Bayesian optimal control of the ultrashort circularly polarized attosecond pulse generation by two-color polarization gating

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Abstract: We present ab initio simulations of optimal control of high-order-harmonic generation spectra that enable the synthesis of a circularly polarized 53-attosecond pulse in a single Helium atom response. The Bayesian optimization is used to achieve control of a two-color polarization gating laser waveform such that a series of harmonics in the plateau region are phase-matched, which can be used for attosecond pulse synthesis. To find the underlying mechanisms for generating these harmonics, we perform a wavelet analysis for the induced dipole moment in acceleration form, and compare the time-energy representation with the quantum paths extracted from the semiclassical calculation. We found that these coherent harmonics are excited along the short trajectories. The proposed method has the potential to migrate to laboratories for generation of isolated circularly polarized ultrashort attosecond pulses.

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1. Introduction

High-harmonic generation (HHG) is a highly nonlinear optical process that transforms an intense infrared laser field into extreme-ultraviolet (EUV) and soft x-ray radiation with pulse durations in the femtosecond-to-attosecond regime on a tabletop [1–3]. It is an ideal tool to capture the fast charge and spin dynamics in atomic, molecular and condensed matter systems on account of its high temporal coherence. The generation of circular or highly elliptic high harmonics and the corresponding attosecond XUV pulses has numerous applications, including the probe of chiral-specific phenomena such as chiral recognition via photoelectron circular dichroism [4], the study of ultrafast chiral-specific dynamics in molecules [5,6], and x-ray magnetic circular dichroism spectroscopy for time-resolved imaging of magnetic structures [7–9] etc. A promising way to produce isolated ultrashort pulses is to achieve the superposition of a broadband supercontinuum in HHG [1,10–16], and the shortest pulse duration of an isolated ultrashort circularly polarized (CP) pulse reported to date is around 150 attosecond (as) [17].

To date, there are several methods developed to generate CP attosecond pulses experimentally. One type of methods is to use an elliptically polarized laser field directly. High-order harmonics with nonzero ellipticity can be generated and the ellipticity of them increases with the degree of the laser ellipticity [18]. However, the HHG yield drops substantially as the ellipticity of the driving laser field increases because the transverse component of the laser field deviates from the trajectory of the electron and prevents it from recombining with the parent nucleus. Another type of methods is to use two-color counter-rotating circularly polarized laser fields in a collinear geometry. Due to the conservation of photon spin angular momentum, the harmonics have alternating helicity. The advantage of this method is that the pure CP harmonics can be generated with considerable intensities. However, the ellipticity of the synthesized attosecond
pulse is limited by the alternating helicity of the spectrum [19–21]. Some approaches have been developed to solve this problem including using a p-state as the initial state [11,22], adjusting the relative intensities between the two driving laser fields [12,23], tuning the frequency ratio between the two driving laser fields [24], using a circular-elliptic laser field [25], and controlling the phase matching [26]. In these methods, the harmonics with either left or right helicity can be suppressed, resulting in the generation of an elliptically polarized or CP attosecond pulse. In the third type method, two single-color CP laser beams with opposite helicity in a non-collinear geometry are used. Recently, Huang et al. demonstrated that two counter-rotating CP EUV beams can be separated in space, and attosecond pulses with pure circular polarization can be obtained [13].

An isolated attosecond pulse can be also achieved with the help of the polarization gating whose polarization changes with time [27]. The HHG efficiency is the highest for linearly polarized pump pulse, and it rapidly decreases with increasing pump pulse ellipticity. Temporal modulation of pump pulse ellipticity confines the XUV emission to a temporal gate where the polarization is very close to linear, as shown in experiments [28]. Such a gate was obtained by superposing two counter-rotating circularly polarized pulses [29]. In this scheme, single attosecond pulses can be produced in the high-order harmonic plateau.

