# Harmonic generation by laser-driven classical hydrogen atoms

Gunadya Bandarage
*Department of Chemistry, Texas A&M University, College Station, Texas 77843-3255*

Alfred Maquet, Thierry Ménis, Richard Taïeb, and Valérie Véniard
*Laboratoire de Chimie Physique, Université Pierre et Marie Curie, 11 Rue Pierre et Marie Curie, F75231 Paris CEDEX 05, France*

J. Cooper
*Joint Insitute for Laboratory Astrophysics and National Institute of Standards and Technology, University of Colorado, Boulder, Colorado 80309-0440*
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We present the results of our investigations of the dynamics of a classical hydrogen atom submitted to an intense classical radiation field with a relatively low frequency as compared to the characteristic Kepler frequency of the atomic system. We discuss here the harmonic spectra which can be deduced from a Fourier analysis of the time evolution of the strongly driven atomic dipole, associated either with a single trajectory or with ensembles of trajectories evolving from an initial microcanonical distribution. The spectra obtained from ensemble-averaged atomic dipoles qualitatively reproduce the main features of those recently observed in experiments and allow, in particular, discussion of the occurrence of a plateau in the distribution of harmonic intensities as a function of their order. The possible competition between harmonic generation and multiphoton ionization, which both take place in the same laser-intensity range, is also discussed.

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## I. INTRODUCTION

High-order harmonic generation by atomic systems submitted to intense laser fields has now been observed in several different experiments [1–3], and general features of the harmonic spectra seem to be independent of the particular atom probed. Among these features, the presence of a plateau in the distribution of the harmonics as a function of their order has aroused interest, since its existence opens the possibility to generate, from an intense laser source, significant numbers of high-overtone photons. Although the presence of a plateau has been reproduced in several model calculations [4–11], no clear picture emerges with regard to its origin, position, and width. This raises questions which are still under active discussion [12].

The main purpose of this paper is to report our investigations of harmonic generation by classical anharmonic systems, driven by a strong, single-mode and low-frequency laser field. We begin by considering the case of a classical hydrogen atom (a preliminary account of this work has already been presented as a Rapid Communication; see Ref. [8]). We shall show that a detailed study of this model can help to shed some light on the above-mentioned features of the harmonic spectrum. As an interesting by-product it provides a clear picture of the possible link existing between harmonic generation and the competing process of multiphoton ionization, which is known to take place in the same laser-intensity range.

Among the findings derived from the classical model, inspired from the Monte Carlo classical trajectory method, used more than ten years ago by Leopold and Percival [13] to deal with the microwave ionization of Rydberg atoms, is the possible connection between the position of the plateau and the (shifted) value of the characteristic Kepler "atomic" frequency of the atom in the presence of the external field. The width of the plateau is likely to be associated with the presence of "satellite" frequencies, shifted from the atomic one by integer numbers of the laser frequency itself. Within this picture, the origin of the plateau appears to be linked to the fact that the shifted atomic frequency can become quasiresonant with some harmonic of the laser, enhancing the intensity of the harmonic, as well as the ones of its neighbors. Another aspect of the physics provided by the classical model is a clear picture of the fact that, for individual trajectories, whenever ionization takes place harmonics are no longer generated, as the outgoing asymptotically free electron can only experience Thomson scattering, i.e., scatter the fundamental laser frequency itself. This prediction, which has been further confirmed by a wavelet analysis of the time-dependent atomic dipole [14], suggests that harmonic generation and multiphoton ionization are competing processes and that at higher laser intensities, when the atomic sample is fully ionized, harmonics cease to be emitted.

The limitations of a classical approach are obvious: atoms are quantal objects and classical mechanics cannot properly account for the presence of resonances in atomic spectra. It is also difficult to discuss the concurrent process of ionization in its finer details, such as the aspects connected to the so-called above-threshold ionization [5], and the resonant effects observed in ionization probabilities [15,16]. However, while precise quantitative predictions cannot be inferred from such an analysis, classical mechanics remains a convenient tool to discuss general features of an otherwise complicated process, as demonstrated when applied to the microwave ionization of Rydberg atoms. Within this context, we have chosen to discuss the case of laser frequencies $\omega_L$ relatively small with respect to the characteristic Kepler frequency $\omega_0$ associated with the free atomic motion, namely $\omega_L \leq 0.15\omega_0$. This choice is motivated by the fact that high-order harmonic generation has precisely been observed in this frequency range. We will also consider relatively small laser intensities typically $F \leq 0.15$ a.u. (atomic unit of field strength intensity: $F_0 \approx 5.14\times10^9$ V/cm). This low-frequency regime partially overlaps the semiclassical one, in which the predictions of the classical approach agree fairly well with the experiments for ionization, with the notable exception of the possible occurrence of resonances which can significantly enhance the ionization yield [15]. As we shall argue below, these discrepancies are not likely to modify appreciably the harmonic spectra.

