High-intensity attosecond high-order harmonic generation driven by a synthesized laser field

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(Received 27 April 2004; published 9 November 2004)

The scheme of high-intensity attosecond high-order harmonic generation driven by a synthesized laser field is proposed. The synthesized laser field is obtained by an appropriate superposition of a few-cycle laser pulse and a relatively long pulse of several tens of femtoseconds. Calculated results show that the intensity of the attosecond high-order harmonic pulse in helium driven by the synthesized laser field with a 8.8×10^{13} W/cm^2/5 fs laser pulse and a 3.51×10^{14} W/cm^2/50 fs laser pulse is several orders of magnitude higher than that driven by a single 8.8×10^{13} W/cm^2/5 fs laser pulse, and it is even stronger than that driven by a single 7.9×10^{14} W/cm^2/5 fs laser pulse, although the single and the synthesized pulses have the same peak electric-field strength.

DOI: 10.1103/PhysRevA.70.053809 PACS number(s): 42.65.Ky, 32.80.Rm, 42.50.Hz

The generation of attosecond pulses is very important and it opens a brand new chapter in ultrafast spectroscopy [1,2]. Just as femtosecond pulses allow the observation of the motion of atoms in molecules, attosecond pulses will allow one to trace electronic motion and electronic relaxation (such as inner-shell dynamics) in atoms and molecules. A number of methods have been proposed for attosecond pulse generation [3–5]. High-order harmonic generation (HHG) is currently the experimentally furthest advanced method that can supply single attosecond pulses, which is essential for many potential applications. Recently Hentschel *et al.* produced single attosecond x-ray pulse with a 7-fs laser pulse as the driving laser for high-order harmonics [2].

In current experiments on the attosecond pulse generation, the few-cycle driving laser pulse (typically 0.7 mJ/5 fs) was obtained by the postamplification spectral broadening technique in which a hollow-fiber chirped-mirror high-power compressor was used [6]. The energy of the few-cycle laser pulse produced by this method is presently limited in the level of 1 mJ due to the ionization of the gas atoms in the hollow fiber. At the same time, the conversion efficiency of the XUV/x-ray pulses in the HHG is very low [6,7]. The XUV/x-ray pulses generated by HHG process are currently too weak to induce measurable nonlinearities of atomic media. As a result, the cross-correlation method is used in the experiment of measuring the duration of the generated attosecond pulses. Although the x-ray pulse duration can be measured with this method, to produce an attosecond pulse with higher intensity is highly desired for other measurements and its application, e.g., the detailed temporal characterization of the x-ray pulse may need higher energy and higher intensity.

In the previous work of HHG [8–11], some methods have been adopted to enhance the conversion efficiency of HHG. With phase-matching scheme [8–10], the conversion efficiency of HHG can be increased by two or three orders. Also, with the shaped pulse scheme [11,12], Bartels *et al.* [11] demonstrated a different method to enhance the conversion efficiency of HHG. With an optimized laser pulse shape, strong constructive interference can be obtained between emissions from different electron trajectories, which can selectively enhance a particular harmonic order. This is so-called "intra-atomic phase-matching." This result shows that a coherent control can be used to enhance the efficiency of HHG.

In this paper we propose a synthesized laser field scheme to enhance the intensity of attosecond pulses produced via HHG. The synthesized laser field is obtained by an appropriate superposition of a few-cycle laser pulse and a relatively long pulse of several tens of femtoseconds whose energy can be increased to the level of several tens of millijoules.

The approach in the calculation of attosecond HHG is similar to that described in previous works [13,14]. We calculated the dipole acceleration by solving numerically the time-dependent Schrödinger equation (TDSE). The synthesized laser pulse in the calculation is described by $E(t)=E_1(t)\cos(\omega t+\varphi_1)+E_2(t-t')\cos[\omega(t-t')+\varphi_2]$, where ω is the frequency of the laser field and φ_i the absolute phase, $E_i(t)=E_i\exp(-t^2/\tau_i^2)$ denotes the envelope of the driving electric field, where E_i is the peak amplitude of the laser pulse and $(2\ln 2)^{1/2}\tau_i$ is its duration, $i=1, 2$. t' is the delay between two pulses. The wavelength of the laser pulse is 800 nm. The dipole acceleration is then Fourier transformed to obtain the spectrum, and a time-frequency analysis (the Gabor analysis [15]) finally provides the temporal profile of the harmonics. We calculated the HHG in helium atoms and the attosecond pulses are generated at the cutoff regime of the HHG.