In order to optimally control the generation of the CP pulses, the parameters of the driving laser beams such as amplitudes, wavelengths, carrier-envelope phases (CEPs), and the relative time delays are all independently adjustable. Since both simulations and experiments of HHG are rather time-consuming, it is impractical to search the entire parameter space either theoretically or experimentally. Previous studies have applied evolutionary algorithms [30,31] and surrogate algorithms [32,33] to design optimal waveforms for HHG in different systems. However, evolutionary algorithms are heuristic, and surrogate methods often assume a local convexity [32,33]. Moreover, uncertainty induced in experiments can make the optimization process more challenging. Recently, the Bayesian optimization (BO) [34–37] has become an attractive option due to its derivative-free feature, capability of handling a black-box or expensive cost functions, and tolerance of stochastic noise in evaluation of cost functions. In addition to a variety of applications including deep neural networks [37–40], BO has been successfully applied to quantum optimal control [41–43]. To the best of our knowledge, BO has not yet been used for the study of HHG control.

In this work, we perform the optimal control of the generation of ultrashort CP attosecond laser pulse using BO. This optimal control scheme is applicable to all kinds of CP attosecond pulse generation methods, and we adopt the bi-chromatic method in a collinear geometry in our study.

2. Theoretical methods

We consider the prototype atomic helium (He) system. In the single-active electron (SAE) model, the time-dependent Schrödinger equation (TDSE) for the electron bound in the He atom and subject to the influence of the two-color polarization gating is written, in atomic units, as

\[ i \frac{\partial \Psi(r,t)}{\partial t} = \left[ \hat{H}_0(r) + \hat{V}(r,t) \right] \Psi(r,t), \]

where \( \Psi(r,t) \) is the wavefunction, \( \hat{H}_0(r) = -\frac{1}{2} \nabla^2 + V_{\text{eff}}(r) \) is the atomic Hamiltonian with a model potential \( V_{\text{eff}}(r) \), in which \( r \) is the electron coordinate, \( r \) is the magnitude of \( r \), and \( t \) is the time [44]. The electron-laser interaction is given by \( V(r,t) = E(t) \cdot r \). In this study, we adopt the bicircular laser field scheme (\( \omega_0 - 3\omega_0 \) fields, \( \omega_0 \) is the fundamental frequency) for the investigation of the HHG helicity control. Earlier studies [24,45] using the \( \omega_0 - 3\omega_0 \) bicircular fields for He atom showed that the harmonic intensity is high and the helicity asymmetry is large. The electric field of the polarization gating \( E(t) \) is the superposition of the left CP (LCP) and the
right CP (RCP) pulses, \( E(t) = E_L(t) + E_R(t) \):

\[
E_L(t) = E_{L0} F(t - T_d/2) [\hat{x} \cos(\omega_0 t + \phi_{CE}) + \hat{y} \sin(\omega_0 t + \phi_{CE})],
\]

\[
E_R(t) = E_{R0} F(t + T_d/2) [\hat{x} \cos(3\omega_0 t + \phi_{CE}) - \hat{y} \sin(3\omega_0 t + \phi_{CE})],
\]

where \( F(t) \) denotes the sine-squared pulse shape, \( T_d \) is the time delay between the two pulses, \( \omega_0 \) is the fundamental frequency, \( \phi_{CE} \) is the CEP, and \( E_{L0}, E_{R0} \) are the peak field amplitudes of the LCP and the RCP pulses, respectively.

The TDSE can be solved efficiently with spectral accuracy by the time-dependent generalized pseudospectral method (TDGPS) in the spherical coordinates [46]. The radial coordinate is discretized by the generalized pseudospectral method [47,48], allowing non-uniform spatial grid spacing: a denser mesh near the origin and a sparser mesh for the outer region. The time propagation of the wave function is achieved by the second-order split operator method in time-dependent techniques using equal-spacing grid discretization.

