Control of the launch of attosecond pulses

Wei Cao, Peixiang Lu,* Pengfei Lan, Xinlin Wang, and Yuhua Li
Wuhan National Laboratory for Optoelectronics and School of Optoelectronics Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074, People's Republic of China
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We propose an approach to steer the launch of attosecond (as) pulses with a high precision. We numerically demonstrate that by adding a weak second-harmonic (SH) field to the fundamental beam the ionization and recollision process of the electron will be perturbed, which can induce a variation of the emission time of high harmonics. Through modifying the relative intensity of the SH and fundamental fields, the launch of as pulses can be manipulated with a resolution less than 40 as. This will show significant potential for ultrafast optics.

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High harmonic generation (HHG), which has been a hot topic at present due to its potential for the generation of XUV light source [1,2] and attosecond bursts [3–9], can be viewed as an interference radiation between the continuous and bound electron wave packets [10]. This process has been well-explained by the simple-man's model [11]. Electrons can be set free in the presence of a strong laser field; and then accelerated to gain additional kinetic energy; finally decelerated towards the parent core and recombine with it to emit a harmonic photon. The conservation of energy leads to the production of harmonic photons with a maximal energy of $3.17U_p+I_p$, where U_p, I_p correspond to the ponderamotive energy of the electron and the binding potential of atoms. Both the classical [11] and quantum mechanisms [12] have demonstrated the dominant electron trajectories contributing to HHG, i.e., the so-called short and long trajectories, characterized by electron travel times in the continuum of the order of one-half and one optical period, respectively. Such electron quantum paths can be applied to describe the properties of the HHG process, including information of the frequency domain, the time domain and the chirp rate of harmonic radiation. Recently, quantum path control has attracted lots of focus, and is significant for the manipulation of HHG. Many pulse generation techniques have been proposed based on HHG [13–16]. In the single atom response, both as pulse train [13] and two-color regime [14] can be applied to quantum path selection, resulting in the generation of regular as pulses. In the macroscopic case, phase-match conditions can be satisfied via propagation effects. Depending on whether the laser is focused before or after the atomic gas volume, phase matching favors contributions from either long or short trajectories, leading to a more regular spectrum of harmonic radiation after propagation [15,16]. Another important effect of propagation is that the short and long trajectories have different radial phase gradients, and therefore different divergence angles. This is used in experiment for synthesizing and controlling attosecond waveforms by adding a small aperture after the harmonic propagation cell [17]. These schemes mentioned above have indicated the significance of quantum path control in the attoscience and recollision science. In this work, we present a feasible method for controlling the launch time of HHG precisely, it is achieved by using a bichromatic field consisting of a fundamental and a second harmonic field. As an efficient pathway for controlling the electron dynamics, two-color scheme is widely used both theoretically and experimentally. The spectral and temporal structure of high harmonic from $\omega-2\omega$ field shows sensitive dependence on the relative phase of the laser field [18,19], then phase-stabilized as pulse train with one pulse per infrared cycle can be generated with a bichromatic field. Furthermore, by using a phase-stabilized bichromatic field with short duration, efficient isolated as XUV pulse with rather short wavelength can be obtained [20,21]. Recently, Dudovich *et al.* proposed a novel approach for measuring and controlling the birth of attosecond XUV pulses [22], by scanning the weak SH field (i.e., changing the phase difference between the bichromatic field), the total output can be transferred between the odd and even harmonics, then the complex wavefront of the as pulse can be fully characterized and controlled. In our work, by adjusting the relative intensity of the two components of the phase-stabilized driving field, the electric field evolution is modulated, then the released and recollision times of the electron, i.e., the electron quantum paths, are perturbed. Consequently, the launch of as pulses can be manipulated with a resolution less than 40 as. It shows another important application of quantum path control.

Figure 1 demonstrates the classical electron trajectories in a 5-fs bichromatic field. It illustrated the perturbation mechanism of electron quantum paths. In monochromatic case, the harmonics are emitted each half-cycle of the driving pulse. The symmetric structure of the electric field results in a period of $T_0/2$ for the electron recollision times, where T_0 represents the optical cycle. While in the bichromatic case, for a phase-stabilized driving field as shown in Fig. 1(a), the including of the SH component will modulate the evolution of the electric field. The asymmetric degree increases with the relative intensity. Due to the breaking of the symmetry of the laser field, the electron shows different dynamics in adjacent half optical cycles, and the interval of the recollision times between the adjacent half cycles varies as adjusting the relative intensity. In addition, the recollision times of electron within the same half cycle, i.e., the launch times of high harmonic, show dependence on the relative intensity of the bichromatic field. A small change of the relative intensity leads to a slight shift of the as pulse in the time domain. The trajectory analysis for various phase differences of the