Figure 1 shows the temporal profiles of the attosecond high-order harmonic pulses produced by a single 5-fs laser pulse and a synthesized laser pulse of 50- and 5-fs pulse pair. The laser parameter in the calculation is 7.9×10^{14} W/cm^2/5 fs (corresponding to the electric-field amplitude $E=0.15$ a.u.) for the single laser pulse case and 8.8×10^{13} W/cm^2/5 fs($E_1=0.05$ a.u.)$+3.51\times10^{14}$ W/cm^2/50 fs($E_2=0.1$ a.u.) for the synthesized pulse case. The carrier-envelope phase and the time delay between two pulses are zero in the calculation. The attosecond x-ray pulses are produced in the cutoff regime of HHG and the photon energies (174 eV in the calculation) of the attosecond x-ray pulses are the same. From this figure one can see that

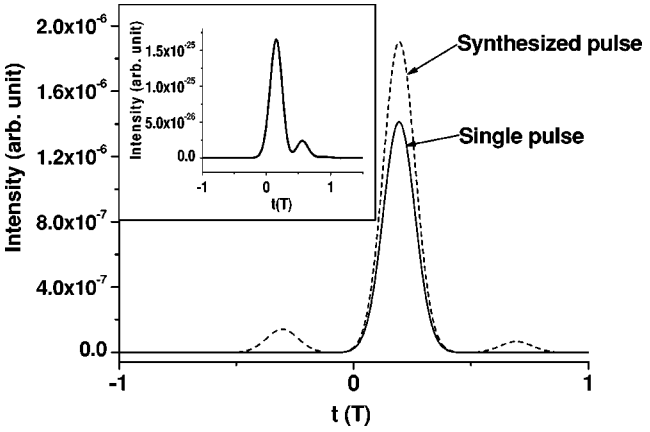

FIG. 1. The temporal profile of attosecond x-ray pulses generated by single few-cycle laser pulse (7.9×10^{14} W/cm^2/5 fs, solid line) and the synthesized pulse (8.8×10^{13} W/cm^2/5 fs+3.51 $\times10^{14}$ W/cm^2/50 fs, dashed line). Although the peak amplitude of the electric field in the two cases are the same, the intensity of the attosecond x-ray pulse generated by the synthesized pulse is higher than that generated by the single 7.9×10^{14} W/cm^2/5 fs laser pulse. The inset shows the temporal profile of the attosecond x-ray pulse with the same photon energy generated by the single laser pulse of 8.8×10^{13} W/cm^2/5 fs in the synthesized pulse.

not only a single attosecond x-ray pulse can be produced with the synthesized pulse, but its intensity is even higher than that generated by a single 5-fs laser pulse with the same intensity. For comparison, the inset in this figure shows the attosecond x-ray pulse with the same photon energy generated by only the 8.8×10^{13} W/cm^2/5 fs part of the synthesized laser pulse. The intensity is lower by 19 orders of magnitude in comparison with that driven with the addition of 50-fs pulse (because there is only the noise in the calculation for the result of the single 8.8×10^{13} W/cm^2/5 fs laser pulse), although the difference in the driving laser electric field is only three folds.

Within this synthesized pulse scheme, the carrier envelope phase (CEP) of the laser pulse is very important. Our calculation shows that, comparing with the case of zero CEP difference, the intensity of the attosecond pulse will be reduced by half if the CEP difference of two laser pulses is $\pi/8$. When the difference of the CEP of two pulses increases, the intensity of the attosecond pulse decreases quickly. But this does not mean that the scheme is difficult to realize in a realistic experiment. According to Ref. [16], the CEP jitter emerging in the waveguide was found to be less than 50 mrad. That means the difference of CEP of two laser pulses is quite stable. To implement this proposal experimentally is therefore straightforward (Fig. 2). One can use a normal chirped pulse amplification laser system to obtain 50-fs laser pulse and split it into two parts, and then send one part into a hollow fiber chirped mirror compressor to obtain a few-cycle laser pulse [6], and then combine and synchronize the two pulses by a suitable delay line before delivering the synthesized laser pulse to the laser atom interaction region for HHG. Figure 3 shows the effect of CEP when the two laser pulses have good time delay. It is similar to the result of a single driven laser pulse with different CEP's [13]. If the

