

Selective Enhancement of a Single Harmonic Emission in a Driving Laser Field with Subcycle Waveform Control

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We experimentally demonstrate a robust scheme to select a single high-order harmonic among the harmonic comb by using a driving laser field with subcycle waveform control, which is synthesized by the fundamental 800 nm laser pulse and two controlling laser pulses at 400 and 267 nm with perpendicular polarizations. By controlling the relative phase among the pulses of different colors, a single high-order harmonic is selectively enhanced while the adjacent harmonics are greatly suppressed with the intensity contrast increased by more than 1 order of magnitude and the peak intensity enhanced simultaneously by more than 2 orders of magnitude compared to the case by using only the fundamental 800 nm laser pulse. Such phenomena can be mainly attributed to the intra-atomic phase matching realized with the sub-cycle waveform controlled field.

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High-order harmonic generation (HHG) [1–9] in gases has been intensively explored in the past decade. The high-order harmonic source usually emits in the form of extreme ultraviolet (XUV) frequency comb in the plateau region with different harmonics at similar intensities, due to the nonperturbative nature of the HHG process. On one hand, the harmonic emission in the form of broadband supercontinuum is desired for the isolated attosecond pulse [2–9] by using a few-cycle or a multicolor laser field. For example, Bandulet *et al.* [9] reported a gating technique to produce an isolated attosecond pulse by combining the 800 nm pulse with other two noncommensurate infrared pulses. On the other hand, the selective generation of a single high-order harmonic emission from the harmonic comb as an intense monochromatic coherent source is also very important for many applications, such as the seeding of an XUV free electron laser for obtaining fully coherent output [10]. The intra-atomic phase matching scheme was proposed [11,12] to select a single harmonic emission from the harmonic comb. Although the intensity of the target harmonic (27th) was maximized with 8 times enhancement, the adjacent harmonic peaks were not suppressed efficiently. The intensity contrast ratio between the target harmonic and the adjacent harmonics was enhanced only by a factor of 4, indicating that the adjacent harmonics were also enhanced simultaneously.

In this Letter, we report a robust scheme to control the intra-atomic phase matching by using a driving laser field with sub-cycle waveform control. The laser field is synthesized with the fundamental, second and third harmonics of an 800 nm multicycle Ti:sapphire laser pulse. The required optical waveform for the successful intra-atomic phase matching is obtained by shaping (both in amplitude and

in polarization) the laser field on the subcycle time scale through simply adjusting the relative time delay among the laser pulses of different colors and polarizations. We experimentally demonstrate that a specific harmonic is selectively enhanced while the adjacent harmonics are dramatically suppressed, with both the harmonic intensity and the selectivity (the intensity contrast ratio between the target harmonic and the adjacent harmonics) significantly improved, for the first time to our knowledge.

The schematic of the experimental setup is shown in Fig. 1(a). A commercial Ti:sapphire femtosecond laser (Coherent, Inc.) is used to produce 2 mJ laser pulses at 800 nm center wavelength with 45 fs pulse duration at a repetition rate of 1 kHz. The output pulses are directed into the vacuum chamber for HHG. In the vacuum chamber, the laser beam is collimated and passes through sequentially a focusing lens (with a focal length of 500 mm), a SH-BBO crystal (β -barium borate crystal, $\theta = 29.2^\circ$, $\varphi = 0^\circ$,

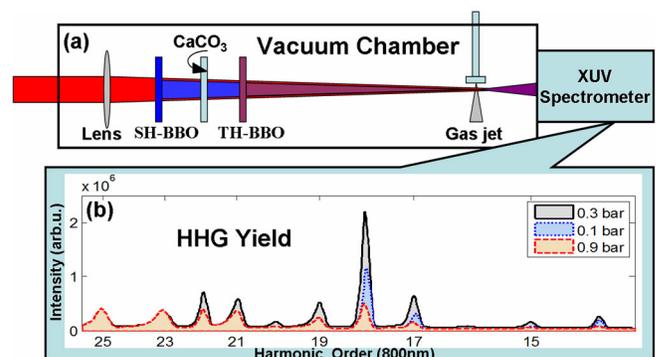


FIG. 1 (color online). (a) Schematic of the experimental setup. (b) Harmonic spectra obtained with different gas pressures.

