Ultraviolet Source Assisted Enhancement of Attosecond Pulse

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(Dated: Received on October 8, 2014; Accepted on November 6, 2014)

A promising method to improve the attosecond pulse intensity has been theoretically presented by properly adding an ultraviolet pulse into the orthogonal two-color field. The results show that by properly adding a 125 nm ultraviolet pulse to the orthogonal two-color field, not only the harmonic yield is enhanced by 2 orders of magnitude compared with the original orthogonal two-color field case, but also the single short quantum path, which is selected to contribute to the harmonic spectrum, results in an ultrabroad 152 eV bandwidth. Moreover, by optimizing the laser parameters, we find that the harmonic enhancement is not very sensitive to the pulse duration and the polarized angle of the assisted ultraviolet pulse, which is much better for experimental realization. As a result, an isolated pulse with duration of 38 as can be obtained, which is 2 orders of magnitude improvement in comparison with the original two-color orthogonal field case.

Key words: High-order harmonic generation, Isolated pulse, Two-color orthogonal field, Ultraviolet source

I. INTRODUCTION

Attosecond extreme-ultraviolet pulse (XUV) generation has been a hot issue due to its potential applications, such as probing and controlling the electron motions inside atoms, molecules, solids [1] and tracing the bound electron motion [2−4] *etc.* Currently, one of the most successful ways to achieve this attosecond pulse is connected with the high-order harmonic generation (HHG) from the rare gases [5−7], which can be well understood by means of the "three-step" model [8], where harmonics are emitted through the steps of tunneling ionization, acceleration, and recombination. Further, an attosecond pulse could be produced by making an inverse Fourier transformation of several harmonics [9−18]. However, the applications of the generated attosecond pulses [9−18] are limited due to the low output energy. Hence, how to enhance the attosecond pulse intensity poses an interesting but challenging issue in attosecond science.

Recently, an alternative technique, named as ultrashort ultraviolet (UV) pulse assisted enhancement of the attosecond sources has attracted much attention [19]. For instance, Li *et al.* [20] obtained an intense 26 as pulse by combination of an 800 nm chirped field and its 27th harmonic pulse. Feng *et al.* [21] obtained an intense 41 as pulse by using the two-color field combined with the ultrashort 29.6 nm pulse.

However, the method mentioned above has some limitations, *i.e.* (i) the high intensity ultrashort UV pulse ($\tau_{\rm UV}$=0.5 fs in the former investigations) is difficult to be obtained in many laboratories, (ii) the model used in all the above theories is for He^+ ion, which is still an experimental challenge for the generation of an ionic gas with sufficient density, (iii) the fundamental fields used in the former investigations are all linear polarization. As we know that the HHG process is highly dependent on the ellipticity of the laser field [22−24], therefore, the time-varying polarization gating is a very important technique to the harmonic selection.

In this work, we present another efficient method to attosecond pulse enhancement by properly adding a UV pulse into the two-color field. Here, the model we used is the neutral He atom, which is much easier to obtain in experiment, and the fundamental field is chosen to be the orthogonal polarization two-color field. It shows that with the introduction of a proper 125 nm UV pulse into the two-color orthogonal field, the harmonic intensity is enhanced by 2 orders of magnitude. Moreover, the harmonic plateau is contributed by the single short quantum path. Further analyses show that this harmonic enhancement is independent of the pulse duration and the polarized angle of the controlling UV pulse. Finally, by properly Fourier transformation of these enhanced harmonics, a 38 as pulse with 2 orders of magnitude improvement can be obtained.

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II. THEORY

In this work, the HHG spectra can be investigated by solving the time-dependent Schrödinger equation (TDSE). In the dipole approximation and the length gauge, the TDSE is given by the following equation, atomic units (a.u.) are used throughout this work unless stated otherwise.

