Phase-Dependent Harmonic Emission with Ultrashort Laser Pulses

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We consider harmonic generation by atoms exposed to an intense laser pulse of a few femtoseconds. Our results, obtained by solving numerically the corresponding three-dimensional time-dependent Schrödinger equation, demonstrate the strong sensitivity of the harmonic spectra to the phase of the laser. These results are explained in terms of both quantum and classical dynamics. We show that this phase sensitivity may be exploited in order to probe the laser phase for ultrashort pulses. Our discussion about this new method of diagnosis takes into account propagation effects. [S0031-9007(98)06977-4]

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Recently, it has been demonstrated by groups at the University of Michigan [1] and at the Technical University of Vienna [2] that ultrashort pulses provide an efficient way to extend harmonic emission up to frequencies well into the so-called water window. The generation of radiation in this spectral region opens the route to future applications, in particular, in biology.

Using ultrashort pulses instead of many cycle pulses has several implications on the harmonic generation process. The main motivation resides in the saturation intensity which is higher when compared to longer pulses. In other words, atoms can survive to higher intensities before being ionized by the laser light. It explains why spectra obtained with short pulses extend farther than those with long laser pulses. Furthermore, starting with a driving laser pulse as short as a few cycles provides emission during a time interval of a few femtoseconds or even less. It is therefore a very attractive way to produce subfemtosecond harmonic pulses because this scheme should be much easier to implement than those based either on an external control of the harmonic emission by means of a time-dependent polarization [3] or on its temporal recompression by a pair of gratings [4].

Finally, a consequence of the ultrashort duration is the rapid variation of the laser intensity, which gives rise to new interesting effects [5]. These are precisely the content of the present Letter. We pay attention to pulses as short as a few cycles for which the laser intensity cannot be considered as a constant within one optical cycle. Theoretical study of the harmonic generation driven by such optical pulses is stimulated by the recent developments in laser technology. Lasers delivering a few optical cycle pulses currently operate in some laboratories [2]. From the theoretical point of view, it is, of course, out of the question to describe the harmonic process with a theoretical model assuming a slowly varying intensity. A fully nonadiabatic approach is required. One has to stress, however, that it does not mean that the semiclassical description [6] provides a wrong picture. On the contrary, its predictions are in very good agreement with our so-called exact model. In the semiclassical picture, the electron first escapes from the core by tunneling through the Coulomb barrier which is lowered by the strong laser field. Once free, it oscillates freely driven by the laser field. If it reencounters the core, it may recombine, emitting a harmonic photon. With a many cycle laser pulse, the maximum kinetic energy gained by the electron is $3.17U_p$, where U_p is the quiver energy of free electrons in an oscillating field. It defines the energy of the highest harmonic of the spectrum to be $I_p + 3.17U_p$, where I_p is the ionization potential. It is worth stressing that the main part of the dynamics of the harmonic emission process is directly determined by the adiabatic phase accumulated by the electron on its trajectory in the continuum. Recently [7], it has been demonstrated that all experimental results obtained with pulses as short as 27 fs can be explained in terms of the intensity variation of the adiabatic phase. A change in laser intensity on the time scale of the optical period alters significantly the harmonic generation process and leads to additional effects.

In this Letter we discuss the most interesting of these effects, namely the sensitivity of the harmonic spectra to the phase of the laser. Depending on this phase, the harmonics in the cutoff region are resolved or not resolved. In addition, the position of the cutoff itself varies and the so-called "plateau" region exhibits two well distinct parts. In the low-frequency part, the spectrum consists of a series of (shifted) well defined harmonics while, in the high-frequency part, the spectrum is composed of a series of broad but well resolved peaks which are no longer separated by twice the laser frequency and whose amplitude changes with the phase. As a result, the amplitude of a given harmonic may change dramatically with the phase up to 3 orders of magnitude depending on the case. This result which does not depend on the pulse shape may be exploited in order to actually probe the phase of the laser in a single shot experiment.