0.3 mm thickness, type I phase matching, used for the second harmonic generation, the polarization of the generated 400 nm pulse is perpendicular to that of the fundamental 800 nm pulse), a CaCO_3 crystal ($\theta = 22.6^\circ$, $\varphi = 0^\circ$, 0.4 mm thickness, used for controlling the time delay between the 400 and 800 nm pulses), and a TH-BBO crystal (β -barium borate crystal, $\theta = 55.5^\circ$, $\varphi = 30^\circ$, 0.1 mm thickness, type II phase matching, for the third harmonic generation, the polarization of the generated 267 nm pulse is parallel to that of the fundamental 800 nm pulse), and is then focused into a continuous argon gas jet emitted from a nozzle with 0.2 mm inner diameter. The measured energy ratio among the 800, 400, and 267 nm pulses is about 100:14:5, with a total laser intensity of $2.0 \times 10^{14} \text{ W/cm}^2$. The generated high-order harmonics are detected by a homemade flat-field grating spectrometer equipped with a soft-x-ray CCD (Princeton Instruments, SX 400). A 500 nm thick aluminum foil is used in the spectrometer to block the driving laser.

Using the above-mentioned three-color laser field, we first optimize the high-order harmonic signal by adjusting the argon gas pressure. The gas pressure is adjusted from 0.1 to 1.0 bar, and the measured harmonic yield is first increased and then decreased. The optimum stagnation pressure of argon gas is about 0.3 bar for obtaining the maximum harmonic yield, which is shown in Fig. 1(b). One can see that several harmonics, especially the 18th harmonic, are selectively enhanced when the gas pressure is optimized. We also measure the HHG with the lower intensity of the 400 and 267 nm pulses through tilting the BBO crystal to the phase-mismatching position, and find that the harmonic yield is decreased and the intensity of even-order harmonics are reduced particularly.

The two-color scheme has been proved to be an effective way to enhance the harmonic yield by about 2 orders of magnitude [13]. For comparison, we measure the harmonic yields by using the traditional two-color scheme with the orthogonally polarized 800 – 400 nm pulses (just remove the TH-BBO crystal, the measured energy ratio between 800 and 400 nm pulses is about 100:17). The measured harmonic yields are shown in Fig. 2(a) as a function of the time delay between the two-color pulses. The measured harmonic yields by using the above three-color scheme are also shown in Fig. 2(b) as a function of the time delay between the 800 and 400 nm fields. The modulation period of the generated harmonics in the two-color case is 1/4 cycle (the optical cycle of 800 nm pulse, the same hereafter), which is determined by the evolution period of the phase difference between the driving laser pulse at 800 nm and the controlling laser pulse at 400 nm. In the three-color case, the harmonic intensity is modulated with a period of about one cycle under the experimental condition since the time delay of the 267 nm pulse is correlated to the time delay between the 800 and 400 nm pulses.

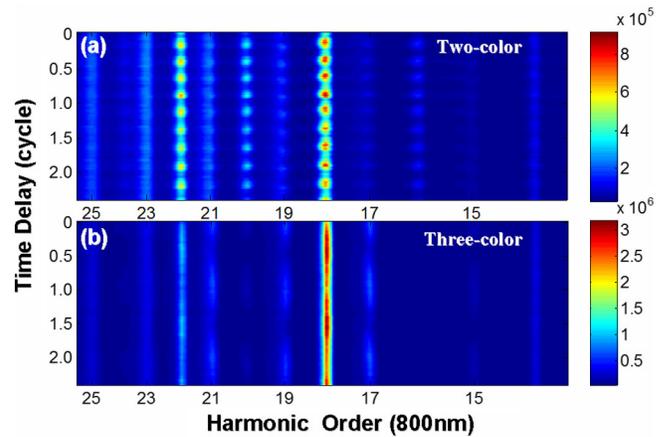


FIG. 2 (color online). Harmonic spectra driven by (a) the two-color laser field and (b) the three-color laser field as a function of the time delay, respectively.

