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Time profile of harmonics generated by a single atom in a strong electromagnetic field

Philippe Antoine and Bernard Piraux
Institut de Physique, Université Catholique de Louvain, 2, Chemin du Cyclotron, B1348 Louvain-La-Neuve, Belgium

Alfred Maquet
Laboratoire de Chimie Physique—Matière et Rayonnement, Université Pierre et Marie Curie, 11, rue Pierre et Marie Curie, 75231 Paris Cedex 05, France

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We show that the time profile of the harmonics emitted by a single atom exposed to a strong electromagnetic field may be obtained through a wavelet or a Gabor analysis of the acceleration of the atomic dipole. This analysis is extremely sensitive to the details of the dynamics and sheds some light on the competition between the atomic excitation or ionization processes and photon emission. For illustration we study the interaction of atomic hydrogen with an intense laser pulse.

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It is now well established that an atom exposed to an intense laser pulse can emit high order harmonics of the driving force. At high field intensity and for a wide range of frequencies, all harmonic spectra share the same qualitative behavior characterized by the existence of a plateau. Until now, the main effort has focused on the study of the overall behavior of the harmonics spectra at high intensity and low frequency, where most of the experiments have been performed. It has been surmised by means of a semiclassical model that in this regime, the highest harmonics of the plateau are produced by electrons which, after tunneling out, return to the vicinity of the ion with a very high kinetic energy [1].

So far, however, no attempt has been made to analyze systematically the dynamical aspects of the harmonic emission process [2]. It is indeed natural to wonder when the emission of a given harmonic occurs during the interaction of the atom with the pulse and whether or not there is a time interval (which corresponds to some intensity range) in which a given harmonic is preferably emitted. The answer to these questions may be obtained by means of a so-called time-frequency analysis (either Gabor's or wavelet) of the acceleration of the atomic dipole. As we show, this type of analysis reveals itself to be extremely useful; it provides new information not only about the fundamental mechanism that leads to harmonic generation, but also about the competition between the process of photon emission and the process of excitation or ionization.

In this paper, we study the time profile of the harmonics emitted by atomic hydrogen exposed to a strong laser pulse. For the sake of illustration, we consider two cases corresponding to two distinct physical situations. In the first case, which corresponds to a low frequency regime, hydrogen initially in its ground state interacts with a strong laser field whose frequency is such that five photons are necessary to ionize the atom. In the second case, hydrogen initially in the $2s$ state interacts with the same laser field. Hence, two photons are enough to ionize the atom; this corresponds to a somewhat higher frequency regime.

In order to study the interactions of atomic hydrogen with an intense laser pulse, we solve numerically the Schrödinger equation:

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

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

$$\mathbf{A}(t)=A_0 f(t)\sin(\omega t)\mathbf{e}_z \ ; \tag{2}$$

A_0 is the amplitude of the potential, $\mathbf{e}_z$ is the unit vector along the z axis, ω is the frequency of the laser field, and $f(t)$ is a slowly varying envelope assumed here to be Gaussian. The numerical procedure is the following: we first expand the wave function $\Psi(\mathbf{r},t)$ of the system in a basis of Coulomb-Sturmian functions [3] 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 then solved by means of a fully implicit Runge-Kutta method of order 7 [4]. Knowing the wave function $\Psi(\mathbf{r},t)$, it is then a simple matter to evaluate the acceleration $a(t)$ of the atomic dipole $d(t)$ along the z axis by means of Ehrenfest's theorem. The power spectrum is obtained by calculating the modulus square of the Fourier transform $\hat{a}(\omega)$ of $a(t)$.

By contrast to this standard Fourier method, a time-frequency analysis provides information about the time localization of a given frequency. This type of analysis consists of introducing a time-frequency transform, which depends on two adjustable parameters denoted by α and β; α refers to the frequency of the transform and β to its position in the signal. A general time-frequency transform has the following form:

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

where $T_{\alpha\beta}(t)$ is a window function oscillating at a frequency depending on α and centered around time β; $\overline{T_{\alpha\beta}(t)}$ denotes the complex conjugate of $T_{\alpha\beta}(t)$.