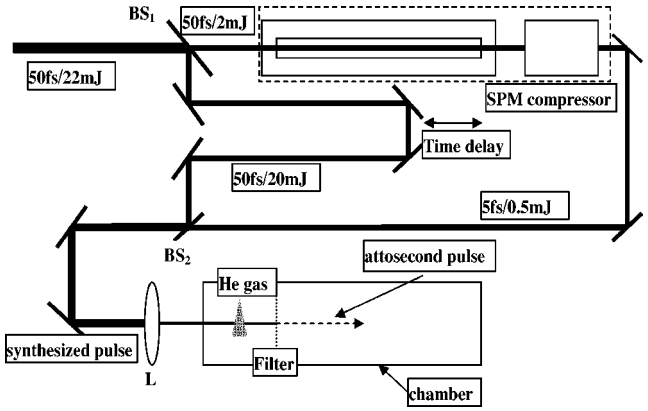

FIG. 2. The schematic layout of the synthesized pulse method. The laser pulse is split into two beams by BS_1. One part is compressed with the hollow fiber chirped mirror compressor and a 5 -fs pulse is generated. Then two pulses are combined and synchronized by suitable delay line at BS_2. Within the synthesized pulse, most energy is offered by the long pulse (50-fs pulse). The synthesized pulse can be focused loosely and more atoms can participate in the interaction.

CEP of the driven laser pulse is stabilized, similar results can be obtained as that in Ref. [16].

Another important issue is the effect of E_1/E_2 on the attosecond pulse generation. When $E_1/E_2=0$, the synthesized pulse is just a long pulse which can only generate a train of attosecond pulse [17]. When E_1/E_2 is infinity, the synthesized pulse is just a few-cycle laser pulse that can be used to generate a single attosecond pulse. To confirm the validation of this scheme, we have also investigated the effect of the electric-field ratio of the 5-fs pulse to the 50-fs pulse on the HHG process and the calculated temporal profiles of the attosecond pulses as functions of the electric-field ratio are shown in Fig. 4. In the calculation, we changed E_1, the amplitude of the electric field of the 5-fs pulse, while E_2, the amplitude of the electric field of the 50-fs pulse, was kept constant as 0.1 a.u.. Other parameters in the calculation are the same as those for Fig. 1.

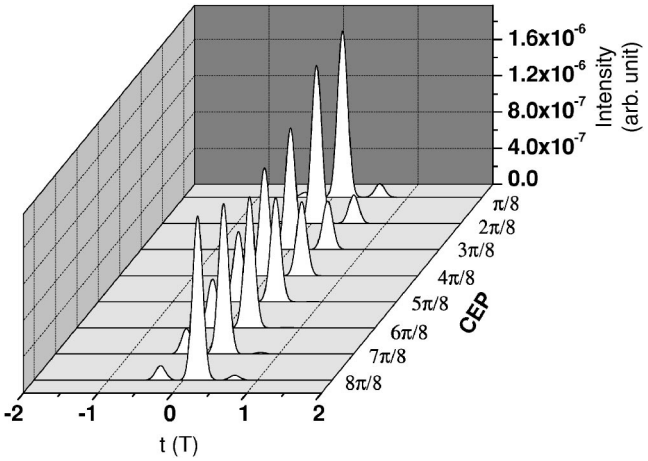

FIG. 3. The attosecond pulse profiles change with the CEP of the synthesized pulse. It is similar to the result of single driving laser pulse.