$$i\frac{\partial\varphi(x,y,t)}{\partial t} = \Big[-\frac{1}{2}\frac{\partial^2}{\partial x^2} - \frac{1}{2}\frac{\partial^2}{\partial y^2} + V(x,y) + xE_x(t) + yE_y(t)\Big]\varphi(x,y,t) \quad (1)$$

$$V(x,y) = \frac{-1.0}{\sqrt{x^2+y^2+0.07}} \quad (2)$$

where $V(x,y)$ is the soft Coulomb potential of He atom. We use the sine basis functions to depict a DVR for the translational coordinates x and y [25, 26] as follows,

$$\langle R_i|n\rangle = \sqrt{\frac{2}{L}}\sin\left(\frac{n\pi R_i'}{L}\right) = \sqrt{\frac{2}{L}}\sin\left(\frac{in\pi}{N+1}\right) \quad (3)$$

here for convenient illumination, we use R to depict the x and y coordinates, where L=$R_{\max}$−$R_{\min}$, R_i'=$i\Delta R$, i=1,2,3,...,N, and ΔR=$L/(N$+1). The DVR basis $|\bar{R}_i\rangle$ can be defined as $\langle\bar{R}_i|n\rangle$=$\sqrt{\Delta R}\langle R_i|n\rangle$. Propagation of the time-dependent electronic wave function $\varphi(x,y,t)$ can be carried out using the standard second-order split-operator method [26−33]. The synthesized laser field is expressed as:

$$\begin{aligned}\mathbf{E}(t) = {} & E_1 f_1(t)\cos(\omega_1 t)\hat{x} + E_2 f_2(t)\cos(\omega_2 t)\hat{y} + \\ & E_3 f_3(t-\tau_{\text{delay}})\cos[\omega_3(t-\tau_{\text{delay}})]\cos(\theta)\hat{x} + \\ & E_3 f_3(t-\tau_{\text{delay}})\cos[\omega_3(t-\tau_{\text{delay}})]\sin(\theta)\hat{y}\end{aligned} \quad (4)$$

here E_i and ω_i (i=1−3) are the amplitudes and the frequencies of the two-color orthogonal field and the UV pulse, respectively. τ_{delay} is the delay time between the orthogonal field and the UV pulse. θ is the polarized angle of the UV pulse (the angle between the two polarization axes). The envelope function is,

$$f_i(t) = \exp\left[-\frac{4\ln(2)t^2}{{\tau_i}^2}\right] \quad (5)$$

where τ_i (i=1−3) are the pulse durations of the three pulses.

According to the Ehrenfest theorem [34], the time-dependent dipole acceleration can be written in the form,

$$\begin{aligned}a(t) &= -\langle\varphi(x,y,t)|\nabla\mathbf{V}(x,y) + \mathbf{E}(t)|\varphi(x,y,t)\rangle \\ &= -\langle\varphi(x,y,t)|\left[\frac{\partial V(x,y)}{\partial x} + E_x(t)\right]\mathbf{e}_x \\ &\quad + \left[\frac{\partial V(x,y)}{\partial y} + E_y(t)\right]\mathbf{e}_y|\varphi(x,y,t)\rangle \\ &= a_x(t)\mathbf{e}_x + a_y(t)\mathbf{e}_y\end{aligned} \quad (6)$$

The HHG spectra can be obtained by Fourier transforming the time-dependent dipole acceleration $a(t)$,

$$\begin{aligned}S(\omega_1) &\approx \left|\int \exp(-i\omega_1 t)a(t)\mathrm{d}t\right|^2 \\ &= \left|\int \exp(-i\omega_1 t)[a_x(t)\mathbf{e}_x + a_y(t)\mathbf{e}_y]\mathrm{d}t\right|^2 \\ &\approx S_x(\omega_1) + S_y(\omega_1)\end{aligned} \quad (7)$$

Finally, the attosecond pulse can be obtained by harmonic superposing as follows,

$$I(t) = \left|\sum_q \int a(t)\exp(-i\omega_1 t)\mathrm{d}t\exp(i\omega_1 t)\right|^2 \quad (8)$$

III. RESULTS AND DISCUSSION

Figure 1 shows the HHG spectra driven by the linear and the orthogonal two-color field, respectively. The two-color field is 5 fs/800 nm, I_1=1.0×10^{15} W/cm^2 and 10 fs/1600 nm, I_2=1.0×10^{14} W/cm^2. Clearly, with the introduction of the polarized angle of the two-color field (here is the orthogonal polarized field), the modulation on the harmonic spectrum is remarkably decreased in comparison with the linear two-color field case, which is beneficial to the isolated pulse selection.