Among all time-frequency methods, two particular ones turn out to be very efficient in the present case: Gabor's transform and the wavelet transform [5]. The main difference

between the two is in the way the parameter α is used (in both cases, β is simply a time translation). In the Gabor analysis,

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

where $G(t)$ is a window function which, in the present case, is a Gaussian $e^{-t^2/2\sigma_0^2}$. $1/\alpha$ defines the frequency of the modulation, whereas the width of the analyzing function σ_0 and, therefore, the frequency width $\Delta\omega$ are fixed. In the case of a wavelet analysis,

$$T_{\alpha\beta}(t)=\frac{1}{\sqrt{\alpha}}W\left(\frac{t-\beta}{\alpha}\right), \tag{5}$$

where $W(t)$ is a windowed oscillating function. The action of α on $T_{\alpha\beta}(t)$ is a dilation when $\alpha>1$ or a contraction when $\alpha<1$ so that the relative bandwidth $\Delta\omega/\omega$ of $T_{\alpha\beta}$ is fixed. In other words, the shape of the analyzing function stays unchanged—it is simply spread out or squeezed. A usual choice for the oscillating function $W(t)$ is (Morlet's wavelet) [6]:

$$W(t)=e^{i\omega_0 t}e^{-t^2/2}, \tag{6}$$

where ω_0 is a fixed parameter.

Basically, the time-frequency transform is significantly nonzero at a given time, when, at that time, the signal oscillates at a frequency close to the frequency of the analyzing function $T_{\alpha\beta}(t)$. So the time-frequency analysis could be viewed as a filter, both in time and in frequency. Following the above discussion, we expect the wavelet analysis to be more adapted than Gabor analysis when the signal contains very high frequencies that appear during a time interval which is short as compared to the driving field period.

Let us now consider the interaction of atomic hydrogen initially in its ground state, with an intense laser pulse whose frequency is 0.118 in atomic units (3.21 eV) and the duration, i.e., its full width at half maximum (FWHM) 20 optical cycles. For a wide range of peak laser intensities I_0, the acceleration of the atomic dipole (not reproduced here) has a typical behavior as a function of time. It exhibits two distinct regions of fast oscillations: at the beginning of the interaction, well before the pulse has reached its maximum, and at the end of the interaction if the atom is not fully ionized. In fact, this second region of oscillations at the end of the interaction with the pulse occurs when the atom is left in a superposition of atomic states. Indeed, in the absence of a decay mechanism (in the present case, the coupling with the vacuum field), the atomic dipole keeps oscillating at frequencies depending on the populated atomic states. On the other hand, the fast oscillations of the atomic dipole at the beginning of the interaction with the pulse are at the origin of the harmonic generation. In Fig. 1, we show the harmonic spectra for two peak laser intensities I_0: 2×10^{14} and 2×10^{15} W/cm^2. We see that except for the first harmonic (the Rayleigh component), harmonics of low order are more pronounced in the lowest peak intensity case (2×10^{14} W/cm^2). The situation is different for the harmonics of high

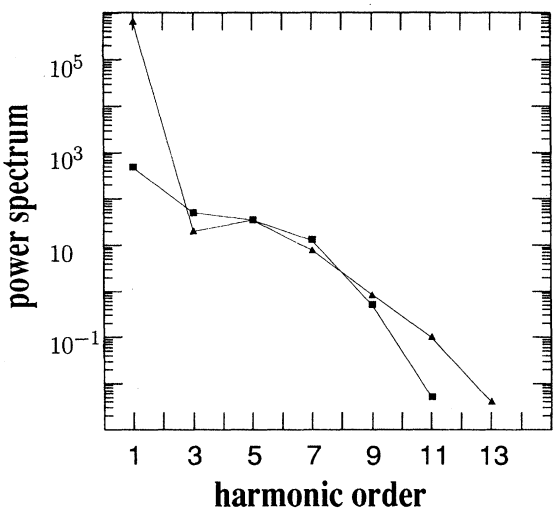

FIG. 1. Spectrum of harmonics produced by atomic hydrogen initially in its ground state and exposed to a laser pulse of frequency $\omega=0.118$ a.u. The pulse is Gaussian and has a FWHM of 20 optical cycles. Two peak laser intensities are considered: ■, $I_0=2\times10^{14}$ W/cm^2; ▲, $I_0=2\times10^{15}$ W/cm^2.

order (11 and 13, corresponding to a photon energy of 35 and 42 eV, respectively) which are more important when the peak intensity is the highest.