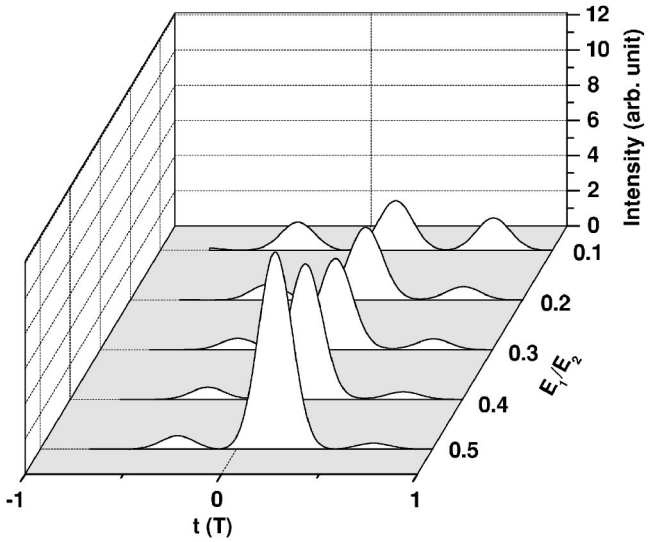

FIG. 4. The calculated temporal profiles of the attosecond pulses as functions of the electric-field ratio of the 5-fs pulse to the 50-fs pulse in the synthesized pulse. E_1, the amplitude of the electric field of the 5-fs pulse, is changed, while E_2, the amplitude of the electric field of the 50-fs pulse, was kept constant as 0.1 a.u. Other parameters in the calculation are the same as those for Fig. 1.

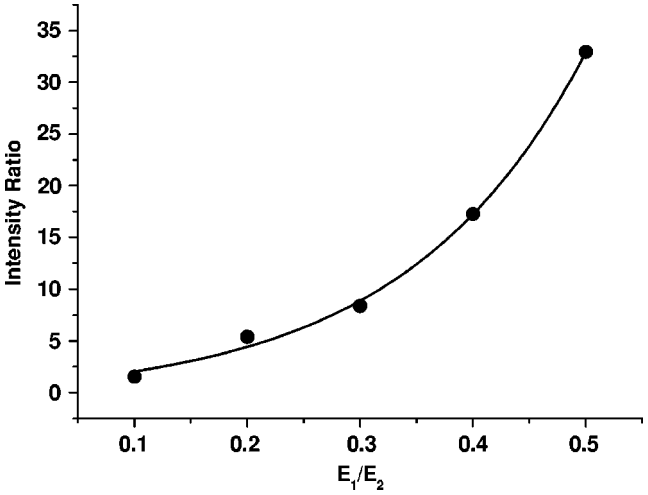

FIG. 5. The intensity ratio of the central attosecond x-ray pulse to its most adjacent attosecond x-ray pulses as the function of the electric-field ratio (E_1/E_2). When $E_1/E_2=0.3$, the attosecond pulse intensity ratio is about 8.4 which suggests the generation of a single attosecond x-ray pulse. In the calculation, E_2 is kept constant as 0.1 a.u. (corresponding to $I_2=3.51\times10^{14}$ W/cm^2) and E_1 is changed.

From Fig. 4 one can see that, when the ratio $E_1/E_2=0.1$, there is an attosecond pulse train generated as in the case of single 50-fs driving laser pulse. With the increase in the ratio, i.e., the increase in the intensity of 5-fs pulse, the intensity of the central x-ray pulse becomes higher and higher, while the pulses in the wing become weaker and weaker, and a single attosecond pulse appears gradually. Figure 5 shows the intensity ratio of the central attosecond x-ray pulse to its most adjacent attosecond x-ray pulses as the function of the driving laser field ratio (E_1/E_2). When $E_1/E_2=0.3$, the attosecond pulse intensity ratio is about 8.4 which suggests the