We consider the interaction of atomic hydrogen with an ultrashort laser pulse. The whole information about harmonic generation is contained in the time-dependent dipole acceleration. This quantity is obtained "without approximation" by solving numerically the time-dependent Schrödinger equation (TDSE) in the velocity form [8]. We define both the intensity and the duration (FWHM of the time-dependent intensity) of our ultrashort laser pulses from the time-averaged magnitude of the Poynting vector (see [9]). For numerical reasons, the electric field $\vec{E}(t)$ is calculated from the expression of the vector potential given by $\vec{A}(t) = \vec{A}_0 f(t) \sin(\omega t + \phi)$, where $f(t)$ is the envelope and ϕ is the phase of $\vec{A}(t)$ at time $t = 0$ when $f(t)$ is maximum. Note that both the energy and the duration of the laser pulse do not depend on this phase ϕ. Various choices for $f(t)$ have been used (Gaussian, even power of sine, etc.). They all lead to very similar results provided that both the peak intensity and the duration are the same. All the results presented here have been obtained with a sin square envelope for $\vec{A}(t)$. The dipole acceleration is "Fourier transformed" to get the spectrum and a time-frequency analysis (Gabor) provides the time profile of the harmonics [10]. We consider pulses whose duration is 2.7 and 5.4 fs at a wavelength of 800 nm. We present results for two laser phases: $\phi = 0$ and $\phi = \pi/2$. In the first case, the electric field reaches its maximum at the maximum of the envelope ($t = 0$), while in the second case, the field is zero.

In Fig. 1, we show two harmonic spectra, one for each laser phase, obtained with a pulse whose duration is 5.7 fs and a peak intensity of 5×10^{14} W/cm^2. In the cutoff region, we see that the harmonics are not resolved any more for $\phi = 0$ by contrast to the $\phi = \pi/2$ case. This difference is understood by comparing the time profile of the 67th harmonic (which belongs to the cutoff) for both phases. The latter one, shown in Fig. 2, reveals only one single relevant emission (during a time interval of 500 as) in the $\phi = 0$ case, whereas for $\phi = \pi/2$, the harmonic is emitted twice. So, by relating the half optical cycle periodicity of the emission to the 2ω structure of the harmonic spectrum, we easily understand why harmonics in the cutoff are not resolved in the $\phi = 0$ case [11]. Phase effects are even more striking for a pulse whose duration is 2.7 fs and a peak intensity of 3.5×10^{14} W/cm^2. In Fig. 3, a redshift of the cutoff position which is strongly phase dependent is observed: when $\phi = \pi/2$ (Fig. 3a), the cutoff occurs at the 47th harmonic and at the 57th harmonic when $\phi = 0$ (Fig. 3b). This contrasts with the prediction of the semiclassical cutoff law (51st harmonic), the peak intensity corresponding to the maximum of the magnitude of the time-averaged Poynting vector. It is also important to notice the significant difference (1 order of magnitude) in the highest harmonic efficiency between the two cases. Finally, the most remarkable effect due to the phase is the apparent new periodicity which occurs at the end of the plateau when $\phi = 0$. In fact, a very sharp transition occurs in the spectrum at about the 31st harmonic. Above this frequency, the spectrum exhibits a series of broad peaks well defined but which are not separated by 2ω any more. By contrast, this 2ω periodicity is preserved below this transition frequency. This apparent new periodicity may be understood by looking at the 43rd harmonic time profile (see Figs. 4c and 4d): the emission occurs 2 times within only one-half of an optical cycle; the time interval between both maxima is roughly equal to a 0.2 period. The inverse of this time defines the distance between the broad peaks in the frequency spectrum. The same Gabor analysis below the transition frequency (at about the 27th harmonic) leads to the well known half optical cycle periodicity of the emission for the harmonics in the plateau.