It is not clear if chaotic dynamics leading to ionization plays an important role in this (frequency and intensity) domain. It seems that it influences, at least indirectly, the harmonic spectra which are less intense and, ultimately, disappear at higher field intensities. Starting from moderate intensities, one observes that the system generates harmonics, whose intensities and number grows steadily with the field amplitude. This indicates that the atomic motion remains dominantly bounded and quasiperiodic. As the field intensity increases, however, one observes that ionization can occur, the time at which it takes place being extremely sensitive on the initial conditions for an individual trajectory. At the same time, the spectra associated to the bounded part of the trajectory become extremely noisy, another indication of the possible role of chaos in the dynamics of the atom-laser interaction. At still higher intensities harmonics are no longer generated, as the motion of the asymptotically free photoelectron becomes perfectly periodic in the field.

With the limitations of the classical model in mind [15–17], we shall present our results along the following lines: Section II will be devoted to a brief presentation of the formalism and of the model used in our work. The dynamics of single trajectories as well as their power spectra will be investigated in Sec. III, as a function of the laser field intensity, thus providing some insight into the origin of harmonics and their intertwining with the survival of atomic frequencies. In Sec. IV, we shall present harmonic spectra generated by ensembles of atoms, initially in a microcanonical distribution of states. This step is necessary to compare our results with those observed with spherically symmetric (quantal) atomic targets [13]. The results of this analysis, as well as their physical implications, shall be briefly discussed in Sec. V.

## II. MODEL AND CALCULATIONS

In the following, we will consider a classical hydrogen atom, with an infinite-mass nucleus fixed at the origin of coordinates, interacting with a strong electromagnetic field with frequency $\omega_L$, linearly polarized along the $z$ axis and whose onset is at $t=0$. The Hamiltonian of the system is thus (atomic units will be used throughout the paper, unless otherwise mentioned):

$$H=H_0=p^2/2-1/r, \quad t<0\ , \tag{1a}$$

$$H=H_0-zF(t)\sin\omega_L t, \quad t\geq 0\ , \tag{1b}$$

where the time-dependent amplitude $F(t)$ of the field can model the envelope of a laser pulse: We have chosen to discuss here the somewhat idealized case of a linear ramp, associated with the laser turn-on, followed by a constant amplitude extending over many cycles of the field. By changing the slope of the ramp one can easily investigate the possible influence of the laser turn-on. This point, as well as the negligible importance of the detailed shape of the ramp, is discussed below.

In order to model a hydrogen atom initially in its ground state, we have chosen initial Kepler trajectories with a fixed energy $E_0=-\frac{1}{2}$ a.u. The corresponding Kepler (or atomic) frequency is $\omega_0=1$ a.u., while the corresponding Kepler (or atomic) period is $T_0=2\pi$ a.u. (1 atomic unit of time $\approx 2.42\times10^{-17}$ s). Once initial coordinates $\mathbf{r}_0$, $\mathbf{p}_0$, compatible with the condition ${p_0}^2/2-1/r_0=-\frac{1}{2}$, have been chosen, one determines the trajectory of the electron in the combined fields of the nucleus and of the laser, by solving Hamilton's equations of motion for $t\geq 0$:

$$\dot{r}_i=\frac{\partial H}{\partial p_i}\ , \tag{2a}$$

$$\dot{p}_i=-\frac{\partial H}{\partial r_i}, \quad i=x,y,z\ , \tag{2b}$$

where $H$ is given by Eq. (1b). These six coupled differential equations have been numerically integrated by using standard routines with variable steps, either fourth-order Runge-Kutta or Bulirsch-Stoer [18]. We have found it convenient to use variable-step integration schemes both for aspects of numerical accuracy and computing time: some trajectories have high eccentricities, giving rise to strong accelerations, which can render fixed-step integration routines very inaccurate. Note also that we have not found any particular difficulty in implementing the numerical routines, in spite of the fact that we have used Cartesian coordinates which are known to lead to difficulties in the one-dimensional model, owing to the presence of the Coulomb singularity.