*Corresponding author. lupeixiang@mail.hust.edu.cn

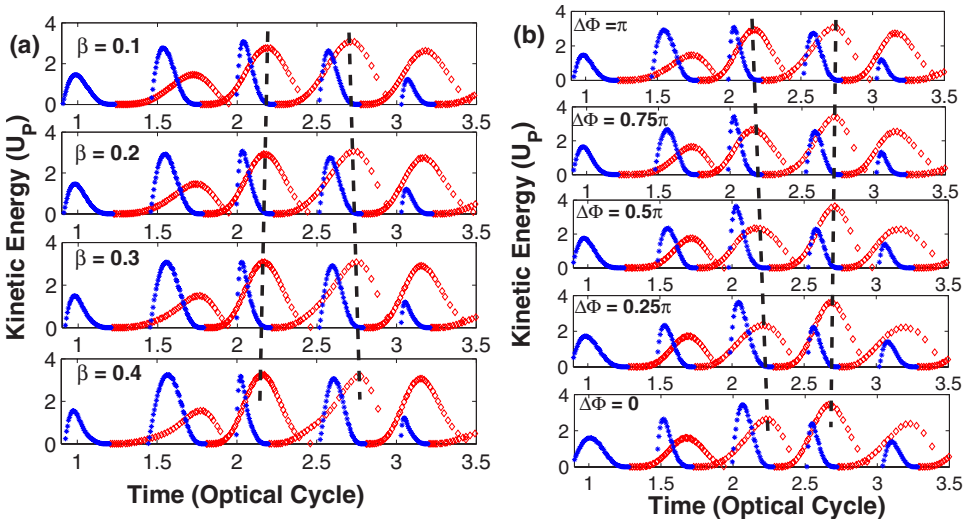

FIG. 1. (Color online) Classical ionization (blue stars) and recombination times (red diamonds) as a function of the return kinetic energy in unit of the ponderomotive potential U_p. The driving field is a 5-fs bichromatic field with the fundamental CEP fixed at π. (a) β denotes the relative intensity of the SH field with respect to the fundamental one, the CEP of the SH is 0. (b) $\Delta\phi$ denotes the phase difference between the bicromatic field, and the relative intensity between the SH and fundamental field is 0.2.

bichromatic field is also made, which is shown in Fig. 1(b). It is shown that the phase difference can also influence the time delay between the adjacent two pulses. Gradually increasing the phase difference results in a increasing of the time delay between the two pulses. Whereas, for a practical experiment, adjusting the laser intensity will be more feasible and easier than precisely changing the phase of the driving field.

Such a classical description provides qualitative and reliable information about the manipulation process of the quantum path control. To obtain a quantitative analysis, we numerically solve the time-dependent Schrödinger equation by means of a splitter-operator method [23]. The ground state is obtained by propagation in imaginary time. An absorption function to each dimension is applied in order to avoid the reflection of wave packets from the boundary. The harmonic spectra are calculated by Fourier transforming the time-dependent dipole acceleration, which is calculated from Ehrenfest's theorem [24]:

$$\ddot{\mathbf{d}} = \langle \Psi | - \frac{\partial \mathbf{V}(\mathbf{r})}{\partial r} + [E(t)] | \Psi \rangle. \tag{1}$$

The single atom response is considered in our simulation. In the analysis, we use the soft-core potential: $V=-1/\sqrt{\alpha+x^2}$, to describe medium with different binding potentials. The electric field in our calculation can be expressed in the form $E(t)=f(t)[E_0 \cos(\omega_0 t+\phi_0)+E_1 \cos(2\omega_0 t+\phi_1)]$, where $f(t)$ presents the profile of the laser field, ω_0 is the fundamental laser frequency, ϕ_0 and ϕ_1 correspond to the carrier envelope phases (CEP) of the fundamental and the SH field, respectively. E_0 and E_1 denote the amplitudes of the fundamental (800 nm) and the SH field (400 nm). In our simulation, the $\sin^2$ pulse profile is adopted with a full width at half maximum of 5 fs.