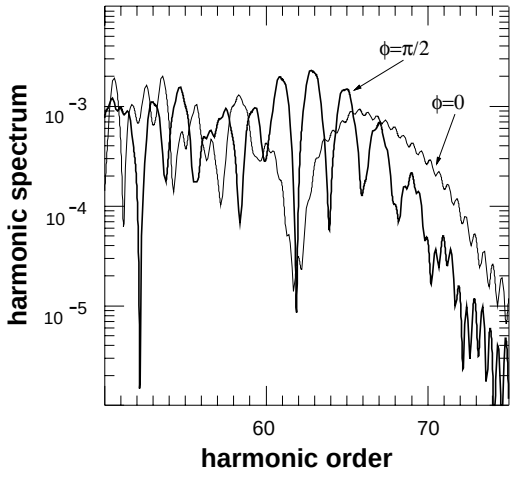

FIG. 1. Harmonic spectrum at 800 nm for a laser pulse whose duration is 5.7 fs and a peak intensity of 5×10^{14} W/cm^2.

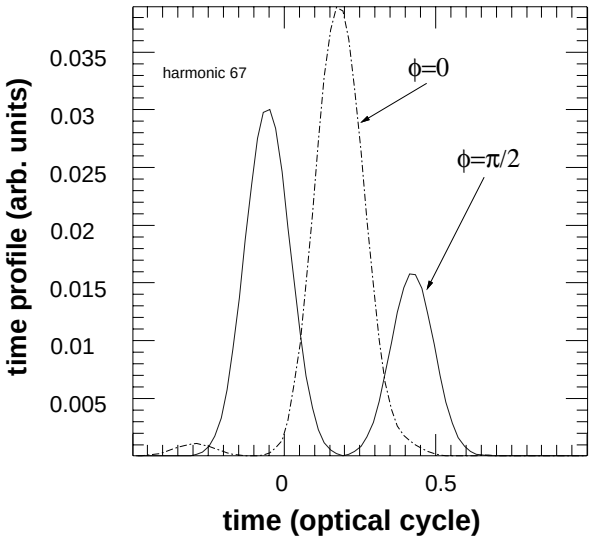

FIG. 2. Time profile of the harmonics at about the 67th one for the same case as in Fig. 1.

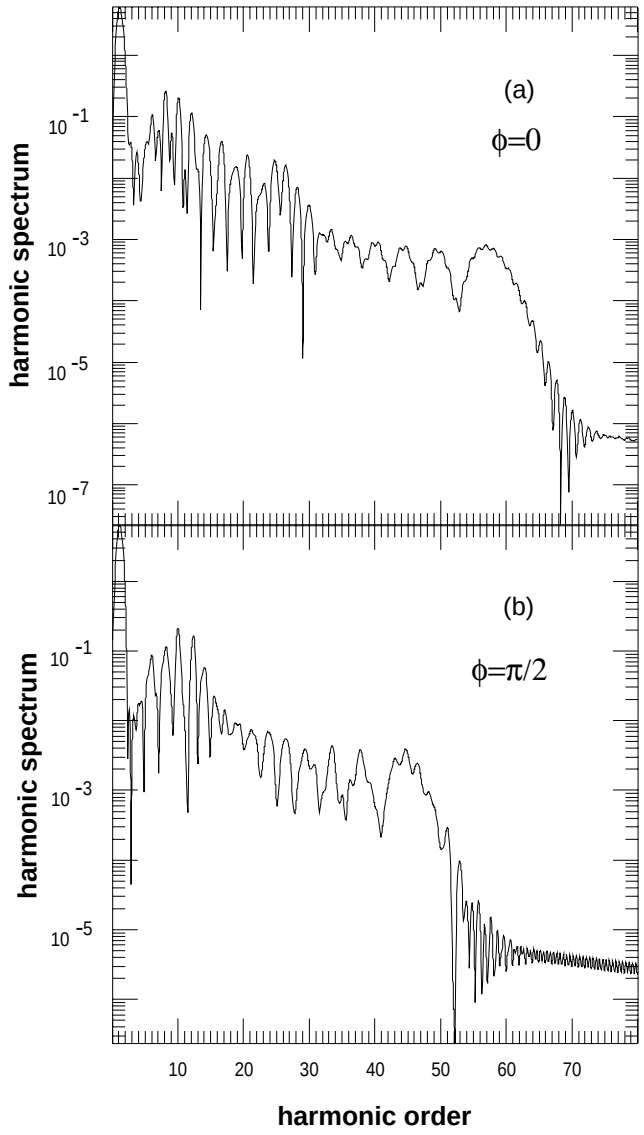

FIG. 3. Harmonic spectrum at 800 nm for a laser pulse whose duration is 2.4 fs and a peak intensity of 3.5×10^{14} W/cm^2.