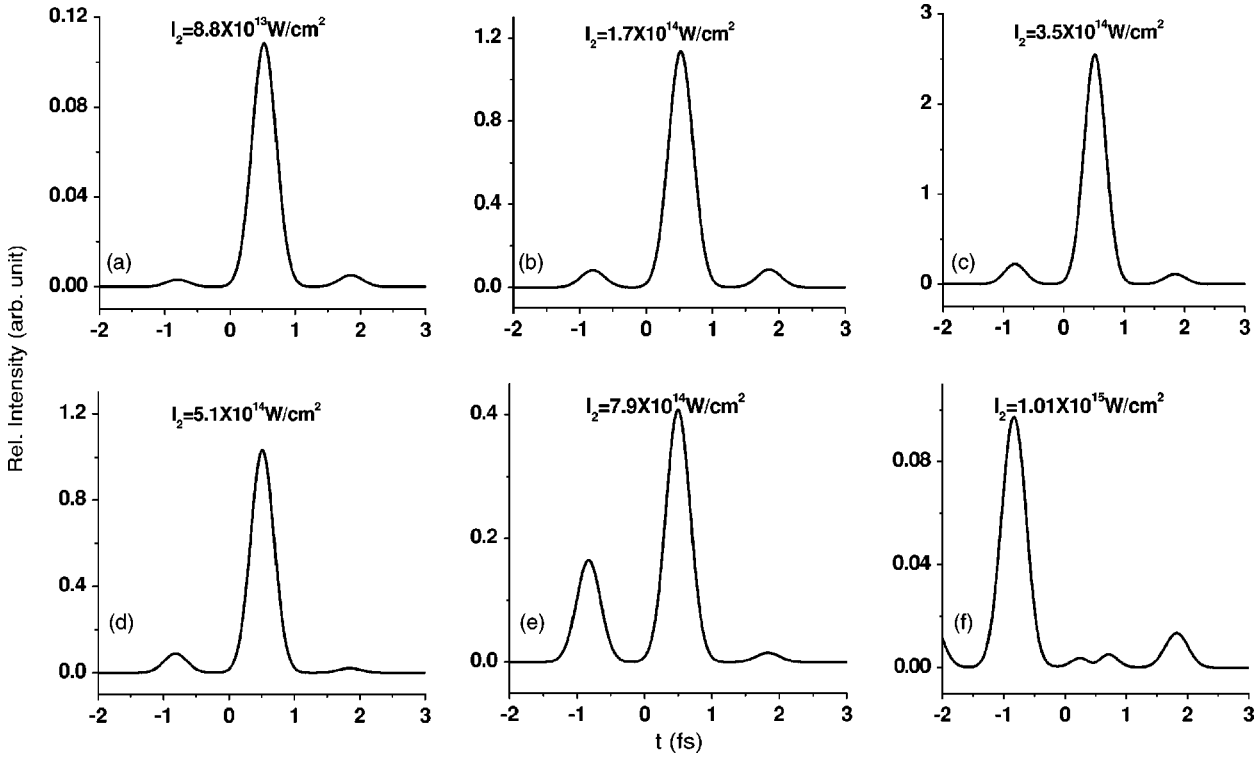

FIG. 6. The attosecond pulse profiles change with different 50-fs driving pulse. From (a) to (c), the intensity of the attosecond pulse increases by about 25 times while the intensity of the 50-fs pulse increases by only four times. But when the intensity of 50-fs pulse increases again, the intensity of the attosecond pulse begins to decrease because the ionization rate becomes very large gradually from (d) to (f).

generation of a single attosecond x-ray pulse. We have also investigated the above-mentioned phenomena with the Lewenstein model [18]. We obtained the similar results that the attosecond pulse intensity ratio is about 5 and 11 when E_1/E_2 equals to 0.4 and 0.5. Based on these results we can conclude that the synthesized laser field has the ability to work as a single few-cycle laser pulse and produce a single attosecond pulse. Even when E_1/E_2 is 0.5, the synthesized laser pulse can work as a few-cycle laser pulse with an increase in pulse energy by about one order of magnitude.

In the synthesized pulse scheme, most of the energy of the synthesized pulse comes from the 50-fs laser pulse. So we also investigated the effect of the intensity of the 50-fs pulse and the calculated temporal profiles of the attosecond pulses are shown in Fig. 6. In the calculation, we changed I_2, the intensity of the 50-fs pulse, while I_1, the intensity of the 5 -fs pulse, was kept constant as 8.8×10^{13} W/cm^2 (corresponding to the amplitude of the electric field $E_1=0.05$ a.u.). Other parameters in the calculation are the same as those for Fig. 1. From Fig. 6 one can see that, when the peak intensity of the 50-fs pulse changes from 8.8×10^{13} W/cm^2 to 3.5×10^{14} W/cm^2 (corresponding to the amplitude of the electric field E_2 change from 0.05 to 0.1 a.u.), a single attosecond pulse is always generated. This is absolutely different from the case of HHG driven by a long laser pulse where only a train of attosecond pulses can be produced and the importance of the weak 5-fs pulse is confirmed. Also, while the intensity of the 50-fs pulse changes from 8.8×10^{13} W/cm^2 to 3.5×10^{14} W/cm^2 by a factor of 4, the intensity of the attosecond pulse increases by about 25 times from Figs. 6(a)–6(c). But if the intensity of the 50-fs pulse increases further, most of the atoms will be ionized and the intensity of the attosecond pulse will be reduced greatly [from Figs. 6(d)–6(f)]. These results show that there is an optimum value for I_2 to generate a single attosecond pulse. For a specified few-cycle laser pulse, the energy of the long pulse has an optimum value.