For a more detailed comparison, we extract the maximum harmonic intensities at the optimum time delay from Fig. 2(a) and 2(b), which are shown in Fig. 3(a), together with the harmonic spectrum obtained by using only the fundamental 800 nm pulse. Compared to the harmonic generation in the fundamental 800 nm pulse, the addition of the 400 nm pulse of perpendicular polarization can enhance the conversion efficiency by about 2 orders of magnitude [13], and the further addition of a weak

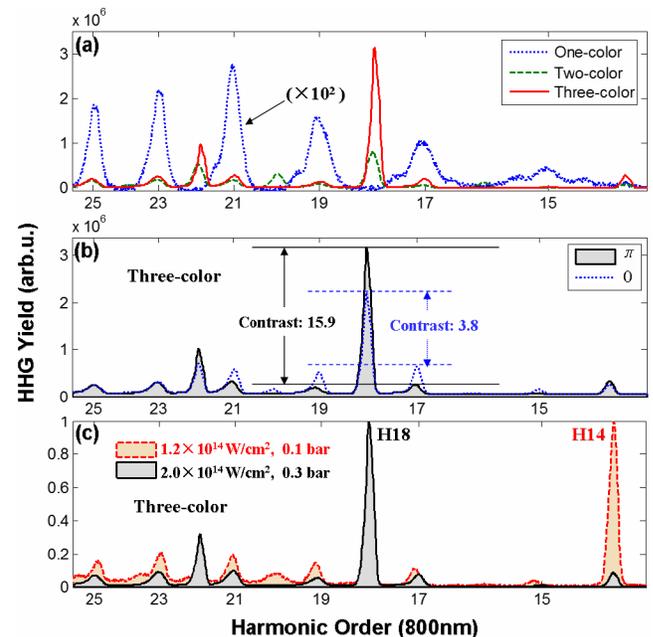


FIG. 3 (color online). (a) Comparison of the maximum harmonic yields driven by different laser fields. (b) Comparison of the harmonic yields driven by the three-color laser field with two different relative phases. (c) Comparison of the harmonic yields driven by the three-color laser field with different intensity and density conditions.

267 nm pulse of parallel polarization further enhances the conversion efficiency by about 5 times. Moreover, compared to the traditional two-color scheme, the addition of the weak 267 nm pulse can greatly enhance the intensity contrast ratio between the 18th harmonic and the adjacent harmonics by over 1 order of magnitude.

In order to identify the role of the relative phase delay between different colors in the three-color scheme, we extract the harmonic intensities from Fig. 2(b) at the time delays of 0 and 0.5 cycle (i.e., the relative phase of 0 and π), which are shown in Fig. 3(b). By optimizing the relative phase, the 18th harmonic intensity is enhanced by 43.7%, while the intensities of the adjacent harmonics are at least suppressed by 65.4%. Then, the intensity contrast ratio between the 18th harmonic and the adjacent harmonics is improved from 3.8 to 15.9. Moreover, the specific harmonic emission can be tuned by controlling the laser intensity and the phase-matching parameters. We realize the selective enhancement of the 14th harmonic at the laser intensity of 1.2×10^{14} W/cm² and the gas pressure of 0.1 bar, as shown in Fig. 3(c) together with the selective enhancement of the 18th harmonic at the laser intensity of 2.0×10^{14} W/cm² and the gas pressure of 0.3 bar.

