We theoretically investigate the high-order harmonic generation and isolated attosecond pulse generation from a two-dimensional model of an Ar$^+$ cluster in a synthesized two-color laser pulse. A scheme to generate an isolated attosecond pulse is proposed by using a 4-fs 800-nm fundamental field in combination with a wavelength-adjustable weaker pulse. When the number of ions surrounding the central atom is 24 and the wavelength-adjustable weaker pulse is chosen to be 1200 nm with the intensity $1 \times 10^{14}$ W/cm$^2$, an almost linearly polarized isolated attosecond pulse with the duration of 45 as is obtained by superposing a bandwidth of 100 eV near the cutoff of high-order harmonic generation. Moreover, we illustrate the interference effect of the paths in terms of the time-frequency analysis of high-order harmonic generation.

DOI: 10.1103/PhysRevA.85.025802  PACS number(s): 42.65.Ky, 42.65.Re, 52.50.Jm

Recently, much effort has gone into high-order harmonic generation (HHG) due to its wide application as a coherent soft x ray [1] and the generation of isolated attosecond pulses [2–5]. In particular, the isolated 80-as pulse of extreme ultraviolet light was obtained by recent experiment [6]. An isolated attosecond pulse is an effective tool for probing the ultrafast dynamics of atoms and molecules [7–9], such as probing the electron motions inside atoms and molecules, above-threshold ionization, and so on. It has been shown that, by superposing continuous harmonics in the plateau near the cutoff region, an isolated attosecond pulse can be generated using driving pulses with few cycles [10], the polarization gating method [11], an orthogonally polarized two-color laser combined with a static electric field [12], and so on. It has been mentioned that a high-signal-to-noise isolated attosecond pulse has been generated by using a few-cycle pulse combined with its second-harmonic field with a proper polarization angle [13]. In general, most of the HHG schemes presented above are used by rare-gas targets, and the conversion efficiency is not so high. Although solid targets can produce photons with energies up to the MeV range [14], amounts of long-lived (nanoseconds) strong x rays are produced because of the hot solid dense plasma.

In order to improve the efficiency of HHG, the neutral clusters have been introduced as a type of efficient medium both experimentally and theoretically [15–17]. Superthermal (keV) electrons and highly charged ions from interactions of intense laser pulses with clusters were generated for a large atom cluster [18,19]; the harmonic conversion efficiency is enhanced and the harmonic plateau is significantly extended from ionized clusters [20]. The ionization probability, which is important for HHG, is enhanced when the electron is in a multiwell system [21]. As is known to all, the ionization process is the first step of Corkum’s semiclassical three-step model [22]. It was proposed in Ref. [23] that there are two different ways to contribute to HHG in clusters: one way is that the HHG is generated by the recombination of the freed electron with its parent ion; another way is that the HHG is generated by the recombination of the freed electron with any neighboring ion.

In this Brief Report, we study the HHG and isolated attosecond pulse generation from a two-dimensional model of an Ar$^+$ cluster based on our previous works, e.g., a one-dimensional model of the He$^+$ ion [24,25] and a two-dimensional model of the He atom [12]. In our method, a few-cycle 4-fs 800-nm fundamental pulse in combination with a weaker assistant pulse is adopted. By adjusting the parameters of the laser field and the number of ions surrounding the central atom, an almost linearly polarized isolated attosecond pulse with a duration of 45 as is obtained. We also investigate the time-frequency distribution and illustrate the interference effect of the paths.

We consider a two-dimensional model of an Ar$^+$ cluster [20,21], which assumes that a sole atom is located at the center of a square grid and multiple singly charged positive ions are distributed on the crossing points of the square grid. The two-dimensional time-dependent Schrödinger equation (in atomic units) is

$$i \frac{\partial \Psi(x,y,t)}{\partial t} = \left[ \frac{\hbar^2}{2m} + V(x,y) + xE_x(t) + yE_y(t) \right] \Psi(x,y,t),$$

where $\Psi(x,y,t)$ is the two-dimensional time-dependent wave function and $V(x,y)$ is the multwell potential used to describe the model of the Ar$^+$ cluster [20,21,26], which can be expressed as