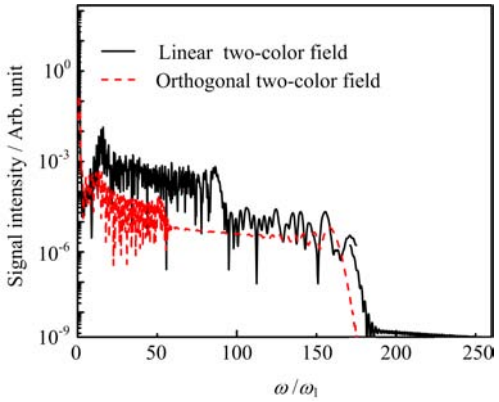

FIG. 1 HHG spectra of the linear two-color field and the orthogonal two-color field. The laser fields are 5 fs/800 nm, I_1=1.0×10^{15} W/cm^2 and 10 fs/1600 nm, I_2=1.0×10^{14} W/cm^2.

DOI:10.1063/1674-0068/28/cjcp1410171

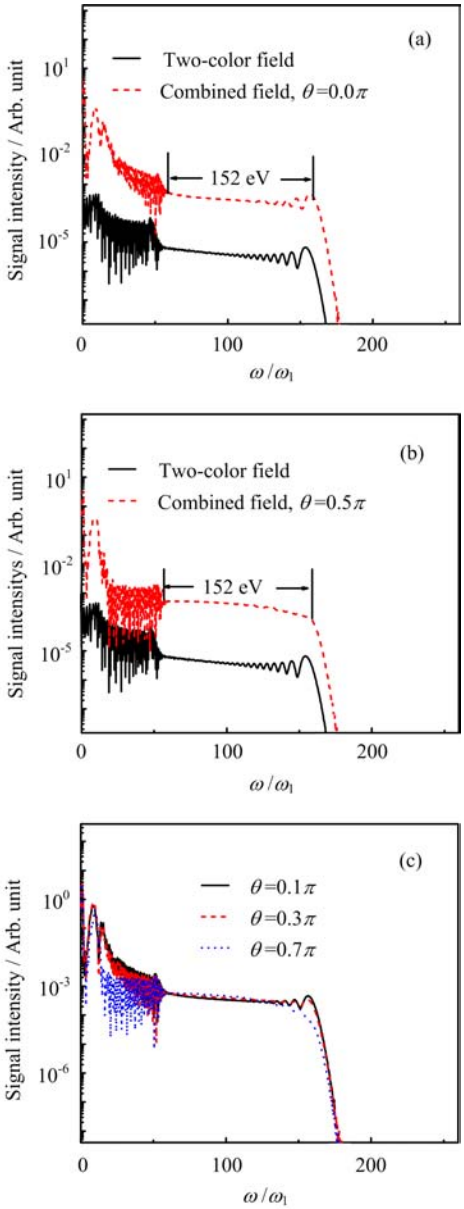

FIG. 2 (a) HHG spectra of the orthogonal two-color field and the orthogonal two-color field combined with the 1 fs/125 nm, I_3=5.0 kW/cm^2 UV pulse with θ=0.0π, $\omega_1\tau_{\rm delay}$=0.8π. (b) HHG spectra of the orthogonal two-color field and the orthogonal two-color field combined with the 1 fs/125 nm, I_3=5.0 kW/cm^2 UV pulse with θ=0.5π, $\omega_1\tau_{\rm delay}$=0.8π. (c) Polarized angle (θ) effect on the harmonic spectra.

Figure 2 (a) and (b) show the HHG spectra of the orthogonal two-color field combined with the 1 fs/125 nm, I_3=5.0×10^3 W/cm^2 UV pulse with θ=0.0π, $\omega_1\tau_{\rm delay}$=0.8π (which means adding the 125 nm pulse in x direction) and θ=0.5π, $\omega_1\tau_{\rm delay}$=0.8π (which means adding the 125 nm pulse in y direction), respectively. Clearly, with the introduction of the 125 nm pulse at $\omega_1\tau_{\rm delay}$=0.8π (this is the optimal time delay

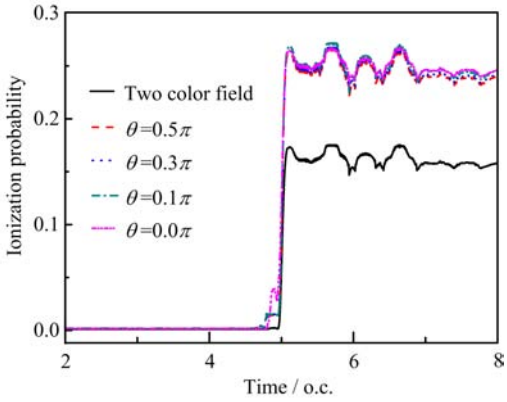

FIG. 3 Ionization probabilities of (a) the orthogonal two-color field and the combined field (two-color field+125 nm pulse) with different polarized angle of $\omega_1\tau_{\rm delay}$=0.8π, θ=0.5π, 0.3π, 0.1π, and 0.0π.

