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Optimal control of attosecond pulse synthesis from high-order harmonic generation

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Numerical solutions of the time-dependent Schrödinger equation for a three-dimensional H atom and an efficient genetic algorithm are used to optimize short intense excitation laser pulses in order to generate high-order harmonics from which we synthesize *single* attosecond pulses. It is shown that chirping of excitation pulses at intensities $\sim 10^{14}$ W/cm^2 and duration of up to $\sim$16 fs can lead to synthesis of single attosecond pulses. The optimal excitation pulses and the phases of the generated harmonics are compared with the nonoptimized ones, showing thus the usefulness of genetic algorithm schemes in the search of optimal conditions for synthesizing single attosecond pulses.

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Optimal laser control of atomic and molecular processes is a new area of research with wide applications to chemistry and physics [1]. Within this framework is the nonlinear response of atoms and molecules to intense laser pulses, where high-order harmonic generation (HOHG) of orders exceeding 250 has been experimentally observed [2,3]. The basic mechanism of HOHG has been established from simple *rescattering* models [4,5]. It has been shown recently that enhancement of single harmonics can be achieved by adjusting the phase (chirp) of the excitation laser pulse and that the enhancement is the result of the single-atom intrinsic phase of the generated harmonic [6,7]. This concept has been verified numerically by a coherent control study of single-harmonic generation using a genetic algorithm optimization scheme [8–10].

We study here another possible application of genetic algorithms with the goal of opening the way to the control of the time evolution of the electron response to intense laser pulses in atoms and molecules for the generation of *single* ultrashort coherent electromagnetic pulses on *attosecond* time scale. Generally, the emitted field consists of train of pulses in the attosecond range [13]. One way to obtain a single pulse of the generated field is the use of the continuum HOHG excited by very short ($\sim$5 fs) intense (10^{14}–10^{15} W/cm^2) 800 nm pulses [11,12]. We address in this work a strategy to obtain single attosecond pulses using longer (duration $\tau_p \sim 8$–16 fs) but appropriately chirped exciting laser pulse. Thus instead of enhancing a single specific high harmonic, as in Refs. [6,9], we study through a genetic algorithm scheme the optimization of the exciting laser phase in order to obtain a single high-frequency attosecond pulse by suppressing the surrounding satellites present in the generated pulse train. We have shown recently that combinations of such intense short pulses with single attosecond pulses [14] can lead to a new route for efficient synthesis of shorter single attosecond pulses and new applications [15].

In this paper we study numerically exact solutions of the three-dimensional (3D) H atom excited by a short intense laser pulse ($\tau_p = 8$ fs or $\tau_p = 16$ fs, $I = 2 \times 10^{14}$ W/cm^2) with a central wave frequency at $\omega_0 \simeq 12\,500$ cm^{-1} ($\lambda = 800$ nm). Control of HOHG spectrum is achieved by means of genetic algorithm optimization [16,17]. The parameters optimally controlled as in previous experimental studies are those entering in the definition of the carrier wave phase $\phi(t)$ or its associated frequency $\omega(t) = d\phi(t)/dt$ [6,7]. The semiclassical rescattering model [4–7] is referred to for the elucidation of the role of the laser phase in control of the motion of the tunneling electron in the chirped oscillating laser field. More precisely the exciting linearly polarized laser pulse is given the following form:

$$\mathcal{E}(t) = \mathcal{E}_0(t)\cos[\phi(t)], \quad \mathcal{E}_0(t) = E_0 \sin^2\left(\frac{\pi t}{T}\right), \tag{1}$$

corresponding to a sine-square pulse of maximum amplitude E_0 and width T [the duration τ_p corresponding to the full width at half maximum of the pulse envelope $\mathcal{E}_0(t)$ is $\tau_p = 0.364T$]. The instantaneous frequency $\omega(t)$ is limited to a cubic chirp:

$$\phi(t) = \omega_0 t + 2\pi \sum_{i=0}^{4} \alpha_i \left(\frac{t}{T}\right)^i, \tag{2}$$

with $|\omega(T/2+\tau_p/2) - \omega(T/2-\tau_p/2)| \leqslant 0.1\omega_0$. Optimal control deals with the search for and adjustment of the chirp parameters ω_0, $(\alpha_i)_{i=0,4}$ to which are included for completeness the maximum field amplitude E_0 and the position ω_c of a Gaussian-shaped filter aiming at the selection of a HOHG spectrum region (as discussed below).