Having determined the actual trajectory of the electron, one can easily obtain the projection $\mu_z(t)$ of the time-dependent atomic dipole along the laser polarization, a projection which coincides with the $z$ component of $\mathbf{r}$. The spectrum of the light emitted by the forced dipole is then straightforwardly obtained from its power spectrum:

$$D(\omega)=\lim_{T\to\infty}\frac{1}{T}\left|\int_0^T \mu_z(t)e^{i\omega t}dt\right|^2\ , \tag{3}$$

valid for stationary correlation functions of the dynamical variable $z(t)$ [19], a condition we shall assume to be met here, as we will discuss the case of constant-amplitude laser pulses and exclude the contributions of the ramp from the Fourier analysis. With regard to the latter we have used standard fast-Fourier-transform (FFT) routines, windowed where appropriate [18]. In order to implement the method, it was necessary to sample the values of $z(t)$ at regular intervals, which obliged us to modify the variable-step integration routine, to force it to stop at the abscissa values needed for the FFT. We have found that such a modified variable-step scheme was still faster and more accurate than fixed-step ones. We used typical window widths of around 30 field periods, which was large enough to get stable power spectra. We note that on such a relatively short time interval, the proportion of ionizing trajectories was kept at a relatively low level, much lower in fact than the one reported in Ref. [15], in which trajectories were followed for up to 300 field periods. We have checked also that, when considering averaged dipoles, no significant changes were found when interchanging the order of averaging and integration.

In general, single Kepler trajectories lack the inversion center of real atoms which are spherically symmetric. As a consequence, the harmonic spectra obtained from a single-trajectory analysis exhibit (unphysical) even harmonics. A natural way to remedy such a difficulty is to consider an ensemble of trajectories, evolving from an initial microcanonical distribution. To this end, we have used the Monte Carlo classical trajectory method [13,20] to define an initial microcanonical ensemble of randomly chosen Kepler orbits with energies $E_0=-\frac{1}{2}$ a.u., thus retrieving the spherical symmetry of a real ground-state hydrogen atom. In the actual implementation of the method we have found it convenient to use astronomical coordinates, which are well suited for the Kepler problem [21]. As we shall show below, the analysis of such atomic dipoles, averaged on large enough sets of randomly chosen trajectories, eliminates the unphysical even components in the harmonic spectra. Nevertheless, we will first discuss the analysis of single trajectories, which provides interesting insights into the dynamics of harmonic generation.

## III. SINGLE-TRAJECTORY ANALYSIS

In the following we shall assume that a laser field, with frequency $\omega_L=0.086$ a.u. ($\approx 2.34$ eV, second harmonic of the Nd laser), is turned on at $t=0$, with a linear ramp ending at $t_1\approx 300$ a.u., i.e., with a field amplitude of the form

$$F(t)=\begin{cases}F_0 t/t_1, & 0\le t\le t_1,\\ F_0, & t>t_1\ .\end{cases} \tag{4}$$

A typical short-time dependence of the corresponding laser-driven atomic dipole, for $F_0=0.12$ a.u., is shown in Fig. 1(a). One observes that the atomic dipole, which is nonzero at $t=0$, is initially oscillating at the Kepler frequency $T_0=2\pi$ a.u. and then, as the laser intensity grows, begins to experience modulations at the laser period $T_L=2\pi/\omega_L\approx 73$ a.u. For this trajectory, as the field amplitude remains constant, the evolution of the dipole follows a regular pattern with both high- and low-frequency components; see Fig. 1(b). Such patterns are independent of the initial conditions chosen at $t=0$. However, as we discuss below, it may happen that the atom ionizes, which strongly modifies the subsequent time evolution of the dipole.

Before discussing the harmonic spectra associated with such oscillating dipoles, it is instructive to consider first the Fourier analysis of the dipole of a pure Kepler trajectory: This Kepler power spectrum consists of regularly decreasing peaks located at frequencies multiples (harmonics) of Kepler's, i.e., at $\omega_n=n\omega_0$, $n=1,2,3,\ldots$; see Fig. 2(a).

