Generation of ultrashort pulses of harmonics

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We study harmonic generation by a single atom exposed to two short perpendicularly polarized laser pulses. The two perpendicular electric fields oscillate at two different frequencies ω_1 and ω_2. Hence, the resultant field has a polarization which depends on time. Since harmonics are emitted when the resultant oscillating field is linearly polarized, it is expected that a short pulse of harmonics may be emitted if the external field is linearly polarized during a short period of time. We show that, indeed, the atom may emit an ultrashort pulse of a *given* harmonic. This result has been obtained by time-frequency analyzing the acceleration of the induced dipole moment with a filter whose frequency bandwidth is smaller than twice the frequency of the external field. Our calculation of the dipole acceleration is based on the numerical solution of the time-dependent Schrödinger equation. We then address the question of how far it is possible to reduce the duration of the emitted pulse of one given harmonic by adjusting both ω_1 and ω_2 and keeping the amplitude of this pulse significant. In order to answer to this question, we used the quantum version of the two-step model [M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994)]. [S1050-2947(96)50109-5]

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An atom that is exposed to a strong electromagnetic field may emit high-order harmonics of the driving field [1]. The physical origin of this phenomenon has been recently attributed to a two-step process [2]: the atomic electron first tunnels out; it is then driven back by the field towards the ion where it is scattered or recombines back to the ground-state emitting harmonics. From this semiclassical interpretation, it is clear that this phenomenon not only depends on the frequency and intensity of the field but also on its polarization. In the case of linear polarization, some of the classical trajectories of the electron pass the ion periodically, allowing for recombination and harmonic generation. There are strictly speaking, no such trajectories for elliptic polarization. Harmonic generation is in that case possible only thanks to the finite extent of the electronic wave packet and quantum diffusion effects. As a result, the harmonic generation efficiency is expected to decrease rapidly with an increase of the ellipticity of the field polarization [3,4].

When the field is linearly polarized, the above interpretation indicates that harmonics are emitted as a train of ultrashort pulses. Every half an optical period, an electron may be emitted and driven back by the field towards the residual ion where it may recombine, emitting an harmonic pulse [5]. Its duration is only determined by the electron oscillations. Since harmonic generation strongly depends on the polarization of the external field, Corkum *et al.* [6] have suggested that one of these ultrashort pulses of harmonics may be selected by using a driving field whose polarization is time dependent. The polarization has to vary in such a way that the polarization is linear during a period of time shorter than half an optical period. A small amount of ellipticity erases the other pulses of the train. A time-dependent polarization is achieved, for instance, when the field is composed of two perpendicular fields that oscillate at two different frequencies ω_1 and ω_2. Adjusting the difference between both frequencies leads to a time control of the polarization. Since the emission of these pulses is periodic in time with a frequency equal to twice the frequency of the field (or the average frequency of both fields in the case of a time-dependent polarization), the observation of these pulses of harmonics requires a filter whose frequency bandwidth is larger than 2ω [7]. In other words, these ultrashort pulses contain frequency components associated with at least three consecutive harmonics. It is important to stress that the idea of Corkum *et al.* for producing extremely short pulses relies on the assumption that all contributing harmonics are in phase *after propagation* through the medium. So far, besides only one theoretical result, [8] there is no experimental evidence that these harmonics are indeed in phase. In this contribution, we address the problem of the pulsed emission of harmonics in a different way: we analyze the atomic dipole acceleration with a filter whose frequency bandwidth is shorter than 2ω in order to select only one given harmonic. Here, our first objective is not the production of the shortest possible pulses. Instead, we want to show that one can produce short pulses of one given harmonic by adjusting the time during which the external field is linearly polarized. In this way, the pulse duration depends on the time the external field is linearly polarized rather than on any phase relation between harmonics.