The selective enhancement of the harmonic emission can be attributed to the phase matching effects. The optimization of phase matching [14] includes both the macroscopic and intra-atomic phase matching. For the macroscopic phase matching, the effective spectral range and the relative weight of the individual harmonic order strongly depend on the species of gas and its pressure, the interaction geometry, the intensity and diameter of the laser beam, and, etc. By properly choosing these parameters, the harmonic emission can be phase matched and confined to a few orders [15]. For the intra-atomic phase matching, the emission of different trajectories in each half-cycle should be constructively interfered [12]. For the driving laser pulse

with a finite duration, the pulse envelope slightly tailors the electric field of each half-cycle, leading to the phase difference of the harmonic emissions from each half-cycle. For an 800 nm laser pulse with the intensity of 10^{14} W/cm², the phase of the harmonic emission is several tens of radian and a small change of the electric field will affect the phase of the harmonic greatly.

In the multicolor laser field, the phase of the harmonic emission can be affected by many parameters, and it is difficult to optimize the electric field with time-dependent Schrödinger equation (TDSE) method. So we use the classical motion of the electron for each trajectory to calculate the phase of the harmonic emission [16] and to optimize the electric field to achieve the intra-atomic phase matching. In the simulation, the laser intensity and the intensity ratios among the laser pulses of different colors are the same as the experiment. The phases of different trajectories are optimized by controlling the relative delays and the linear chirps of the multicolor laser field. As shown in Fig. 4(a), when the fundamental 800 nm pulse is used to drive the HHG, the phase of each trajectory of the 18th harmonic will change significantly even for a multicycle laser pulse (the blue dotted line). When the 400 nm pulse is added, the phase difference of the trajectories from -4 to 4 cycle can be decreased greatly (the red dashed line). When the 267 nm pulse is added, the phase difference of almost all the trajectories from -3 to 2.5 cycles can be decreased to be about or less than one π , which means a good intra-atomic phase matching (shown as the black solid line in the lower panel, together with those for the adjacent 16th and 17th harmonics. Those for the adjacent 19th and 20th harmonics are similar). One can also see that phase difference for the adjacent harmonics is close to or larger than 2π and thus the phase mismatching occurs. Although the simulation is simple, it is demonstrated that the intra-atomic phase matching can be achieved by using the three-color scheme.

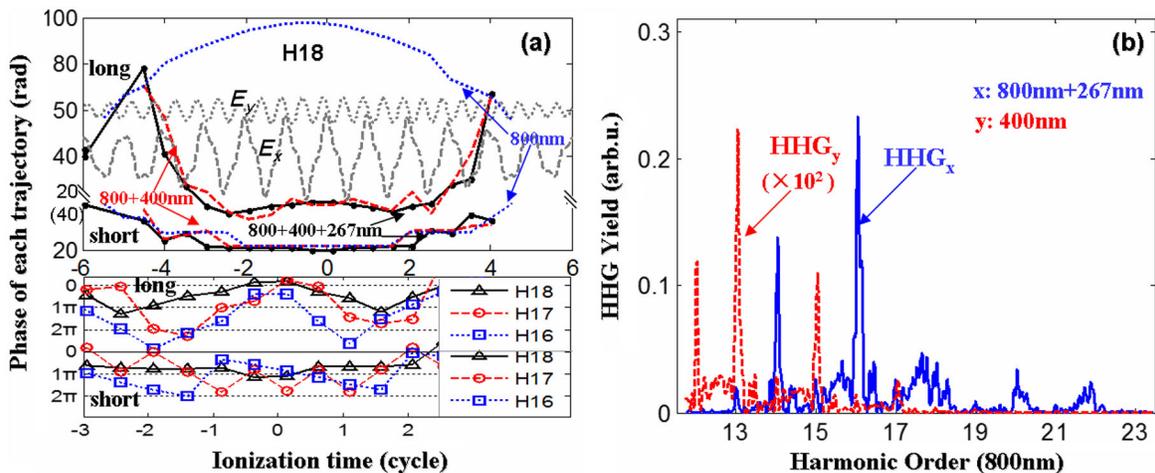


FIG. 4 (color online). (a) The phase of the 18th harmonic in different laser fields (upper panel) and the comparison with the adjacent 16th and 17th harmonics (lower panel). (b) The harmonic spectra in two polarization directions (HHG_x, HHG_y).