The power spectra computed from laser-driven atomic dipoles, for $t>t_1$ and for a large number of laser periods, are modified, with respect to the field-free case, in a way which strongly depends on the laser amplitude $F_0$. This point is illustrated in Figs. 2(b)–2(d), where we show the power spectra of typical trajectories, with the same initial conditions, and increasing field amplitudes $F_0$ comprised between 0.005 (low-intensity regime) and 0.14 a.u. (high-intensity regime). Note that these spectra correspond to trajectories which do not ionize over a quite large number ($\approx 30$) of laser periods: in the above range of intensi-

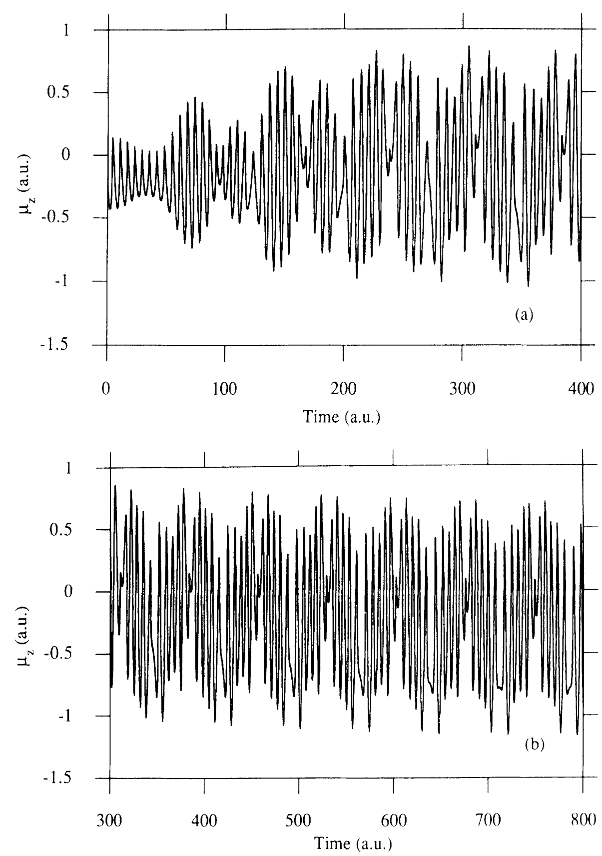


FIG. 1. Temporal variations of the atomic dipole moment $\mu_z$ of a trajectory along the laser polarization direction: $\omega_L=0.086$ a.u., $F_0=0.12$ a.u. (a) corresponds to the turn-on of the laser field, while for (b) the field amplitude remains constant.

ties few trajectories lead to ionization, at least when the initial conditions are chosen at random.

In addition to the multiples of the Kepler frequency the spectra present a dominant line at the laser frequency $\omega_L$, which corresponds to the Rayleigh component in the light scattered by the atom. Harmonics ($\omega_{L,n}=n\omega_L$, $n=2,3,\ldots$) of the laser do appear, their number and intensities growing with the field amplitude. As already mentioned, even harmonics are also present, as a result of the fact that general Kepler orbits with nonzero eccentricities have no inversion center. As expected, at low laser intensities [Figs. 2(b) and 2(c)], corresponding to a perturbative regime, the harmonic intensities decrease steadily with the order (note the logarithmic scale on the figures). However, if one considers higher field amplitudes, Fig. 2(d), more harmonics are present in the spectra and their relative intensities cease to decrease with the order, which gives rise to a plateau, or even a bump, in the spectrum (see also below).

Another interesting feature of these spectra is the presence of the atomic Kepler frequency $\omega_0$ ($=1$ a.u. $\approx 11.63\omega_L$) which is still present and displays, in addition, a number of satellites separated from it by integer numbers of laser frequency: $\omega_{0,n}=\omega_0 \pm n\omega_L$, $n=1,2,3\ldots$. The number of these satellite lines, which would correspond to Raman components in the light scattered from the atom, grows with the laser amplitude and their relative intensities follow an approximately bell-shaped distribution centered on $\omega_0$. We note that the presence of a damping term in our equations of motion would lead, after a long enough time, to the suppression of these atomic components. However, within the framework of the classical model adopted here there is no natural way to introduce such a term. We have checked, in particular, that the classical radiation reaction would produce only negligible changes on the time scale considered here, when proceeding to the Fourier analysis of our trajectories [22].