The harmonic spectra as well as the corresponding as pulses from a bichromatic field with different relative intensities are plotted in Fig. 2. The peak intensity of the fundamental field is 5×10^{14} W/cm^2. The CEPs of the fundamental and SH fields are π and 0, respectively. The atom is H by choosing the soft core parameter $\alpha=2$. The intensity of the driving pulse is above the saturation intensity. Then the ionization level near the peak of the laser field shows independence on the amplitude of the pulse [25]. As a result, the ionization step plays a slight role in the yield of harmonic radiation within different half cycles. Due to the nonadiabatic effect of the few-cycle driving pulse, harmonics in the cutoff region are contributed from electron trajectories within two adjacent half cycles. As a result, two pulses are generated in the cutoff region of the spectrum $(65-80\omega_0)$. The interference of the two pulses leads to a modulated structure in the spectrum, and the periodicity of the modulated structure shows evidence of the interval between the two pulses. The relationship between the time interval (τ) and the spectral periodicity $(\Delta\omega)$ can be expressed in the

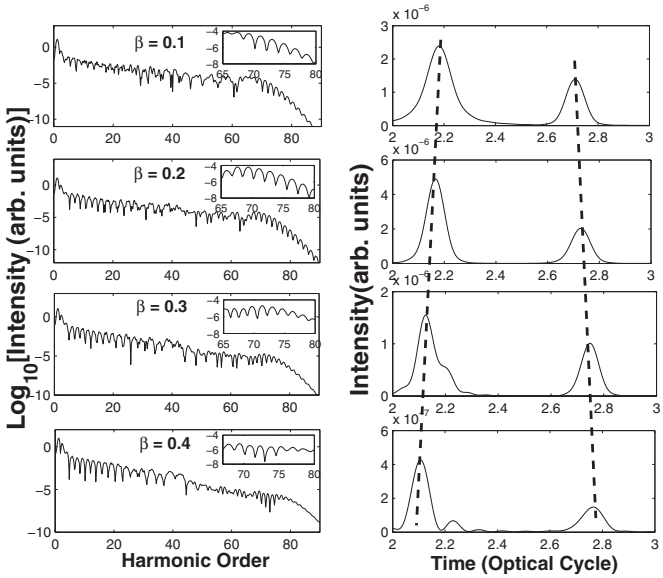

FIG. 2. Harmonic spectra of H atoms driven by bichromatic fields with different relative intensities (the left sets of the figures), and the corresponding as pulses generated in the cutoffs (the right sets of the figures). The peak intensity of the fundamental field is 5×10^{14} W/cm^2. The CEPs of the SH and the fundamental fields are 0 and π, respectively. The insets are amplified harmonic spectra in the cutoffs.

form $\Delta\omega\tau=2\pi$. It can be seen from Fig. 2 that the gradual increasing of the relative intensity induced a variation of the modulated periodicity of the spectrum in the cutoff. As the relative intensity $\beta=E_1/E_0$ increases from 0.1 to 0.4, the modulated periodicity in cutoff decreases from $1.92\omega_0$ to $1.61\omega_0$. According to the relationship mentioned above, the corresponding pulse interval will increase from $0.52T_0$ to $0.62T_0$ accordingly, where T_0 represents the optical cycle of the fundamental field. The dashed lines in Fig. 2 are used to guide the eyes to see the tendency of the launch of as pulses, which shows remarkable agreement with the prediction of the classical picture as shown in Fig. 1(a). Note that in the few-cycle regime, the CEP plays a key role in modulating the electron dynamics. A change of 0.5π of the fundamental CEP will inverse the tendency of the launch of as pulses. That is, the increasing of the relative intensity β results in the decreasing of the pulse interval for the cutoff regime. Although the $\sin^2$ profile is adopted in the simulation, our calculations show that the time interval of the two attsecond pulses is robust to the shape variation of the driving field. Changing the driving pulse profile (e.g., Gaussian or $\sin^4$ profiles) will not influence the launch positions of the as pulses but modulate the photon energies of the harmonics slightly. Therefore, a two-color scheme can be efficiently applied to manipulating the electron recollision wavepackets and consequently controlling the high harmonic generation in time domain. The twin pulse with an expected delay obtained in our approach can serve as an efficient and simple tool in a pump-probe experiment. This will open new ways of time-resolved studies with unprecedented resolution. Additionally, the stability of the pulse-to-pulse relative intensity can be detected from the modulated spectral structure at the cutoff regions, which shows potential for precise measuring and controlling in the field of laser-matter interaction.