Once the time-dependent wave function is obtained, the LCP and the RCP components of the HHG spectrum can now be computed, respectively, as

\[
P_L(\omega) = \left| \frac{1}{\omega_f - \omega_i} \int_{\omega_i}^{\omega_f} \frac{1}{\sqrt{2}} [a_x(t) + ia_y(t)] e^{-i\omega t} dt \right|^2 \equiv |\tilde{a}_L(\omega)|^2, \tag{4}
\]

\[
P_R(\omega) = \left| \frac{1}{\omega_f - \omega_i} \int_{\omega_i}^{\omega_f} \frac{1}{\sqrt{2}} [a_x(t) - ia_y(t)] e^{-i\omega t} dt \right|^2 \equiv |\tilde{a}_R(\omega)|^2, \tag{5}
\]

where \( a_x(t) \) and \( a_y(t) \) are the induced dipole moments in acceleration form for the x- and y-directions, respectively. By superposing different harmonics over a frequency window \([\omega_i, \omega_f]\), the attosecond pulse can be calculated as follows:

\[
I_L(t) = \left| \int_{\omega_i}^{\omega_f} d\omega \tilde{a}_L(\omega)e^{i\omega t} \right|^2, \tag{6}
\]

\[
I_R(t) = \left| \int_{\omega_i}^{\omega_f} d\omega \tilde{a}_R(\omega)e^{i\omega t} \right|^2. \tag{7}
\]

In order to produce an isolated ultrashort CP pulse, we need to control the chirality of the HHG spectrum. Determined by our designated helicity of the CP pulse, the HHG can be manipulated via optimization of a properly chosen cost function such that the harmonics are dominated by the preferred helicity. To this end, we will consider the ratio between the RCP and the LCP components as the cost function in our HHG simulations. The resulting optimized HHG spectrum is characterized by a dominant right-handed helicity, either with enhanced harmonic intensities, or an extended cutoff frequency, or both, which in turn leads to a strong and short attosecond pulse polarized in the controlled helicity.

Next, we determine the controlling variables of the two laser pulses. To accommodate the experiments, we choose \( \omega_0 = 0.05695 \) a.u. (800 nm) and fixed the laser field envelopes such that the full width at half maximum (FWHM) of both pulses is 8 femtoseconds (fs). As a result, there are four controllable laser field parameters, namely, the field amplitudes \( E_{L0} \) and \( E_{R0} \), the time delay \( T_d \) and the CEP \( \phi_{CE} \). The four parameters are denoted by \( \mathbf{x} = (E_{L0}, E_{R0}, T_d, \phi_{CE}) \in \mathbb{R}^4 \). In a nutshell, the design of an optimal RCP HHG spectrum can be formulated as the optimization
problem of the following cost function:

\[ f(x) = \frac{\int_{\omega_1}^{\omega_2} P_R(\omega) d\omega}{\int_{\omega_1}^{\omega_2} P_L(\omega) d\omega}. \]  

(8)

where \( \omega_1 = 100\omega_0 \), \( \omega_2 = 260\omega_0 \) are the lowest and the highest frequencies, respectively, of the selected frequency window in the plateau region. Our given window size \([100\omega_0, 260\omega_0]\) is purposely chosen to be sufficiently large by taking into account the fact that in most cases, without optimization, the HHG cutoff frequency will be around 200\(\omega_0\), beyond which the harmonic intensities are 3 orders smaller in magnitude. After the optimization, the cutoff frequency can be extended beyond 220\(\omega_0\), with enhanced harmonic intensities on par with those in the plateau region.

We adopt the BO to seek the optimal waveform in our study. In BO, we first build a probabilistic surrogate model of \( f(x) \) using a Gaussian process based on randomly selected observations of \( f(x) \). To propose the next set of the laser parameters, we construct an acquisition function from the surrogate function, which balances the exploitation and the exploration. The exploitation allows the optimization process to search for the optimal value in the surrogate model, while the exploration prompts the search toward region where the variance of the surrogate model is large. The proposal of the next parameter can be estimated efficiently by optimizing the acquisition function with convex optimization schemes (see Supplement 1 for the details).