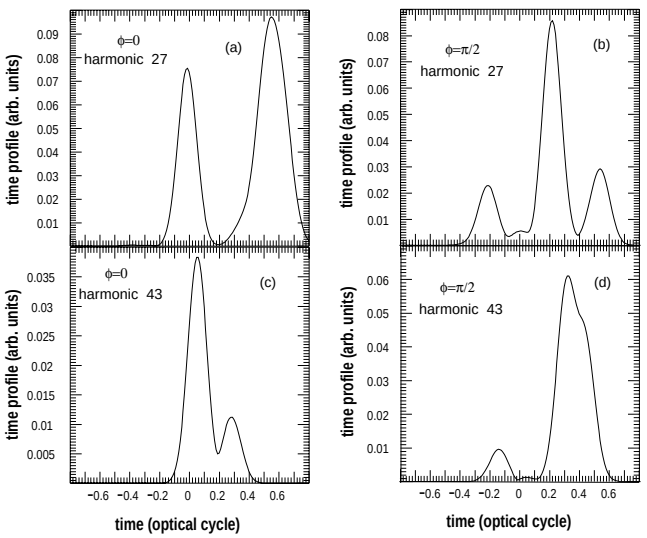

FIG. 4. Time profile of the harmonics at about the 27th one and the 43rd one for the same case as in Fig. 2.

It is known that the fully quantum theory of a high-harmonic generation [12] recovers the classical interpretation [6]. Even in the case of an ultrashort pulse, this classical interpretation allows one to explain the characteristic behavior of the above time profiles and the existence of the transition frequency. According to the classical interpretation, the electron is emitted in the external field at the origin with zero velocity at time t_0 (ionization time). Under the influence of the external field, the electron may come back to the origin at time t (recombination time). The kinetic energy of the fastest electrons determines the frequency of the highest harmonics which are emitted at time t (also called the emission time). This kinetic energy is estimated by solving the corresponding Newton's equation with the relevant boundary conditions. Therefore, a plot of the kinetic energy as a function of the emission time provides information on the frequencies which are emitted and on the time when they are emitted. Besides, the ionization time t_0 as a function of the emission time t provides information on the efficiency of the emission since the latter one increases with the ionization probability which in turn depends on the electric field at time t_0.

Our results for the kinetic energy and the ionization time t_0 as a function of the emission time t are presented in Fig. 5 for a pulse whose duration is 2.7 fs and a peak intensity of 3.5×10^{14} W/cm^2. The kinetic energy is expressed in units of the ponderomotive potential U_p given by $E^2/4\omega^2$. The ionization and the emission times are expressed in units of the laser period. For $\phi = 0$ (Fig. 5a), the maximum kinetic energy equals $2.5U_p$ (which corresponds to the 57th harmonic) while, for $\phi = \pi/2$ (Fig. 5b), this maximum kinetic energy is only $2.2U_p$ (which corresponds to the 47th harmonic). This result for the cutoff frequencies in the harmonic spectrum is in perfect agreement with those presented in Fig. 3. In the $\phi = \pi/2$ case, the kinetic energy profile consists of two broad peaks separated by about half of an optical period. As a result, we recover half of an optical cycle period even for the highest frequencies in the spectrum. By contrast, in the $\phi = 0$ case, this periodicity is absent for frequencies above $0.9U_p$. Figure 5a shows that two electron trajectories contribute to the emission within this frequency range, in agreement with Fig. 4c. In addition, we clearly see that the time interval between the two emissions increases with decreasing kinetic energies. This explains why the frequency interval between two adjacent peaks at the end of the plateau increases with the frequency (see Fig. 3a). In Fig. 5a, the height of the second peak defines the position of the transition frequency in the spectrum. In the $\phi = \pi/2$ case, the kinetic energy profile (Fig. 5b) clearly shows the