It should be noted also that, due to the presence of the external oscillatory field, the periods of orbits change and the lines identified above as atomic Kepler frequencies in Figs. 2(b)–2(d), are in fact *shifted* with respect to the corresponding field-free ones. They have frequencies $\tilde{\omega}_0 \neq \omega_0$, which depend mainly on the laser frequency and intensity, on the one hand and, as discussed above, slightly on the trajectory's initial conditions, on the other hand. This holds also for the satellite lines which are in fact located at frequencies $\tilde{\omega}_{0,n}=\tilde{\omega}_0 \pm n\omega_L$. Apart from the above-mentioned variations dependent on the initial conditions, this (ac Stark) shift grows with the laser inten-

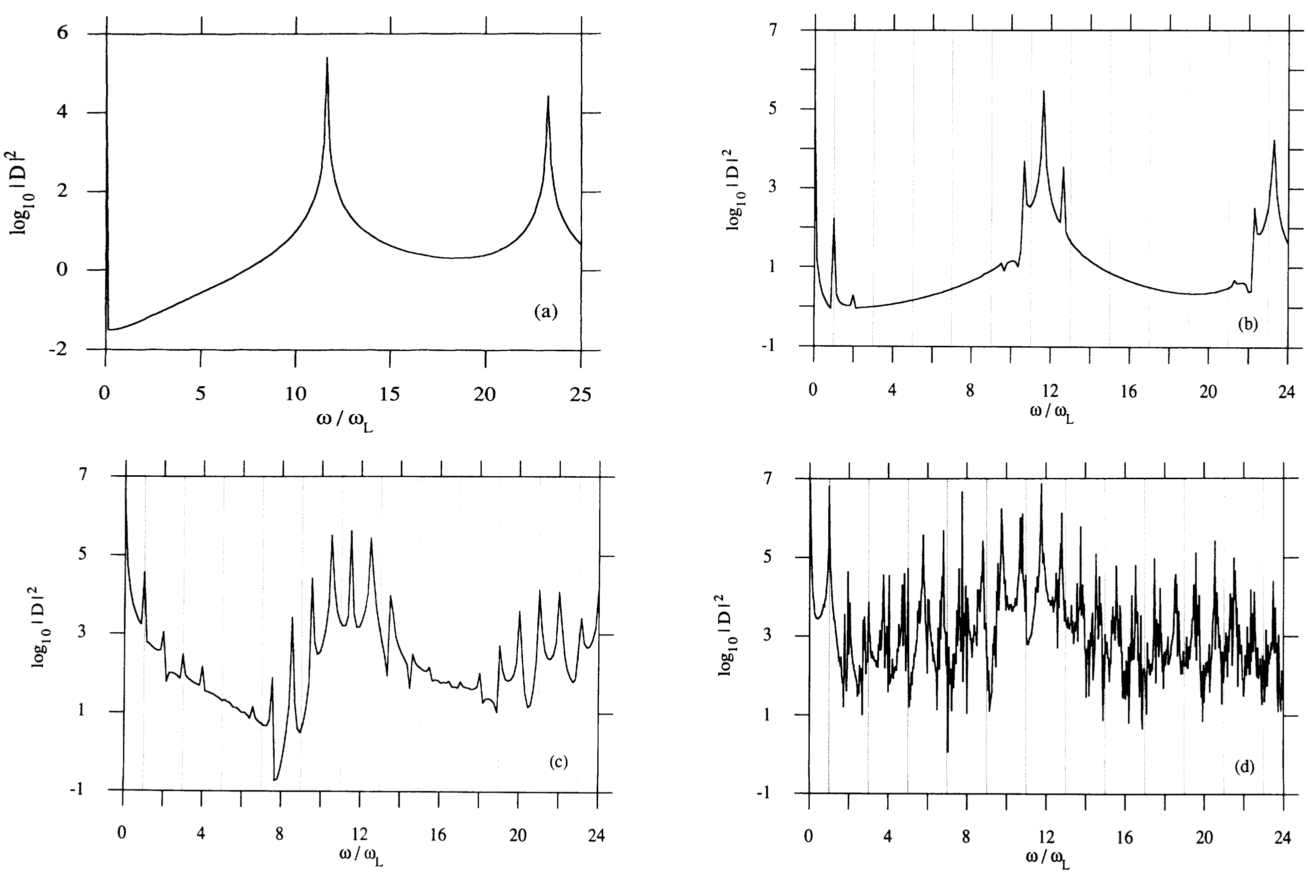


FIG. 2. Power spectra $D(\omega)$ of the dipole $\mu_z$ for typical trajectories. (a) corresponds to a pure Kepler trajectory (field-free case) and (b), (c), and (d) are associated to laser-driven atomic dipoles for increasing field amplitudes, with $\omega_L=0.086$ a.u., (b) $F_0=0.005$ a.u., (c) $F_0=0.05$ a.u., and (d) $F_0=0.14$ a.u.

sity and can even bring the atomic lines in quasiresonance with a laser harmonic, thus enhancing the intensities of high-order harmonic components. Such a situation is illustrated in Fig. 3 for $\omega_L = 0.086$ a.u. and $F_0 = 0.12$ a.u. in which harmonics and atomic lines can no longer be distinguished.