Such a scheme shows a great potential for steering the electron quantum paths. Through perturbing the recollision step related with the ionization step, the emission times of the as pulses can be controlled. Next, we will demonstrate the scheme for precise control of the launch of as pulse. It is well-known that a few-cycle driving field can be used to generate single attosecond pulse [3]. Due to the intensity gate near the peak of the field, electrons with the maximum kinetic energy can only return to the parent core once. This leads to an isolated as pulse production. In our few-cycle bichromatic scheme, a twin pulse is generated for specific CEPs employed in Fig. 2, which shows disadvantage for direct application compared with single as pulse. However, such problem can be circumvented by using the medium with higher potential. Since the intensity of the driving field is below the saturation intensity, the ionization possibility shows sensitive dependence on the laser intensity. Therefore, in the few-cycle regime, the ionization levels in two adjacent half cycles differs remarkably due to the nonadiabatic effect. This will seriously influence the yield of HHG, and the left one of the two-pulse in Fig. 2 will be smeared remaining a single as pulse in the cutoff region. The parameters used in Fig. 3 are the same as Fig. 2 except that the Ne medium is employed. It is shown that a supercontinuous structure appears in the harmonic spectrum instead of a modulated one in Fig. 2. By superposing the harmonics in the cutoff region

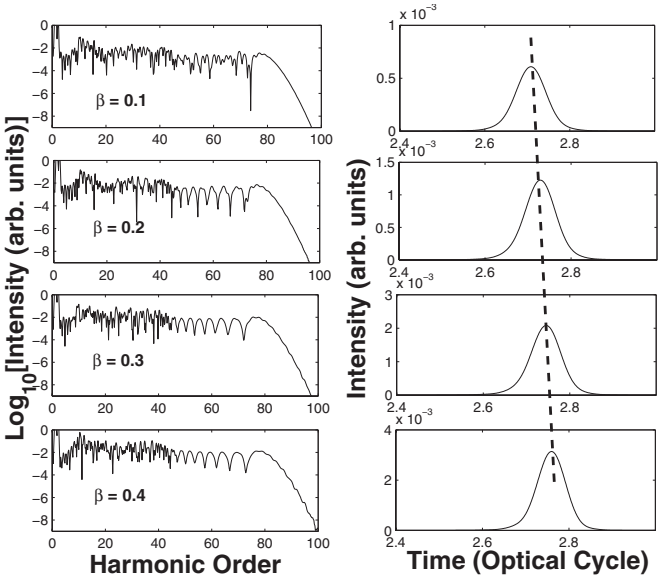

FIG. 3. Harmonic spectra of Ne atoms driven by bichromatic fields with different relative intensities (the left sets of the figures), and single attosecond pulses generated by superposing the harmonics in the cutoff region (the right sets of the figures), the parameters are the same as in Fig. 2.

$(73-90\omega_0)$, single as pulses can be obtained. The launch times of the as pulses show a monotonous dependence on the relative intensity between the SH and the fundamental fields. As increasing β from 0.1 to 0.4, the emission time of the as pulse shifts towards the positive direction gradually. The variation of β as much as 0.1 will induce about a 40-as shift of the as pulse. It can be expected that upon a smaller variation of the relative intensity, the emission time of isolated as pulse can be manipulated with a higher resolution less than 40 as. A question should be addressed is that isolated as pulse can also be obtained by adjusting the phase difference of the driving field without changing the gas medium. However, the yield of the harmonic is about three orders of magnitudes lower than that in the case of Ne medium. Therefore, it is more efficient to generate single as pulse by using medium with higher potential. For a typical pump-probe experiment, the precise control of the probing pulse delay plays a key role in determining the accuracy of the record information. The general technique for adjusting the pulse delay based on the precise positioning stage will shows limitation upon resolution below a few attosecond. In our scheme, the precise control of the launch of as pulses has the potential to breakthrough the limitation and reveals significant application in probing ultrafast dynamics in a rather accurate time scale. By adding a weak SH harmonic field to a strong phase-stabilized laser field, the generation position of the as pulse can be perturbed. The expected accuracy of the launch time is dependent on the minimum variation of the relative intensity.

In conclusion, we have demonstrated a scheme for controlling the launch of attosecond pulses with a rather high accuracy. By adding a weak SH field to the fundamental one. The electron dynamics is perturbed due to the change of the evolution of the electric field. Both the ionization step and

the recollision wave packets can be manipulated through adjusting the relative intensity of the two components of the combined CEP-stabilized laser field. Consequently, the launch of an isolated pulse can be steered with a resolution less than 40 as. Such scheme based on electron quantum path control has significant applications in ultrafast optics.

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