$$V(x,y) = - \sum_{i=-N}^{N} \sum_{j=-N}^{N} \frac{1}{\sqrt{(x+ir)^2 + (y+jr)^2 + 0.62^2}}.$$
cycle of the fundamental field that corresponds to an 800-nm laser pulse; \( \varphi_1 = \pi/6, \varphi_2 = 0.3\pi \) are the carrier-envelope phases, and \( \theta = \pi/6 \) is the complementary angle of the polarization angle between the two pulses. \( E_{10} \) and \( \omega_1 \) are the amplitude and frequency of the fundamental field, and \( E_{20} \) and \( \omega_2 \) are the amplitude and frequency of the wavelength-adjustable weaker pulse, respectively.

We solve numerically the Schrödinger equation by using the second-order splitting-operator fast Fourier transform algorithm [27] by choosing \( \omega_2 = 0.114 \text{ a.u.} \), which corresponds to \( \lambda_2 = 400 \text{ nm} \), and \( I_1 = 3 \times 10^{14} \text{ W/cm}^2, \ I_2 = 1.5 \times 10^{13} \text{ W/cm}^2 \). By summing up the intensities of both \( x \) and \( y \) components, we can obtain the harmonic spectrum of \( N_x = 8 \) and \( N_y = 24 \), which is shown in Fig. 1(a). We can see from Fig. 1(a) that when the number \( N_x \) of the surrounding the central atom is 8, an obviously harmonic plateau is observed, and the cutoff is about 140 eV. As the number \( N_x \) is increased to 24, the cutoff is broadened to 200 eV, and the intensity is also enhanced. Due to the fact that with the increase in the number of ions surrounding the central atom [21] the binding energy of the Ar\(^{+} \) cluster is increased and the ionization probability is also enhanced, the plateau is broadened and the intensity is enhanced.

To explain the HHG spectrum shown in Fig. 1(a), we investigate the emission time of harmonics in terms of the time-frequency analysis method [28]. We take the \( y \) component as an example to explore the physical mechanism of the HHG. Figure 1(c) presents the time-frequency distributions of HHG for the case of \( N_y = 8 \). As shown in Fig. 1(c), there are three photon-energy peaks and the photon energy is smaller and the intensity is weaker for peaks \( A_1 \) and \( C_1 \), which indicate the harmonic plateau is contributed mainly by the peak \( B_1 \), and the maximal energy is 140 eV, which is agreement with the cutoff shown in Fig. 1(a). In detail, several paths on the peaks can be seen, which is because the ionization electron in the Ar\(^{+} \) cluster can be recombined with several nuclei. For the case of \( N_y = 24 \) (not shown here), there are more paths for the peak than for peak \( B_1 \). According to Ref. [23], the freed electron may recombine to the nucleus around the center argon atom. The larger the number \( N_y \) is, the easier the freed electron recombines with a parent or any neighboring ion. So the interference in the case of \( N_y = 24 \) is stronger than that in the case of \( N_y = 8 \). We investigate the attosecond pulse generation by superposing a bandwidth of 45 eV near the cutoff of the HHG plateau area [12]. An attosecond chain is obtained for the case of \( N_y = 8 \), and for the case of \( N_y = 24 \), two satellite attosecond bursts with strong intensity are generated with an attosecond pulse. The figures are not shown here for simplicity.