both in x and y directions), the harmonic yields have been enhanced by 2 orders of magnitude compared with the original two field case, which is beneficial to the isolated pulse enhancement. Figure 2(c) shows the polarized angle (θ) effect on the harmonic enhancement. The other parameters are the same as those in Fig.2 (a) and (b). It shows that the harmonic enhancement phenomenon is not very sensitive to the polarized angle (θ). It means that one can add this UV pulse with an arbitrary polarized angle, and the harmonic enhancement can always happen, which is much better for experimental realization.

According to the "three-step model" [8] and the previous investigations [20, 21], we know that the harmonic yields are strongly dependent on the ionization probabilities (IPs). Therefore, to understand the physical origin of the harmonic enhancement, in Fig.3, we calculate the IPs of the above cases. We see that with the introduction of the UV pulse, the IPs are enhanced compared with the original two-color field case, thus leading to a largely augmented harmonic efficiency [21]. Moreover, the enhanced IPs produce no distinct changes with the variety of the polarized angle, which is responsible for the harmonic insensities to the polarized angle.

To better understand the harmonic emission process, in Fig.4, we present the time-frequency of harmonic spectra, obtained by using the wavelet transformation of the dipole acceleration $a(t)$ [35]. For the linear two-color field case, as shown in Fig.4(a), there are five energy bursts on the harmonic emission process. Moreover, each burst receives similar contributions from the right path (called the long quantum path having earlier ionization and later recollision) and the left path (called the short quantum path with later ionization but earlier recollision) [36], which is responsible for the large modulation on the harmonic. For the orthogonal two-color field case, as shown in Fig.4(b), we see that for the maximum energy burst, the contribution of the short quantum path is much larger than that of the long quantum

DOI:10.1063/1674-0068/28/cjcp1410171

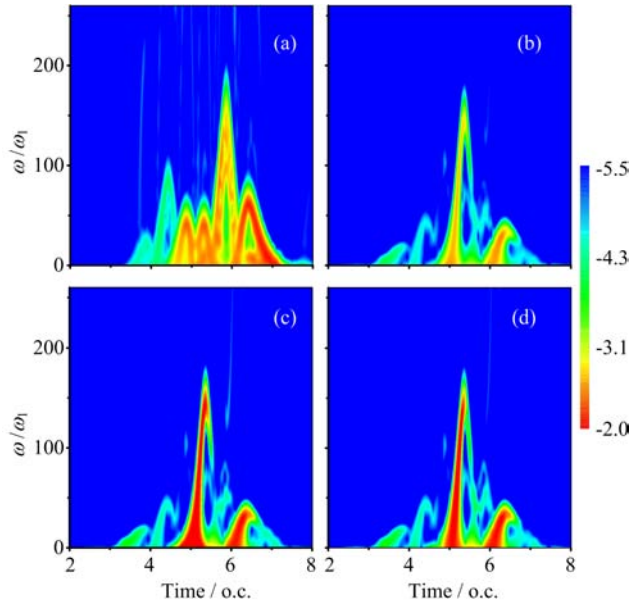

FIG. 4 The time-frequency distributions for (a) the linear two-color field, (b) the orthogonal two-color field, (c) the orthogonal two-color field combined with the 1 fs/125 nm, I_3=5.0 kW/cm^2 UV pulse with θ=0.0π, $\omega_1\tau_{\rm delay}$=0.8π, and (d) the orthogonal two-color field combined with the 1 fs/125 nm, I_3=5.0 kW/cm^2 UV pulse with θ=0.5π, $\omega_1\tau_{\rm delay}$=0.8π. o.c. means the optical cycle of 800 nm pulse in all the following figures unless stated otherwise.