For the intra-atomic phase matching by using only one-color laser field [11,12], it seems that the adjacent harmonics will be slightly enhanced when the target harmonic is optimized, leading to the low intensity contrast. However, in the three-color laser field, the adjacent harmonics (especially the odd-order harmonics) are greatly suppressed when the target harmonic intensity is maximized. In order to understand the underlying physics, we further simulate the harmonic emission by using the simplified Lewenstein model [1] with the three-color laser field. In the simulation, the intensity ratios and the relative delays between the different colors including the chirps of them are considered. The optimized result is shown in Fig. 4(b). One can see that only the 14th and 16th harmonics dominate in the spectra while almost all the odd-order harmonics are dramatically suppressed. Although it is a little different with the result of our experiment (where only a single harmonic is selected) and it is strange that the polarization direction of the 14th and 16th harmonics is parallel to that of the 800 and 267 nm pulses while the even-order harmonics can only be generated in the polarization direction of 400 nm pulse in the two-color scheme [17], it is demonstrated that an isolated harmonic or two can be generated while the adjacent harmonics can be suppressed.

Shaping the laser field waveform by using the combination of multi-color laser pulses [18,19] can lead to not only a broadened XUV supercontinuum for isolated attosecond pulse generation but also the confinement of the harmonic emission in a small spectral region for narrow-bandwidth XUV radiation [20] and even a single-order harmonic emission as a monochromatic XUV source in the present work. In our waveform shaping with three-color laser field, the fundamental 800 nm laser pulse is the main driving laser field for the HHG, the two controlling pulses of different colors with perpendicular polarization help to improve the harmonic yield and spectral selectivity simultaneously. The above three-color combination is the basic configuration for controlling the electron trajectory in both the perpendicular and parallel directions. Although a finer control is possible by using the laser field of more than three colors, the improvement would be very limited and it is difficult to implement experimentally. As we know, the ability of the waveform shaping with multicolor field is dependent on the spectrum-spanning range. Such three-color laser field spans from 800 to 267 nm, and the addition of the fourth harmonic will extend only from 267 to 200 nm.

Therefore, the improvement in intensity contrast can be mainly attributed to the intra-atomic phase matching realized with the sub-cycle waveform controlled driving laser field consisting of a main driving laser pulse and two controlling laser pulses of perpendicular polarizations, with a total spectrum-spanning range of 1.6 octaves from 267 to 800 nm. In fact, the maximum intensity contrast

ratio of 15.9 is obtained in the three-color scheme, which is much better than the optimized results obtained by the pulse-shaping technique [11]. These results can be understood as follows: First, the harmonic emission is selectively enhanced and confined to only a few harmonic orders by controlling the macroscopic phase matching conditions. Second, the target harmonic (18th) is further enhanced and the adjacent harmonics (16th, 17th, 19th, and 20th) are greatly suppressed by controlling the intra-atomic phase matching conditions. The photon number of the 18th harmonic in each shot is estimated using the method [21] to be larger than 9.2×10^6 . A conversion efficiency of about 1.0×10^{-6} is expected if the loss of spectrometer slit is taken into account.

In summary, we have experimentally demonstrated the generation of intense single high-order harmonic emission by using the multicolor laser field with subcycle waveform control, which is consisting of the fundamental laser pulse and two controlling laser pulses of different polarizations. We observe that a single high-order harmonic is selectively enhanced while the adjacent harmonics are dramatically suppressed, with both the peak intensity and selectivity of the target harmonic significantly improved. Moreover, the single-order harmonic source can be tuned by controlling the driving laser intensity and the phase matching parameters. This scheme is promising for the development of compact monochromatic coherent XUV sources.

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