Let us examine the corresponding time profile [given by $\tilde{a}(\alpha,\beta)$, see Eq. (3)] of the harmonics as deduced from a Gabor time-frequency analysis. We see in Figs. 2(a) and 2(b)

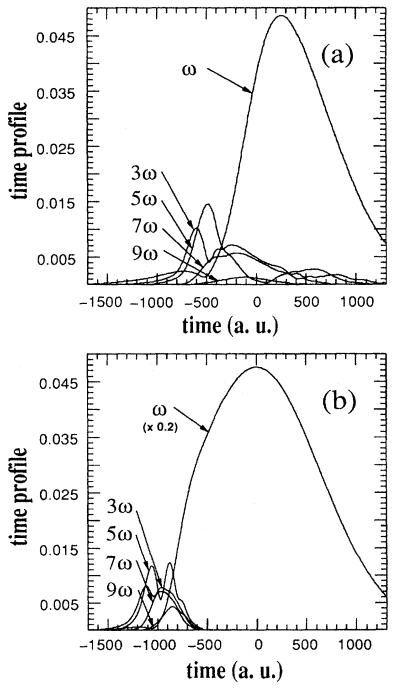

FIG. 2. Time profile [given by the coefficient $\tilde{a}(\alpha,\beta)$] of the odd harmonics (1–9) for the same cases as in Fig. 1. (a) $I_0=2\times10^{14}$ W/cm^2 and (b) $I_0=2\times10^{15}$ W/cm^2.

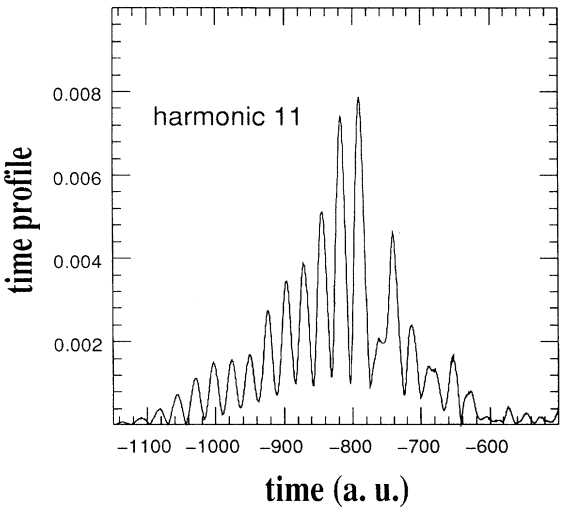

FIG. 3. Time profile [given by the coefficient $\tilde{a}(\alpha,\beta)$] of the 11th harmonic emitted by atomic hydrogen initially in its ground state and exposed to a Gaussian pulse of $I_0=2\times10^{15}$ W/cm^2, FWHM=20 optical cycles, and laser frequency $\omega=0.118$ a.u.

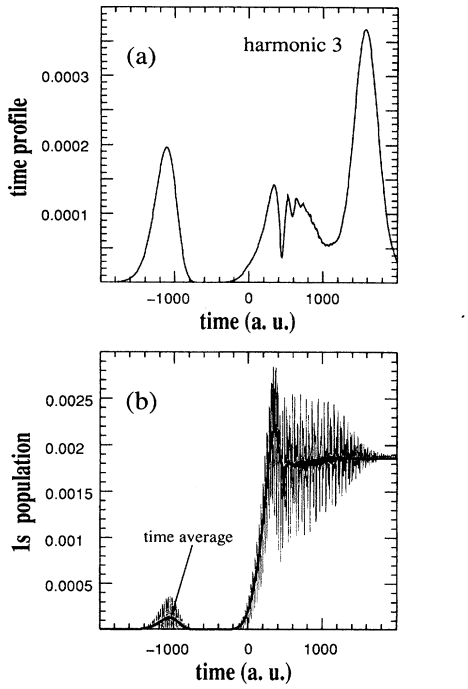

FIG. 4. (a) Time profile of the third harmonic emitted by atomic hydrogen initially in its $2s$ state and exposed to a Gaussian pulse of $I_0=2\times10^{14}$ W/cm^2, FWHM=20 optical cycles, and laser frequency $\omega=0.118$ a.u. (b) $1s$ population (i.e., the projection of the full wave function on the bare $1s$ state of atomic hydrogen) as a function of time in atomic units for the same case as in (a). The thick line represents the time average of the $1s$ population.

that high-order harmonics are emitted during a short time interval which moves toward the beginning of the pulse, well before its maximum when the peak laser intensity increases. It is interesting to note that recent experimental data in a lower frequency regime show the same trend [7]. In fact, we have checked that the emission process stops when most of the population is transferred from the ground state toward excited states (the atom does not need to be ionized). In other words, the time localization of the emission is associated with a strong space localization, as expected, since it is only close to the nucleus that the electron being accelerated is likely to emit light. In addition, it is clear from Fig. 2 that in both cases, each harmonic is emitted at a given time corresponding to a given intensity. The possible existence of a characteristic threshold intensity, associated with the emission of each harmonic, will be discussed in detail elsewhere.