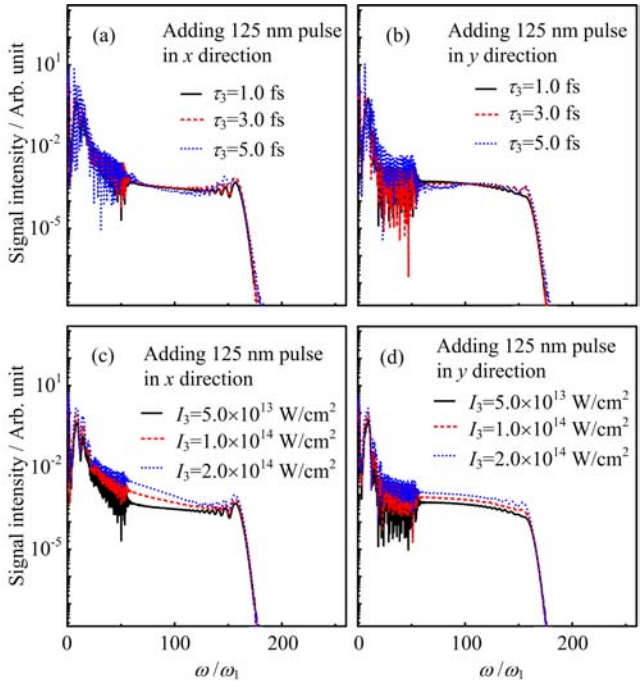

FIG. 5 (a) and (b) Controlling pulse duration (τ_3) effect on the harmonic generation. (c) and (d) Controlling pulse intensity (I_3) effect on the harmonic spectra.

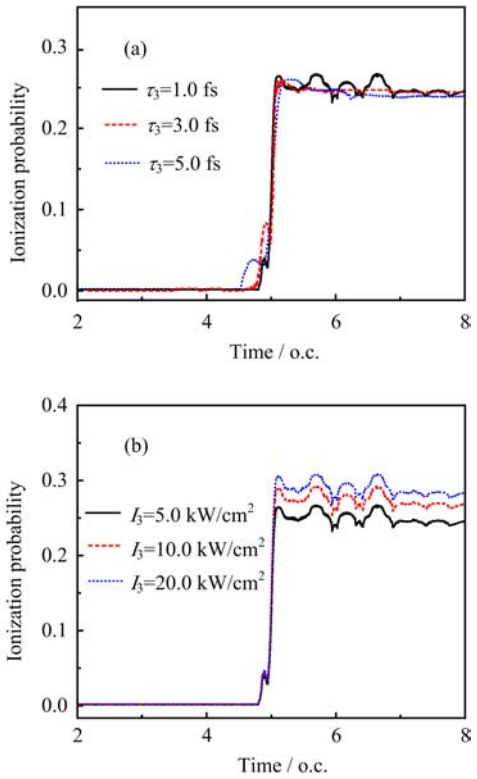

FIG. 6 (a) Ionization probabilities of the combined field with different controlling pulse duration. (b) Ionization probabilities of the combined field with different controlling pulse intensity.

path, which is responsible for the small interference on the harmonic spectrum. While for the cases of the orthogonal two-color field combined with the UV pulse in x and y directions with $\omega_1\tau_{\rm delay}$=0.8π, as shown in Fig.4 (c) and (d), the harmonic intensity has been remarkably enhanced in comparison with the orthogonal two-color field case (see the color-bar). Moreover, the single short quantum path has also been selected to contribute to the maximum harmonic emission burst, which is beneficial to the isolated attosecond pulse generation.

Figure 5 (a) and (b) show the controlling pulse duration (τ_3) effect on the harmonic enhancement. The other parameters are the same as those in Fig.2 (a) and (b), respectively. Clearly, the HHG spectra are almost invariable with the variety of the pulse duration of τ_3, suggesting that HHG is rather independent of the pulse duration. Therefore, based on the practical application, choosing the longer pulse duration (τ_3=5 fs) is much better for experimental realization. Figure 5 (c) and (d) show the controlling pulse intensity (I_3) effect on the harmonic spectra. It shows that with the increasing of the controlling pulse intensity, the harmonic yields are further enhanced, however, the enhanced ratio is less than 1 order of magnitude. Thus, in this work, we choose I_3=5.0×10^{13} W/cm^2 as a proper controlling pulse intensity and a 152 eV supercontinuum with 2 or-