3. Results and discussions

To perform BO, the range of the parameter space need to be specified first. If the intensity of the polarization gating is too high, it will lead to ionization of the electron and thus a fall of yield in HHG. Therefore, the field amplitudes \( E_{\ell 0} \) and \( E_{r 0} \) are set to be in the range of 0.0534 a.u. (1.0 \times 10^{14} \text{W/cm}^2) to 0.267 a.u. (2.5 \times 10^{15} \text{W/cm}^2). Similarly, there is an upper bound for the time delay at around 413 a.u., because the intensity at the middle of the polarization gating would be too low if the time delay is too large. In order to produce the circularly polarized pulse, we maximize the ratio between the RCP and the LCP components, as shown in the objective function, Eq. (8). The optimal waveform obtained by BO is presented in Fig. 1, in which the optimized parameters are \( E_{\ell 0} = 0.118 \) a.u. (4.89 \times 10^{14} \text{W/cm}^2), \( E_{r 0} = 0.240 \) a.u. (2.02 \times 10^{15} \text{W/cm}^2), \( T_d = 283.98 \) a.u. and \( \varphi_{CE} = 0 \), which is converged with 600 trials.

![Fig. 1. The optimized polarization gating. The red solid and blue solid lines represent the electric fields in the x- and y-directions, respectively. The black dotted lines represent the envelopes of the two laser pulses. Each envelope has a duration of 8 femtoseconds (fs) for both the \( \omega_0 \) and 3 \( \omega_0 \) components, respectively. Note that the time unit is optical cycle (o.c.).](image-url)
The mechanism of the generation of the ultrashort attosecond pulse can be understood by performing the time-energy analysis and the semiclassical simulations. Figure 2 shows the wavelet time-energy transform \cite{49} of the RCP and the LCP components, respectively. It is found that the spectral density of the RCP component from the 180th to the 235th harmonics is much larger than that of the left component at around 0.48 optical cycle (o.c.). Figure 2(c) presents the HHG spectra for both RCP and LCP components. Note that the indicated window size is used in the BO process. This ultrabroad band of harmonics emits simultaneously. We thereby superimpose the harmonics ranging from the 182nd to the 233rd to synthesize an isolated 53-as CP pulse, which is by far the shortest CP attosecond pulse ever simulated, as shown in Fig. 3(a). The electric field of the synthesized attosecond pulse rotates in the x-y plane is presented in Fig. 3(b).

![Wavelet time-energy representations of (a) the RCP component and (b) the LCP component of the HHG spectra. (c) The RCP and LCP components of the total HHG spectrum with marked frequency window that was used in optimization process.](image)

To investigate the mechanism that generates a series of strong harmonics emitted at around 0.48 o.c. in Fig. 2(a), we perform a semiclassical simulation \cite{50}. Figure 4(a) shows several typical electron trajectories subject to the optimal laser waveform, on which the electron ionization time and returning time are marked by open circles and solid circles, respectively. We examine the trajectories with returning energy in the range of the 182nd to the 233rd harmonic orders, and with the returning time in the interval between 0.45 o.c. and 0.5 o.c. We found that the trajectories that contribute to the quantum path in Fig. 2(a) are mainly short trajectories (blue curves) and multiple returning short trajectories (green curves). Note that the trajectory types are classified by the ionization time and the returning time of the electron.