Another interesting point is that as their order increases, the harmonics are emitted during shorter time intervals. For instance, when the peak field intensity is 2×10^{15} W/cm^2[see Fig. 2(b)], the fifth harmonic is emitted within roughly 800 a.u. ($\approx$19 fs), while the seventh lasts roughly 600 a.u. ($\approx$14.5 fs). If confirmed, this tendency indicates that the linewidth of higher harmonics should broaden with their order. This point will be also addressed in a forthcoming paper.

In order to follow the harmonic emission on a finer time scale, one has to use a wavelet analysis instead of Gabor's [8]. The results of such analysis are shown in Fig. 3, where the time profile of the 11th harmonic is given for the same pulse as in Fig. 2(b). It clearly appears that during about 450 a.u. ($\approx$ 11 fs), a train of subfemtosecond pulses of high frequency light ($\approx$35 eV) is emitted. It is interesting to observe that the emission of these pulses is periodic with frequency 2ω.

Let us now consider the interaction of a laser pulse with atomic hydrogen initially in its metastable state $2s$. The pulse has a duration of 20 optical cycles, its frequency is 0.118 a.u. (3.21 eV), and its peak intensity is 2×10^{14} W/cm^2. At this intensity, ionization is significantly suppressed because the system is coherently excited into a linear superposition of various Rydberg states (mainly $8p$, $9p$, and $10p$) which is stable against ionization [9]. As a result, the atomic dipole does not vanish at the end of the interaction with the pulse.

In addition to the odd harmonics, the spectrum contains in this case a large amount of hyper-Raman lines [10] whose origins may be traced back without particular problems. We refer to [11] for a discussion and, instead, focus our attention here on the time profile (Gabor analysis) of the third harmonic, which has a typical behavior. The results are presented in Fig. 4(a). By contrast to the previous cases, it is clear that the dynamics of harmonic emission are more complex as a consequence of the excitation of many atomic states. In particular, we note that the maximum present in Fig. 4(a) around 1300 a.u. of time is actually due to an atomic frequency which is quasidegenerate with 3ω and present during the free evolution of the dipole after the interaction with the pulse. More physical insight is gained by looking at the population dynamics; in Fig. 4(b), we show the time evolution of the $1s$ population (i.e., the projection of the total wave function on the bare atomic state). We observe that this $1s$ population is only significant during two distinct time intervals: first, around 20 optical periods ($\approx$1000 a.u.) before the maximum of the pulse, and again during the second part of the pulse. A more detailed analysis indicates that

the transfer of population towards the $1s$ state occurs concomitantly with a significant exchange of population between the $2s$ and the excited p states.

The fast oscillations (at 2ω) of the $1s$ population indicate that the total wave function oscillates very rapidly in a region of space very close to the nucleus. In Fig. 4(b), we also show the time average of the $1s$ population. The striking similarity that exists between this curve and the time profile of the third harmonic [see Fig. 4(a)] clearly demonstrates that the emission occurs only when the electron is close to the nucleus (as expected) and gets suppressed when the system is either in an excited state or in the continuum.

In this paper, we have studied the time dependence of harmonic generation. We have considered the case of atomic hydrogen in either the ground or the metastable $2s$ state in the presence of short pulses of an intense radiation field. Our motivation was to consider both low frequency regime [$H(1s)$, $\omega=0.118$ a.u.] and a (relatively) higher frequency regime [$H(2s)$, $\omega=0.118$ a.u.], the second case being known to exhibit dynamical suppression of ionization. Our results indicate that harmonic generation takes place during a short time interval, when the field reaches a critical intensity, which depends on the harmonic order. At higher intensities, i.e., when atomic excitation or ionization takes place, harmonic generation is suppressed. Moreover, the wavelet analysis indicates that harmonic emission can be periodic in time at a frequency 2ω. A similar analysis for atomic hydrogen initially in the $2s$ state shows that the time profile of harmonic emission exhibits a more complex behavior. This results from the fact that the atom is excited into a superposition of states. More detailed results on the dynamics of harmonic generation will be presented in a forthcoming paper.

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