In Ref. [12] Bartels *et al.* obtained an optimum 27th harmonic with a shaped pulse and the results can be explained by "intra-atomic phase-matching." The interference between emissions from different electron trajectories can induce the enhancement in a special order harmonic emisssion. For the generation of an attosecond pulse, the emission should be concentrated into a very short time period. As shown in our previous work [13,14], a train of attosecond pulse is always generated for any laser pulse. Only when the laser pulse is short enough, the intensity ratio of the adjacent attosecond pulses will be large enough and a single attosecond pulse can be generated. In the synthesized pulse scheme, the 5-fs laser pulse works as a very fast temporal switch for producing a single attosecond pulse. The enhancement of the attosecond pulse comes from two parts. One is as shown in Fig. 1, which can be explained by the Corkum model. Figure 7 shows the electric-field curves of the single 5-fs laser pulse

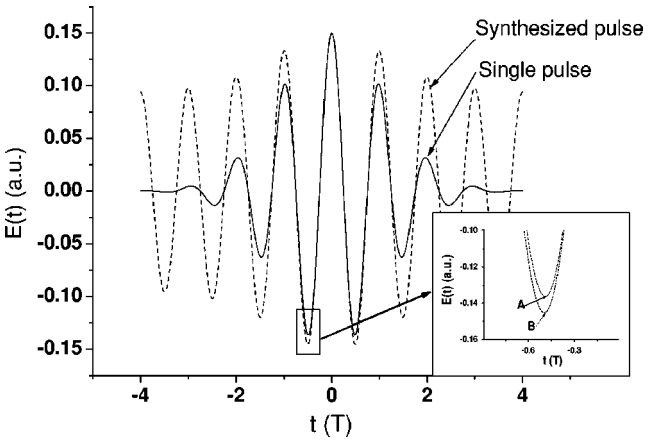

FIG. 7. The electric-field curves of the single 5-fs laser pulse (solid line) and the synthesized (50 fs+5 fs) pulse (dashed line) used in obtaining the results shown in Fig. 1.

(solid line) and the synthesized (50 fs+5-fs) pulse (dashed line) used in Fig. 1. According to Corkum's model of HHG [19], before the attosecond x-ray pulses are generated, the electrons will be ionized at position A (solid line, single laser pulse) and B (dashed line, synthesized pulse scheme). The electric field at position B is a little bit stronger than that at position A. Therefore in the synthesized pulse scheme, more electrons will be ionized. Then after the free motion in the laser field, more electrons will recombine with the atom to generate the attosecond x-ray pulse. This is the reason why the intensity of the attosecond x-ray pulse generated by the synthesized pulse scheme is higher than that generated by a single laser pulse as shown in Fig. 1, although the peak electric fields of the two cases are the same. Another reason is as shown in Fig. 6. There is an optimum intensity for the atom to generate the attosecond pulse. For a specified few-cycle pulse, the synthesized pulse can work as a few-cycle laser pulse whose energy is increased by about one order of magnitude. This induces the enhancement in the attosecond pulse intensity.

In conclusion, we have proposed a different scheme to generate high-intensity attosecond high-order harmonic by using a synthesized laser field. The synthesized laser field is obtained by an appropriate superposition of a few-cycle laser pulse and a relatively long pulse of several tens of femtoseconds whose energy can be increased to the level of several millijoules. With the synthesized pulse, the intensity of the attosecond pulse can be increased by about 25 times.

This work was supported by the National Natural Science Foundation of China (Grant Nos. 69925513 and 19974058), the Chinese National Major Basic Research Project (Grant No. G1999075204), the Chinese Academy of Sciences, and the Major Basic Research Project of Shanghai Commission of Science and Technology.

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