These results seem to indicate that the origin of the plateau in the harmonic distribution is linked to the presence of the (shifted) atomic frequency, the value of which coincides approximately with a local maximum in the harmonic spectrum, the relative height of the plateau being higher in the case of quasiresonance, as mentioned above. Similarly, the width of the plateau and, more precisely, the position of its high-frequency edge is likely to be related to the disappearance of the high-frequency satellites of the atomic line. To illustrate this point we have presented in Fig. 4 the relative harmonic intensities in terms of their order [Fig. 4(a)], and the ones of the satellites of the atomic components [Fig. 4(b)] in a case in which they can be distinguished.

This interpretation of the existence and width of the plateau is supported by our previous calculations performed at a frequency that is one-half of the one discussed here [8]. Indeed, if one considers a frequency $\omega_L = 0.043$ a.u. ($=1.17$ eV for the Nd laser), one observes the presence of a shifted atomic frequency accompanied by satellites, the width of the harmonic spectrum being again approximately determined by the number of satellites; see Fig. 2 of Ref. 8. This indicates that the plateau is approximately twice as wide, when measured in terms of laser-frequency units, if the laser frequency is divided by a factor of 2. This can be checked by comparing Fig. 4 of Ref. 8, in which the plateau extends up to around the 27th harmonic at $\omega_L = 0.043$ a.u., and our present data for $\omega_L = 0.086$ a.u., which show that the plateau extends up to the 15th harmonic; see below, Fig. 7(b). Further evidence is also shown below in Fig. 8, in which we present the harmonic spectrum obtained at a slightly higher frequency $\omega_L = 0.129$ a.u. (third harmonic of the Nd laser): the plateau which extends then up to the ninth harmonic, spans the same frequency range as in the previous cases.

Our results suggest also that, as a consequence of the existence of harmonics of the atomic Kepler frequency itself, the same analysis should hold also for the second, the third, etc. of these Kepler harmonics. This implies that our model predicts the existence of successive plateaus, less and less intense, the position, and intensity of each of them being governed by the ones of the harmonics of the fundamental atomic frequency.

We have checked that the spectra depend on the actual eccentricity and orientation of the Kepler orbit chosen to start the integration scheme: the intensities of the harmonic lines can differ depending on the initial conditions. In order to illustrate this point we show in Figs. 5(a)–5(b) the harmonic spectra obtained at a fixed field intensity, namely $F_0 = 0.11$ a.u., and for two extreme cases corresponding to circular orbits pertaining to planes either perpendicular [Fig. 5(a)] or parallel [Fig. 5(b)] to the laser polarization $\epsilon$. At the chosen intensity, if the orbit plane is perpendicular to $\epsilon$, the motion remains bounded and almost centered so that no even harmonics are present in the spectrum [Fig. 5(a)]. On the contrary, if the orbit plane is parallel to the polarization, the symmetry is broken and even harmonics do appear. The spectrum is also

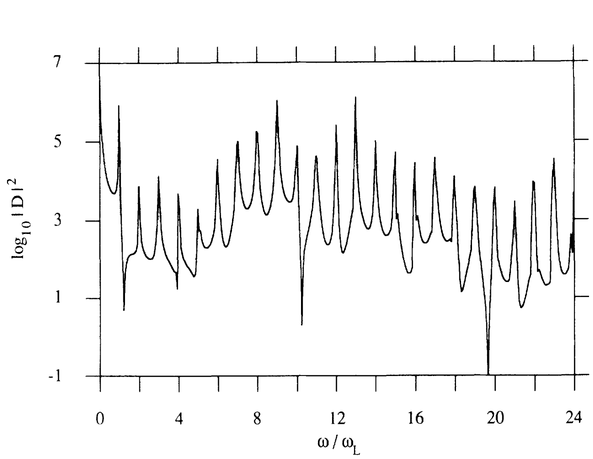


FIG. 3. Power spectrum $D(\omega)$ of the dipole $\mu_z$ with $\omega_L = 0.086$ a.u. and $F_0 = 0.12$ a.u. Note that the harmonics and the atomic lines can no longer be distinguished.