In the case of a time-independent polarization and if the frequency bandwidth of the filter is shorter than 2ω [7], the time profile of the harmonics depends essentially on the shape of the external laser pulse and does not exhibit any fast

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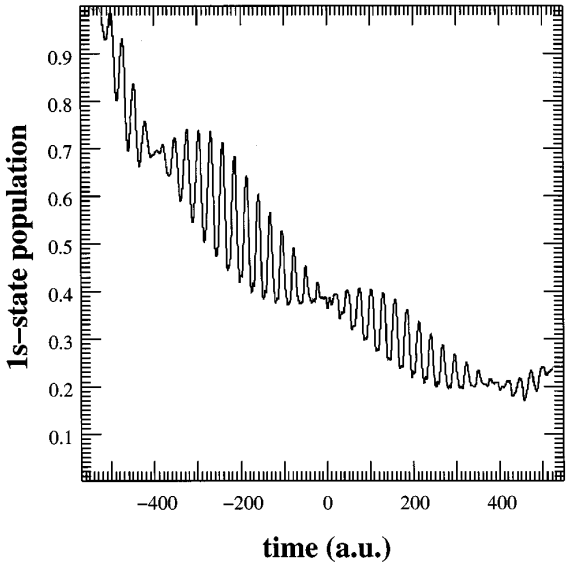

FIG. 1. 1s-state population, i.e., the projection of the full wave function on the bare 1s state of atomic hydrogen, as a function of time in a.u. in the case where the atom is exposed to two perpendicularly polarized laser pulses of 10^{14} W/cm^2. The photon energies are 0.118 and 0.110 a.u. Both pulses have a flat top and sine square turn-on and -off. In both cases, the total duration of the pulse is 20 optical cycles, and it is turned on and off over two optical cycles.

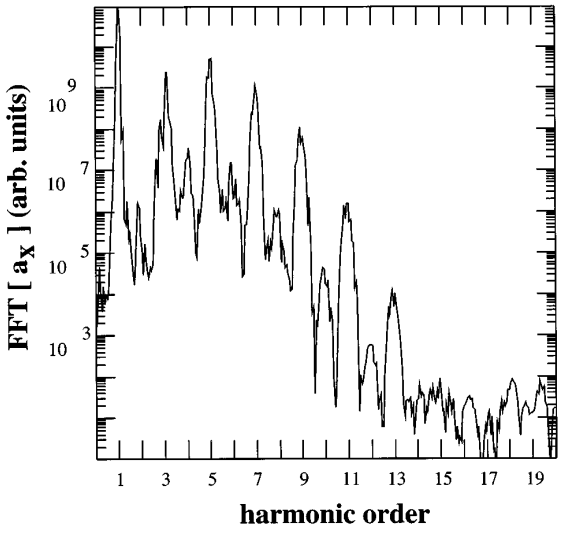

FIG. 2. Fast Fourier transform in arbitrary units of the x component of the acceleration of the atomic dipole moment for the same case as in Fig. 1. The y component, not shown here, has the same behavior. It is important to stress that the harmonic order given in the abscissa refers to the average frequency $\overline{\omega}$.

oscillation. In the present case we use an external field whose polarization is time dependent and study in detail the possibility of observing the emission of a short pulse of harmonics and how its width and amplitude can be controlled with the difference between both frequencies ω_1 and ω_2.

In the case of a time-dependent polarization, all the calculations that have been carried out so far [6] are based on the quantum-mechanical version of the two-step model [9], which includes the effects of quantum tunneling, quantum diffusion, and interference. The validity of this model has only been checked in the case of low frequencies, high-field intensities, and linear polarization. Here, we present results obtained by solving the corresponding three-dimensional Schrödinger equation. We consider the interaction of atomic hydrogen with a short electromagnetic pulse whose field polarization is time dependent. We solve numerically (and without any approximation) the time-dependent Schrödinger equation [10]:

$$i\frac{\partial}{\partial t}\Psi(\vec{r},t)=[H_0+\vec{A}(t)\cdot\vec{p}]\Psi(\vec{r},t), \tag{1}$$

where H_0 is the atomic Hamiltonian and $\vec{A}(t)$ the vector potential associated with the field

$$\vec{A}(t)=A_0 f(t)[\cos(\omega_1 t)\vec{e}_x+\sin(\omega_2 t)\vec{e}_y]; \tag{2}$$