DOI:10.1063/1674-0068/28/cjcp1410171

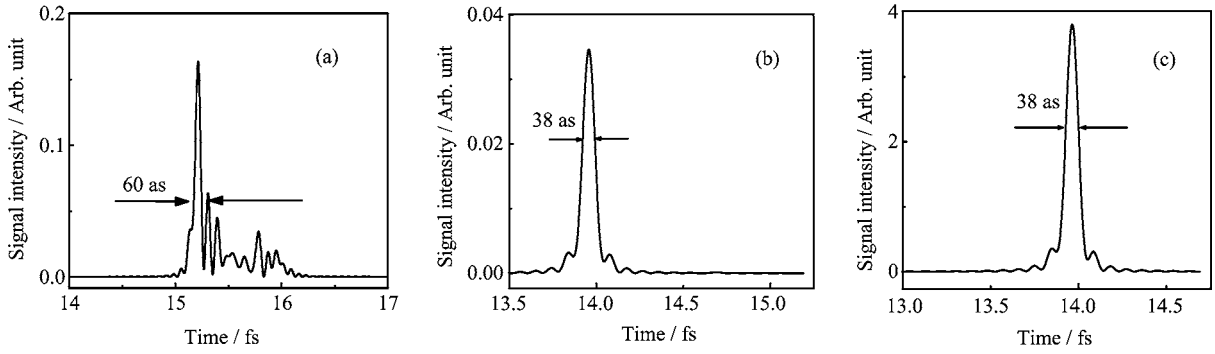

FIG. 7 The temporal profiles of the attosecond pulses by superposing harmonics of (a) the attosecond pulse generation from the linear two-color field, (b) the orthogonal two-color field from the 70th to the 150th orders, and (c) the orthogonal two-color field combined with the 125 nm UV pulse with θ=0.0π, $\omega_1\tau_{\rm delay}$=0.8π from the 70th to the 150th orders.

ders of magnitude improvement can be obtained, which will be favorite to support attosecond pulse enhancement.

Figure 6(a) shows the IPs of the combined field with different τ_3. The other parameters are the same as those shown in Fig.2(a) dash line. It shows that with the increasing of τ_3, the IPs are slight decreased. But the decreased grade is very small, thus we almost can not see that changes on the harmonic spectra shown in Fig.5 (a) and (b). Figure 6(b) shows the IPs of the combined field with different I_3. The other parameters are the same as those shown in Fig.2(a). Clearly, with the increasing of the controlling pulse intensity, the IPs are enhanced due to the increasing of the amplitude intensity of the combined field, which is the main reason for the harmonic enhancement shown in Fig.5 (c) and (d).

Figure 7 shows the temporal profiles of the attosecond pulses. Firstly, by superposing the harmonics of the linear two-color field from the 100th to the 190th orders, a 60 as pulse with accompanying satellite pulses can be obtained, as shown in Fig.7(a). Clearly, this pulse is not beneficial to practical application. Further, by properly superposing the harmonics of the orthogonal two-color field from the 70th to the 150th orders, an isolated 38 as pulse can be produced, as shown in Fig.7(b). Finally, if we choose the harmonic spectra of the orthogonal two-color field combined with the UV pulse with θ=0.0π, $\omega_1\tau_{\rm delay}$=0.8π (here, only one condition is shown because that the others are similar to it) and by properly superposing the harmonics from the 70th to the 150th orders, a 38 as isolated pulse with 2 orders of magnitude improvement can be obtained, as shown in Fig.7(c).

IV. CONCLUSION

We theoretically present an effective method to enhance the attosecond pulse intensity. The results show that with the proper introduction of a 125 nm UV pulse into the orthogonal two-color field, not only the harmonic yield is reinforced by 2 orders of magnitude, but also the single short quantum path is selected to contribute to the harmonic emission. As a result, an isolated 38 as pulse with 2 orders of magnitude improvement has been obtained. Moreover, through analyzing the laser parameters, we find that the harmonic and the attosecond pulse enhancement phenomena are independent of the pulse duration and the polarized angle of the introduced UV pulse, which is good for experimental realization and application. Thus, it is expected that our proposed scheme may bring useful insight into practical generation and application of the intense ultrashort attosecond pulses.

V. ACKNOWLEDGMENTS

The authors thank Professor Ke-li Han for providing us the computational code used in the present work. This work was supported by the Scientific Research Fund of Liaoning University of Technology, China (No.X201319 and No.X201312) and the Scientific Research Fund of Liaoning Provincial Education Department (No.L2014242).

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DOI:10.1063/1674-0068/28/cjcp1410171