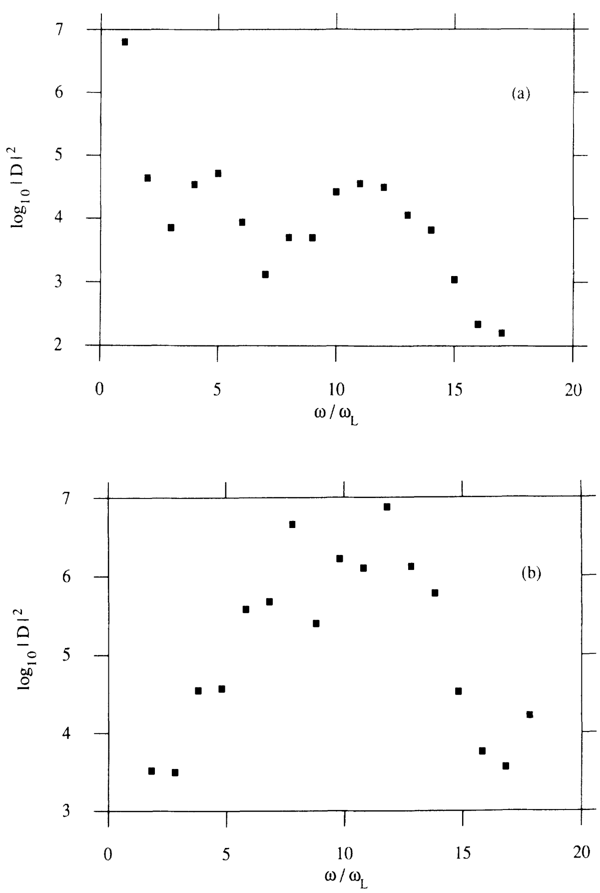


FIG. 4. (a) Relative intensities of the harmonic lines as a function of their order, for $\omega_L = 0.086$ a.u. and $F_0 = 0.14$ a.u. (b) same as (a) for the satellites of the "atomic" components.

more intense and noisier, the atomic lines being much broader, with subcomponents, and almost in resonance with multiples of the laser frequency. This certainly is responsible for the observed overall enhancement of the intensities of the spectral lines. Between these two extreme cases, and depending on the orientation and eccentricity of the initial orbit, one obtains different spectra ranging between those shown in Figs. 5(a) and 5(b). In fact, when randomly sampling the initial conditions as described in the following section, such extreme cases occur very rarely and the spectra for different trajectories, although they differ in the details, display in most cases similar features as illustrated in Figs. 2–4. Let us note also that some orientations, for instance, the semimajor axis being parallel to the field polarization, lead very easily to ionization and almost do not give rise to harmonic generation. This point is addressed next.

Indeed, another feature of this single-trajectory analysis is the possibility of investigating to what extent ionization can affect harmonic generation. For instance, at field amplitudes $F_0 > 0.12$ a.u. some trajectories lead to ionization, i.e., the electron goes very far away from the nucleus, becoming asymptotically free. We show, in Fig. 6(a), the time dependence of the atomic dipole $\mu_z$ for one of these trajectories, in which ionization takes place at approximately $t \approx 1600$ a.u.. $=21.90T_L$. Beyond this time, the time dependence of the dipole becomes approximately