A_0 is the amplitude of the potential, $\vec{e}_x$ and $\vec{e}_y$ the unit vectors along the x and y axes, respectively, ω_1 and ω_2 the frequencies, and $f(t)$ is a slowly varying envelope. We expand the total wave function on the basis of complex Coulomb-Sturmian functions [11] in the radial coordinate and spherical harmonics in the angular coordinates. As a result, we obtain a set of coupled first-order differential equations in time for the expansion coefficients. These equations are solved numerically by means of a fully implicit Runge-Kutta method of order 7. It is important to note that due to the lack of cylindrical symmetry, the azimuthal quantum number m of the electron is not conserved and the problem is now a fully three-dimensional one. This implies that within our method, the number of atomic states involved is extremely high. In order to get convergence in the case treated here, we have included 82 000 atomic states. The implicit character and the high order of our method ensure a stable time propagation of the solution. In order to minimize the execution time, the time step as well as the total number of angular momenta included are adjusted by the code itself. Knowing the wave function $\Psi(\vec{r},t)$, it is a simple matter to evaluate the dipole acceleration $\vec{a}(t)$ by means of Ehrenfest's theorem.

The time profile of a given harmonics may be obtained by means of a time-frequency analysis (either Gabor or wavelets) of the dipole acceleration $\vec{a}(t)$ [13]. Here, we use a Gabor analysis in order to get a time resolution that is independent of the harmonic order [12]. This type of analysis consists of introducing a time-frequency transform that depends on two adjustable parameters denoted by α and β:

$$\vec{a}(t)\rightarrow\vec{a}(\alpha,\beta)=\int_{\infty}^{\infty}T_{\alpha\beta}(t)\vec{a}(t)dt, \tag{3}$$

where the analyzing function $T_{\alpha\beta}(t)$ is given by

$$T_{\alpha\beta}(t)=e^{-it/\alpha}G(t-\beta). \tag{4}$$

$G(t)$ is a window function, which in the present case is a Gaussian. $1/\alpha$ defines the frequency of oscillation of the analyzing function and β the position of the window function in the analyzed signal. This time-frequency transform is significantly nonzero at a given time when the signal oscillates at a

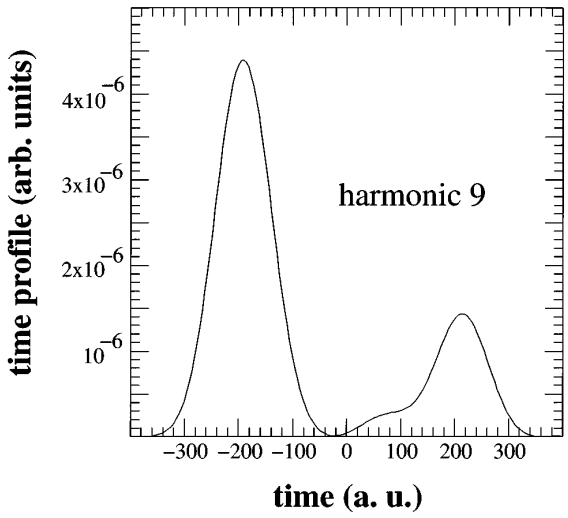

FIG. 3. Time profile in arb. units of the ninth harmonic, emitted by atomic hydrogen exposed to the same pulses as in Fig. 1.

frequency close to the frequency of the analyzing function $T_{\alpha\beta}(t)$ and therefore, acts as filter, both in time and frequency.