We discuss the case of \( N_y = 24 \) in the following, which obtains a broadened supercontinuum harmonic plateau and is beneficial in generating an isolated attosecond pulse. We fix other parameters as those in Fig. 1(a) and just increase the wavelength of the weaker laser pulse. The cutoff is extended to 220 eV for the case of \( \lambda_2 = 1200 \text{ nm} \) as shown in Fig. 1(b) (solid black line), and the plateau is smooth and continuous. As \( \lambda_2 \) increased to 1600 and 2000 nm, the cutoff is extended to 225 and 235 eV, as shown by the dashed (green) line and dotted (red) line, respectively, but the HHG plateau is not so smooth compared to the case of \( \lambda_2 = 1200 \text{ nm} \); some modulations appear. In a word, the HHG plateau is broadened but modulated as the wavelength of the weaker pulse increases. Figure 1(d) shows the time-frequency distributions of HHG in the case of \( \lambda_2 = 1200 \text{ nm} \). To compare with Fig. 1(c), the harmonic plateau is also contributed mainly by the middle peak \( B_1 \) but it is broader and stronger. For the cases of \( \lambda_2 = 1600 \text{ nm} \) and \( \lambda_2 = 2000 \text{ nm} \), which are not shown here for simplicity, the phenomena are similar except that more paths are obtained in the time-frequency distribution. That indicates the interference for the case of \( \lambda_2 = 1200 \text{ nm} \) is weaker than in the other two cases.

In addition, we also investigate the attosecond pulse generation by superposing the bandwidth of 45 eV (175–220 eV) near the cutoff for the case of \( \lambda_2 = 1200 \text{ nm} \), and an isolated attosecond pulse with duration of 90 as is obtained as shown in Fig. 2(a). When we superpose a longer bandwidth of 50 eV
(170–220 eV), a main attosecond pulse with a duration of 103 as accompanied by a stronger satellite attosecond burst is generated, which is shown in Fig. 2(b). For the cases of \( \lambda_2 = 1600 \text{ nm} \) and \( \lambda_2 = 2000 \text{ nm} \), an attosecond pulse with several stronger satellite bursts is obtained by superposing a bandwidth of 45 eV as shown in Figs. 2(c) and 2(d). A longer duration is obtained when we filter a broader bandwidth and the stronger satellite burst is produced, all of which are because of the interference of the quantum paths. To clearly show the polarization characteristics of the attosecond pulses, we draw the three-dimensional (3D) electric fields of the attosecond pulses, which are shown in Fig. 3. Figure 3(a) shows the 3D electric field corresponding to Fig. 2(a) and indicates that the attosecond pulse is nearly linearly polarized and Fig. 3(b) shows the 3D electric field corresponding to Fig. 2(c) and indicates a chaotic pulse. The chaotic behavior is because the polarization directions of the two pulses are different.

To obtain a shorter isolated attosecond pulse, we investigate the HHG for the wavelength \( \lambda_2 = 1200 \text{ nm} \) of the weaker laser pulse and increase its intensity to \( 1 \times 10^{14} \text{ W/cm}^2 \) and \( 2 \times 10^{14} \text{ W/cm}^2 \); other parameters are the same as those in Fig. 1(b) (solid black line). Figure 4 presents the high-order harmonic spectra generated with different intensities of the weaker pulse. The harmonic plateau is supercontinuous and the bandwidth is broadened. The cutoff of the HHG is extended to 275 and 340 eV, respectively. By superposing a bandwidth of 80, 90, and 100 eV near the cutoff, we obtain isolated attosecond pulses with durations of about 55, 50, and 45 as, which are shown in Figs. 5(a), 5(b), and 5(c), respectively, for the case of \( I_2 = 1 \times 10^{14} \text{ W/cm}^2 \). The satellite attosecond bursts around the isolated attosecond pulse are too small to be ignored. All the attosecond pulses are nearly linearly polarized.

In conclusion, we theoretically investigate the HHG and the isolated attosecond pulse generation for different number of ions surrounding the central atom in a synthesized two-color laser pulse combined with a wavelength-adjustable weaker laser pulse. When the wavelength of the weaker laser field is
1200 nm with an intensity of $1 \times 10^{14}$ W/cm$^2$ and the number of ions surrounding the central atom is 24, a continuous plateau with a bandwidth of 100 eV could be obtained, and an isolated attosecond with a duration of about 45 as could be generated, which is almost linearly polarized. In addition, we also investigate the time-frequency distribution and illustrate the physical phenomenon in the quantum path interference.

This work was supported by the National Natural Science Foundation of China (Grants No. 11174108, No. 11104108, and No. 10974068), and the Graduate Innovation Fund of Jilin University (Grant No. 20111032). We also acknowledge the High Performance Computing Center (HPCC) of Jilin University for supercomputer time.