The blue curve represents a short trajectory that returns to the nucleus one time. Note that there can be numerous of short trajectories due to various initial conditions. It ionizes at 0.23 o.c. as indicated by the blue open circle in Fig. 4(b); it first moves in the negative y-direction and then gradually turns around as the the electric field of the laser is circulating. Finally, it returns to the origin at 0.46 o.c. with the total energy in the interval of the harmonics from the 182nd to...
the 233rd orders. The green curve represents a short trajectory that returns to the nucleus two times. It ionizes at 0.26 o.c. as indicated by the green open circle in Fig. 4(b). Similar to the blue trajectory, it first moves in the negative y-direction, but it turns back to the nucleus at 0.35 o.c., which is earlier than that of the blue trajectory. After the first return, it goes out again, moving in the positive y-direction; as the direction of the electric field of the laser is reversed, it returns to the origin again at 0.5 o.c., with the total energy also in the interval of the harmonics from the 182nd to the 233rd orders. Both of them rotate in the counter-clockwise direction, the same way as the electric-field vector of the attosecond CP pulse [Fig. 3(b)], and they mainly contribute to the RCP component. In addition to the short trajectories, some long trajectories may also contribute to the quantum path in Fig. 2(a). The red curves represent some of the long trajectories and are more complicated. The red solid trajectory ionizes at 0 o.c. as indicated by the red open circle in Fig. 4(b) and returns to the origin at 0.6 o.c. with the total energy in the interval of the harmonics from the 182nd to the 233rd orders. These trajectories rotate in the clockwise direction and mainly contribute to the LCP component. However, since most of these long trajectories do not return to the origin in the time interval 0.45-0.5 o.c., they have small contribution to the generation of the attosecond CP pulse [cf. Figure 3(a)]. In a nutshell, within a short interval between 0.45 and 0.5 o.c., short trajectories are predominant with returning energy ranging from the 182nd to the 233rd harmonics, which constitute a broadband of plateau spectrum and are beneficial for attosecond pulse synthesis.

We further show that the constituent harmonics that form the 53-as pulse are not only synchronized but also phase-matched. The time profile \( a_{\omega q}(t) \) of the harmonic \( \omega_{q} (= q\omega_{0}) \) can be sliced from the time-energy representation from the wavelet transform of the induced dipole
moment in acceleration form \([15,49]\). The time profiles as well as the dynamical phases \([15,49]\) at their peak time \(t_e\) are presented in Fig. 5. Note that the peak emission time can be interpreted as the electron-ion recollision (or recombination) time in a semiclassical sense. The dynamical phase difference \(\Delta \theta\) between adjacent harmonics, separated by \(\Delta \omega = \omega_0\), is nearly constant at \(\Delta \theta \approx 3\) radians, suggesting that these harmonics are phase-matched \([51–53]\). We can estimate the emission time \([54]\) of these harmonics from the dynamical phase difference as follows:

\[
t_e = \frac{\Delta \theta}{\Delta \omega} \approx 0.48 \text{ o.c.},
\]

which is consistent with the peak time of the attosecond pulse in Fig. 3(a), as well as the emission time of the short trajectory from the semiclassical simulation in Fig. 4, which is also 0.48 o.c. The emission time from each individual harmonic originating from the short-trajectory electron does not significantly vary in time from harmonic to harmonic, indicating that all these harmonics are emitted virtually at the same instance and their phases are coherent.

**Fig. 4.** (a) The electron trajectories on the x-y plane. The red lines with different line style represent long trajectories with different initial conditions. The blue and the green lines represent short trajectories. The arrows along these trajectories indicate the motion of the electron. (b) The black curve represents the electric field vector \(E(t)\) for \(-0.4 \text{ o.c.} \leq t \leq 0.6 \text{ o.c.}\) in the direction specified by the arrows. For the beginning and the end points on the curve, the time \(t\) is indicated. The times for which the investigated orbits in (a) start and terminate are marked by open circles and solid circles, respectively, with the corresponding color.
4. Conclusion

In summary, we have successfully demonstrated numerically the synthesis of a 53-as RCP pulse that is the shortest ever simulated in a helium single atom response. The single atom response was performed by solving the three-dimensional Schrödinger equation of helium atom driven by a two-color polarization gating. We apply the Bayesian optimization to obtain an optimal waveform. The resultant HHG spectrum consists of a series of phase-matched harmonics in the plateau region that emit simultaneously and can be used to synthesize an isolated CP attosecond pulse. By comparing the wavelet analysis and semiclassical trajectories, we found that these phase-matched harmonics are predominantly generated along short trajectories that return simultaneously. Our study enables a deeper understanding of the mechanism and electron dynamics of the HHG, and provides a fresh new insight to this important field of ultrafast science and technology. The proposed BO method is expected to serve as a viable means in generating ultrashort attosecond pulses experimentally in the future.

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Disclosures. The authors declare that there are no conflicts of interest related to this article.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

References