Let us now examine the interaction of an atomic hydrogen with a flat electromagnetic pulse with a two-optical-cycle sine square turn-on and -off. The total duration of the pulse is 20 optical cycles. The electric field is composed of two perpendicular fields (in the x-y plane) that oscillate at two different frequencies. The corresponding photon energies are 0.118 and 0.110 a.u. In the present case, the electric field is circularly polarized at time $t=\pm 400$ a.u. and $t=0$, and linearly polarized at time $t=\pm 200$ a.u. The maximum intensity associated with both fields is 10^{14} W/cm^2. In Fig. 1, we represent the ''1s-state population,'' i.e., the square of the projection of the total wave function on the bare atomic 1s state as a function of time. This projection gives information on the motion of the atomic electron wave packet during the interaction of the atom with the field. The ''squeezes'' of the curve around 0 (middle of the input pulse) and ± 400 a.u. of time arise because at these times, the polarization of the field is circular. In that case, the wave packet does not oscillate through the nucleus. At time ± 200 a.u., the field is linearly polarized and the 1s-state population exhibits fast oscillations associated with the back and forth motion of the electron wave packet through the nucleus. These oscillations occur at the average frequency $\overline{\omega}=(\omega_1+\omega_2)/2$. We have checked that the population that leaves the ground state is ionized rather than transferred to an excited state. However, it is important to note that ionization is much slower than in the case where the field stays linearly polarized during the entire duration of the input pulse [7]. This has an interesting consequence: we may force the system to emit harmonics when the input pulse intensity is maximum if the electric field is linearly polarized in the middle of the pulse by preventing ionization during the increasing edge of the pulse. The first tests indicate that in our regime of laser frequencies and intensity, we could increase the intensity of each harmonic significantly.

In Fig. 2, we show for the same case as before, the fast Fourier transform of the x component a_x of the dipole acceleration (the y component, not shown here, behaves exactly in the same way). This spectrum of harmonics of the averaged frequency $\overline{\omega}$ exhibits the usual features, namely, a ''plateau'' and a relatively sharp cutoff. By means of a Gabor time-frequency analysis of $\vec{a}$ (the frequency bandwidth of the filter is roughly equal to $\overline{\omega}/2$), we have studied the time profile of the ninth harmonic. The results are presented in Fig. 3. We clearly see that two well-defined pulses are emitted around ± 200 a.u. of time when the external field is linearly polarized. The second maximum is smaller due to the partial ionization of the atom. In both cases, the time width at half maximum is equal to 3 fs, which is about twice the laser period. All these results have been obtained by solving numerically the time-dependent Schrödinger equation. It is worth mentioning that, qualitatively, the results for the harmonic spectrum are in reasonable agreement with those obtained with the two-step model, although, strictly speaking, the present conditions do not correspond to its validity domain. In fact, the cutoff of the harmonic spectrum is sharper in the case of the two-step model.

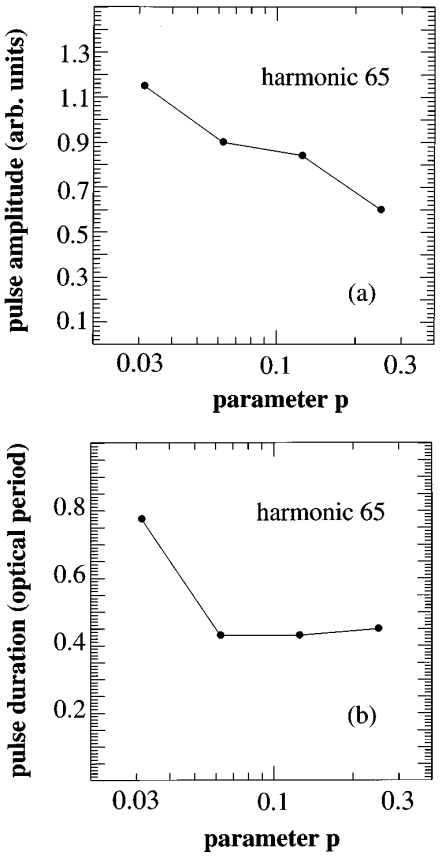

FIG. 4. Amplitude (a) and duration (b) of the pulse of the 65th harmonic emitted by a model atom of neon exposed to two perpendicular fields. The total intensity is 6×10^{14} W/cm^2. Both amplitude and duration are given as a function of the parameter p, the frequencies ω_1 and ω_2 being defined in terms of p as $\omega_1=\overline{\omega}(1-p)$ and $\omega_2=\overline{\omega}(1+p)$.