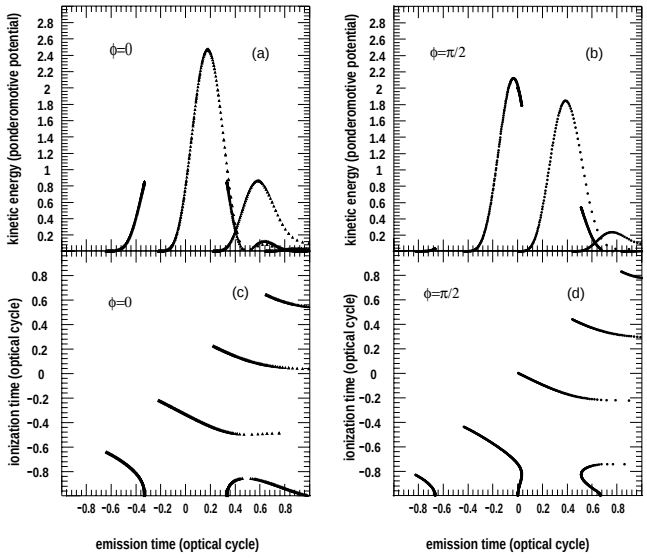

FIG. 5. Kinetic energy and ionization time as a function of the harmonic emission time for the same case as in Fig. 2.

existence of three dominant trajectories leading (without restriction on the frequency) to three peaks in the time profile (Figs. 4b and 4d). From the classical ionization times, we also understand the relative magnitude of the peaks in the time profile of a given harmonic. For instance, in Fig. 4c, the first peak in the time profile of the 43rd harmonic for $\phi = 0$ is dominant because it comes from the recombination of electrons which are ionized when the field is important (equal to its half maximum). The same argument may be used to explain why in Fig. 3 the second part of the plateau is higher for $\phi = \pi/2$ than for $\phi = 0$.

The laser pulses used in the Vienna experiment [2] have a duration of about two optical cycles. It turns out that, in this case, the key parameter for optimizing the x-ray source for maximum brightness at high photon energies is the phase ϕ of the laser field. However, this phase which affects the x-ray flux in the cutoff region is not controlled in the experiment. It is therefore of great interest to be able to probe this phase ϕ. The present results about the phase sensitivity of the harmonic spectra demonstrate that it is indeed possible to develop such a diagnosis method. The behavior of a given harmonic as a function of the phase ϕ is very characteristic. In the cutoff region, the intensity of the harmonics is maximum for $\phi = 0, \pi$ and has a deep minimum (3 orders of magnitude lower) for $\phi = \pi/2$. In the high frequency part of the plateau, the intensity of the harmonics as a function of ϕ presents a similar dependence with a very deep minimum around $\phi = \pi/3$. Again, there are more than 3 orders of magnitude between this minimum and the maximum at about $\pi/2$. These results do not depend significantly on the pulse shape provided that both the pulse duration and the peak intensity are the same (in an experiment, the phase determination has to be correlated with the peak intensity measurement). Therefore, it exists as a relation between the phase ϕ and the number of photons emitted within a narrow frequency bandwidth. This method is efficient provided that the two following conditions are fulfilled. First, the number of photons must be high enough because the phase ϕ has to be estimated in a single shot experiment. This condition is satisfied if we extrapolate the results from Ref. [2] where they observe about 10^3 photons per shot in a 10% bandwidth at 0.3 keV. Second, the phase dependence must be preserved after propagation. Indeed, focusing the laser beam introduces a geometrical phase which may wash out the expected effect. However, a gas jet located well after the focus (3 or 4 times the confocal parameter) ensures that the geometrical phase stays constant longitudinally across the atomic beam. We have checked that the intensity and phase distribution in the plane perpendicular to the laser axis do not significantly affect the present conclusions. This has been performed by averaging the single atom response over the electric field distribution in the transversal direction; this approximates the macroscopic response because there is no longitudinal variation of the electric field along the gas jet. Although the intensity is much lower than the peak intensity at the focus, it is not a practical restriction because our diagnosis method does not require such high intensities and because, in this geometry, very good phase matching conditions are achieved [13].

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