The *objective* function J to be optimized is determined directly by a time profile $\mathcal{E}^a(t)$ of a synthetized attosecond

pulse. The calculation is based on the dipole acceleration $\ddot{d}(t)$ [18] obtained from the wave function $\psi(t)$, which is the exact solution of the time-dependent Schrödinger equation (TDSE) for a hydrogen atom in the field $\mathcal{E}(t)$ [Eq. (1)] within a 3D model. This is given (in atomic units $\hbar=1$, $e=1$, $m=1$) by

$$\ddot{d}(t) = \frac{d^2}{dt^2}d(t) = -\langle\psi(t)|\frac{\partial}{\partial z}V(\vec{r},t)|\psi(t)\rangle, \tag{3}$$

$$i\frac{\partial}{\partial t}|\psi(t)\rangle = [\hat{H}_0 + z\mathcal{E}(t)]|\psi(t)\rangle, \tag{4}$$

$$V(\vec{r},t) = V_c(\vec{r}) + z\mathcal{E}(t), \tag{5}$$

$$|\psi(t=0)\rangle = \psi_{1s}(\vec{r}), \tag{6}$$

where $\hat{H}_0$ is the field-free H atom Hamiltonian and $V(z,t)$ is the total potential, Coulomb potential $V_c(\vec{r})$ plus the field-atom interaction $z\mathcal{E}(t)$ for an initial state where the electron is in the ground 1*s* orbital. A third-order unitary split-operator propagation technique [19] is used to obtain a highly accurate nonperturbative time evolution of the electron wave packet $\psi(t)$ on a 3D grid ($0\leqslant r\leqslant 256$ bohr, $0\leqslant\theta\leqslant\pi$, $0\leqslant\varphi\leqslant 2\pi$) and using absorbing boundary conditions. The highly accurate wave function $\psi(t)$ leads after Fourier transform to the HOHG power spectrum $|\ddot{d}(\omega)|^2 = \omega^4|d(\omega)|^2$ with its typical frequency dependence: an initial intensity decline corresponding to ground-state dipole emission [20] followed by a plateau with an energy cutoff around $3U_p$, where $U_p=E_0^2/4\omega_0^2$ is the electron ponderomotive energy [4,5]. The emitted field is constructed as

$$\hat{\mathcal{E}}^a(\omega) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}\ddot{d}(t)e^{-i\omega t}dt. \tag{7}$$

As has previously been mentioned, a frequency filter $f_{\omega_c}(\omega)$ centered at ω_c (optimized between the 11th and the 31st harmonics) is introduced to ultimately reach the dipole emission pulse:

$$\mathcal{E}^a(t) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}f_{\omega_c}(\omega)\hat{\mathcal{E}}^a(\omega)e^{i\omega t}dw. \tag{8}$$

The precise shape of the filter used is $f_{\omega_c}(\omega)=g_{\omega_c}(\omega)h_{\omega_c}(\omega)$ where $g_{\omega_c}(\omega)$ is a simple Gaussian filter given by $g_{\omega_c}(\omega) = e^{-\beta[(\omega-\omega_c)/\omega_0]^2}$ and $h_{\omega_c}(\omega)$ is given by

$$h_{\omega_c}(\omega) = \begin{cases} \dfrac{|\hat{\mathcal{E}}^a(\omega_c)|}{|\hat{\mathcal{E}}^a(\omega)|} & \text{if } g_{\omega_c}(\omega)|\hat{\mathcal{E}}^a(\omega)| > |\hat{\mathcal{E}}^a(\omega_c)|, \\ 1 \quad \text{otherwise.} \end{cases} \tag{9}$$

Ideally, to synthetize a Gaussian-like attosecond pulse one should use for $\hat{\mathcal{E}}^a(\omega)$ Gaussian frequency profile as well. This filter, which is still technologically reachable, has a Gaussian shape with the additional property of trimming harmonics exceeding a chosen central value.

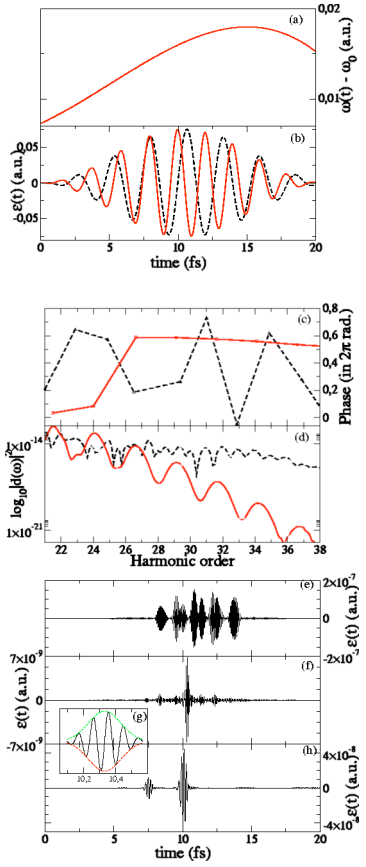

FIG. 1. Optimized emitted pulse using direct criterion J_2 in a full 3D model: (a) the chirped function; (b) the excitation laser (dotted line, without chirp; solid line, with chirp) with a duration of eight cycles; (c) phases calculated at the spectrum peaks; and (d) amplitudes of the HHOG spectrum (dotted line, without chirp; solid line, with chirp); (e) and (f) emitted laser pulse without and with optimization, respectively; (g) optimally obtained attosecond pulse (solid line) with a Gaussian-type fitted envelope (dotted line); (h) emitted laser pulse with optimization using a simple Gaussian filter.