So far, we have shown by means of an ''exact'' calculation that we can produce ultrashort harmonic pulses by using a time-dependent polarization field. Whether or not the duration of these pulses can be further reduced is an interesting question. However, in order to avoid long execution times of

the code, we tackled this problem by using the two-step model. Because the validity of this model is restricted to lower frequencies, we have considered the following case: the average frequency is 0.057 67 a.u., the total intensity of the field is 6×10^{14} W/cm^2, and the ionization potential is the one of the neon atom. Depletion of the ground state is still negligible at the intensity considered. We introduce the parameter p such that

$$\omega_1=\overline{\omega}(1-p), \tag{5}$$

$$\omega_2=\overline{\omega}(1+p). \tag{6}$$

The values of p we choose are 1/32, 1/16, 1/8, 1/4, and 1/2; this means that for a given p, the resultant electric field is linearly polarized at time $t=nT/4p$, where n is an integer number and $T=2\pi/\overline{\omega}$. In the following, we analyze the time profile of the 65th harmonic, which for the case considered here, is in the ''plateau'' region of the harmonic spectrum. The time resolution of our Gabor analysis is $T/3$. In Fig. 4, we study both the amplitude and the duration of the pulse of the harmonic 65 as a function of p. As expected, the amplitude of the pulse decreases as p increases but its variation stays relatively small; in other words, the harmonic conversion efficiency depends only slowly on the rate of change of the light ellipticity. Moreover, as shown in Fig. 4(b), the pulse duration decreases until it reaches the limit value of 0.42 optical cycles for $p=1/16$. The pulse duration for $p\leqslant1/16$ may be understood as follows: if we assume that the harmonic efficiency is reduced by a factor of 2 when the ellipticity is equal to ±0.1 [14], the harmonic emission should last $1/40p$ of an optical period [15]. When $p=1/8$ or 1/4, the pulse duration is roughly constant. We also checked that this duration is not limited or imposed by the time resolution of our analysis. In order to understand this behavior, it is important to remember that within the quasiclassical model the atomic dipole is expressed (in the plateau region of the spectrum) as the sum of contributions corresponding to two dominant electron trajectories in the continuum. Furthermore, the time interval between the emissions of light resulting from these two trajectories is of the order of 0.20 of an optical period; when convoluted with the analyzing filter, it leads to a duration of $\sim0.45T$. Although we are at the limit of the time resolution of our analysis, this result indicates that it is not possible to select only one electron trajectory by simply increasing the rate at which the ellipticity changes. For larger rates corresponding to $p>1/4$, the electron trajectories are strongly affected and the pulse amplitude decreases rapidly. The point corresponding to $p=1/2$ is not reproduced in Fig. 4 because the ellipticity changes too quickly in comparison to the time resolution of the analysis.

In this paper, we have analyzed the harmonic generation by a single atom exposed to an oscillating field whose polarization depends on time. Since harmonics are emitted when the field is essentially linearly polarized, we expect that a short pulse of harmonics may be emitted if the time interval during which the field is linearly polarized is sufficiently short. We have shown that it is actually possible for the atomic system to emit an ultrashort pulse of a *given* harmonic, whose time duration is of the order of the optical period. It is important to stress that this short pulse duration can be controlled externally. All the calculations presented here are based on the numerical solution of the time-dependent Schrödinger equation and on a generalization of Lewenstein's model.

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[14] Ph. Antoine, A. L'Huillier, M. Lewenstein, P. Salières, and B. Carré, Phys. Rev. A **53**, 1725 (1996).

[15] The field is linearly polarized every $1/4p$ of an optical period and in good approximation, the ellipticity changes from -0.1 to 0.1 within 1/10 of the time needed for the field polarization to evolve from σ^+ to σ^- circular polarization.