First we performed calculations using a relatively short ($T=8$ cycles, $t_p=7.8$ fs) unchirped 800 nm laser pulse, i.e., we used Eq. (2) with $\alpha_i=0$, $i=1.4$. The resulting attosecond pulse, shown in Fig. 1(e), contains five distinct pulses. Since our goal is to extract one pulse from such a train of pulses we used following criteria J_1 and J_2 in our genetic algorithm scheme. The mathematical implementation of such criteria is as follows:

$$J_1 = I_1/I_2,$$

with

$$I_1 = \int_{t_c-t_{pulse}}^{t_c+t_{pulse}}|E^a(t)|^2dt \tag{10}$$

and

$$I_2 = \int_0^{t_c-t_{pulse}}|\mathcal{E}^a(t)|^2dt + \int_{t_c+t_{pulse}}^{T}|\mathcal{E}^a(t)|^2dt, \tag{11}$$

or another criterion

TABLE I. Optimally adjusted parameters as obtained from the genetic algorithm, corresponding to Figs. 1 and 2.

τ_p (fs)	I (W/cm^2)	ω_c/ω_0	α_0	α_1	α_2	α_3	α_4
7.8	3.74×10^{14}	30.3	0.37	0.99	0.99	0.92	−0.94
15.5	2.03×10^{14}	30.9	0.98	1.0	−0.73	1.0	0.72

$$J_2 = [|\mathcal{E}^a(t_c)| / |\mathcal{E}^a(t_{\bar{c}})|], \tag{12}$$

where t_{pulse} taken as 20 a.u. is the expectedly short duration of the emitted pulse. As for t_c and $t_{\bar{c}}$ they are, respectively, defined as $t_c = \text{argmax}_{[0,T]}(\mathcal{E}^a(t))$, and $t_{\bar{c}} = \text{argmax}_{\{t \leqslant t_c - t_{pulse} \text{ and } t \geqslant t_c + t_{pulse}\}}(\mathcal{E}^a(t))$. The goal of J_1 is to maximize the time integrated pulse intensity profile $|\mathcal{E}^a(t)|^2$ with respect to the integrated intensity of the remaining part, whereas the goal of J_2 is to maximize the amplitude of the central pulse as compared to the maximum of the other part of the pulse. The optimization loop starts with some initial candidate fields $\mathcal{E}(t)$, solves the TDSE for a H atom excited by such a field, and gets the power spectrum $\hat{\mathcal{E}}^a(\omega)$. Next, by a windowed Fourier transform, the desired temporal shape of the emitted pulse $\mathcal{E}^a(t)$ is obtained [Eq. (8)]. Using one of the criteria J_1 or J_2, Eq. (10) or (12), respectively, as a fitness function, the genetic algorithm proceeds through crossovers and mutations to generate offsprings and select the parameters of the next generation of fields. This procedure is repeated until convergence is achieved (typically after 200 generations). Best results were reached with the use of J_1 (rather than J_2), which is finally retained for full 3D calculations. Although filters as the one under consideration in Eqs. (8) and (9) are experimentally reachable, we have also considered other simpler, rectangular, or Gaussian filter shapes. The rectangular one due to its discontinuities leads to nonadiabatic dynamics with, as a consequence, more marked satellite peaks in the generated pulse. The optimization with an adiabatic Gaussian filter, within standard experimental possibilities, has finally been retained.

The most convincing result of a full 3D model with criterion J_1 is displayed in Fig. 1. The numerical values of all adjusted parameters by the optimization are collected in Table I. As for the fixed parameters, the following values are used: $T=20.6$ fs, $\beta=2\ \ln(2)/25$, $\omega_0=5.87\times10^{-2}$ a.u. The optimization strategy turns out to be particularly efficient in producing an ultrashort emission pulse well isolated from its satellites [almost one order of magnitude, see Fig. 1(f) resulting from the filter of Eq. (9) and 1(h) from a simple Gaussian filter]. It is interesting to observe that the control scheme in consideration as compared to the previous strategy brings deep modifications to the nonoptimized emission field, when comparing panels (e) and (f). It is also worthwhile noting that the phases of the harmonics, calculated at the spectrum peaks extending from 27 to 35, are nearly equal in Fig. 1(c). A more detailed analysis of the emitted pulse is presented in Fig. 1(g). This can be conducted by the consideration of the mean pulse (neglecting all satellites and limited to the time interval $[t_1=10.09\ \text{fs}, t_2=10.57\ \text{fs}]$) and by performing a Gaussian-type fit of its envelope. Three remarks are in order:

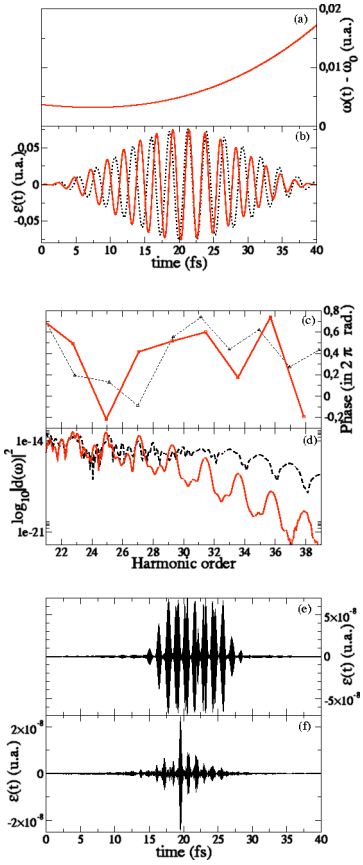

FIG. 2. Optimized emitted pulse using direct criterion J_2 in a full 3D model: (a) the chirped function; (b) the excitation laser (dotted line, without chirp; solid line, with chirp) with a duration of 16 cycles; (c) phases calculated at the spectrum peaks; and (d) amplitudes of the HHOG spectrum (dotted line, without chirp; solid line, with chirp); (e) and (f) emitted laser pulse without and with optimization, respectively.

(i) An attosecond pulse production strategy is definitely reached, in the sense that the envelope of the emitted field extends no more than about $t_2-t_1\simeq200$ as.

(ii) Four to five oscillations of the carrier wave, corresponding to a frequency $\omega=1.59$ a.u., are within this envelope. We note that this frequency is in the range of the neighboring 27th and 29th harmonics (i.e., $\omega_{27}=1.54$ a.u. and $\omega_{29}=1.65$ a.u.).

(iii) As expected for a freely propagating electromagnetic field, the time average of $\mathcal{E}^a(t)$ limited to the main (atto) pulse is very close to zero [2]. More precisely

$$\int_{t_1}^{t_2} \mathcal{E}^a(t)dt \simeq 4.0\times10^{-12}\ \text{a.u.}, \tag{13}$$

to be compared to the area of the Gaussian envelope, i.e., 3.6×10^{-9} a.u. We thus emphasize that the isolated emitted pulse satisfies the standard characteristics of a laser with an attosecond time shape. A more challenging optimization is conducted with a full 3D calculation, still using criterion J_1, but with a $T=16$ cycles excitation laser field ($\tau_p=15.6$ fs).

Using a longer exciting laser field makes the optimization more difficult since the emitted field presents, prior to optimization, more marked satellites [see Figs. 1(e) and 2(e)]. Despite this difficulty our algorithm succeeds and provides a well isolated central pulse of the emitted laser [see Fig. 2(f)]. Note that now the resulting harmonic spectrum, Fig. 2, has very distinct pics (much better separated than before optimization) and slightly narrower than in Fig. 1. We also note that the variation of harmonic phase is now much more significant than in the case of shorter 800 nm pulse. We believe that phases are constant in Fig. 1 because the pulse duration is only 8 fs, which means that this intermediate case is still close to that used in Refs. [11,12] (t_p=5 fs). In this case the harmonic spectrum is nearly continuous and for such case one can synthetize single attopulse using constant phases as in the case of extremely short pulses. However, for longer pulses $t_p \simeq 16$ fs (Fig. 2), the algorithm finds a special phase relationship between harmonics and the harmonic spectrum is quasidiscrete, with each peak slightly narrower than that in Fig. 1. Clearly, chirping provides a complementary way for generating attosecond pulses, which allows us to use much longer pulses than that used previously [11,12].

In conclusion, we have shown from numerical solutions of TDSE's that zero-area single attosecond pulses can be synthetized from quasidiscrete harmonics generated by intense short pulses optimized by genetic algorithm schemes applied to single atom. Up to date the study of single atoms constitutes the general context [10], but strictly speaking the spatial properties of the pulse imply propagation over an ensemble of atoms. Optimization of such a system is currently not feasible numerically but needs eventually to be addressed. The optimization was achieved by finding the appropriate pulse phase (chirping) which broke the periodicity of the generating laser field and allowed us to use much longer $\tau_p \sim 16$ fs than that used in Refs. [11,12]. We thus have shown that optimization schemes used previously in the coherent control of photophysical processes [21] should be also applicable to synthesizing single attosecond pulses.

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