$$\mu_z(t) \approx v_z t + \alpha_0 \sin(\omega_L t + \phi) , \quad (5)$$

where $\alpha_0 = F_0/\omega_L{}^2$, with the dimension of a length, is the

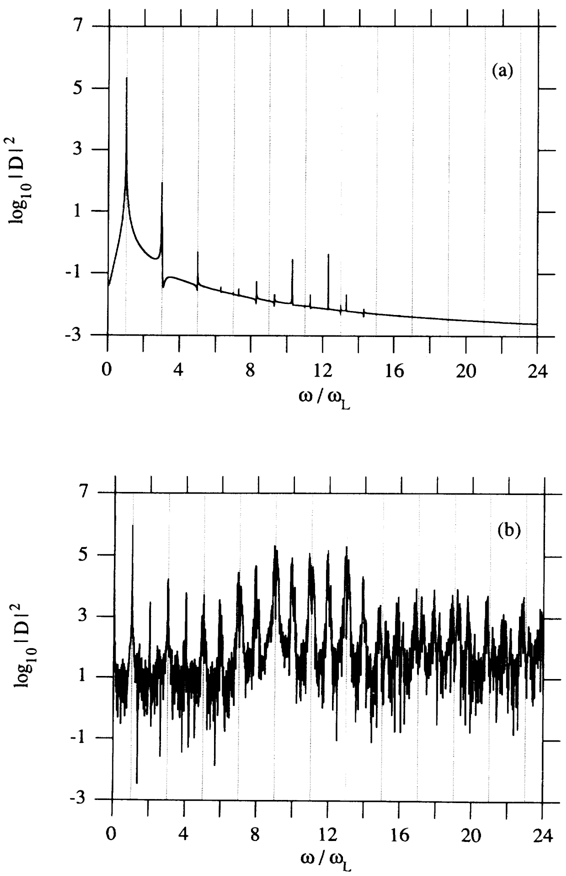


FIG. 5. Power spectra $D(\omega)$ of the dipole $\mu_z$ for two particular trajectories with $\omega_L = 0.086$ a.u. and $F_0 = 0.11$ a.u. For (a) the plane of the initially circular trajectory is perpendicular to the laser polarization, while for (b) it is parallel to the laser polarization.

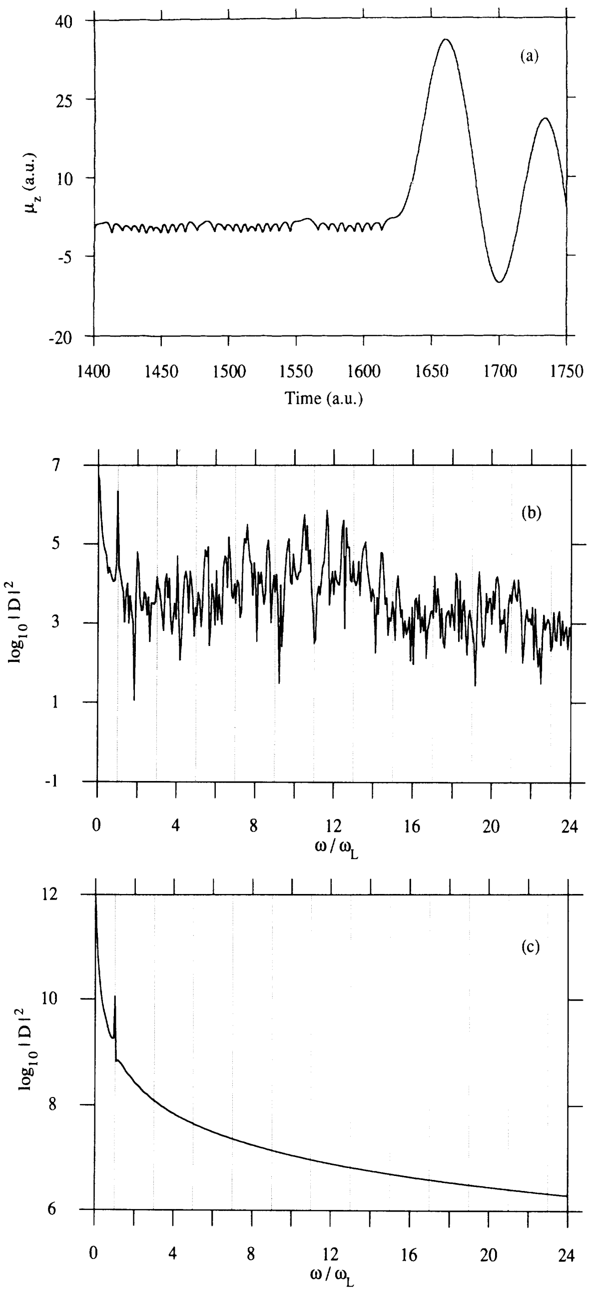


FIG. 6. (a) Temporal variations of the atomic dipole moment $\mu_z$ of an ionizing trajectory, with $\omega_L = 0.086$ a.u., $F_0 = 0.15$ a.u. Ionization takes place at approximately $t \approx 1600$ a.u. (b) shows the power spectrum $D(\omega)$ of $\mu_z$ corresponding to the bound motion of the trajectory, i.e., $t < 1600$ a.u., while (c) shows the power spectrum corresponding to the unbound motion, i.e., $t > 1650$ a.u.

classical excursion length of an electron driven by a single-mode field.

We show also in Fig. 6(b) the corresponding power spectrum, as obtained from the (windowed) Fourier analysis of the nonionizing part of the trajectory: It is very similar to the ones shown above and features a large number of harmonics and the atomic line accompanied by its satellites. If one then includes part of the ionizing section of the trajectory in the Fourier analysis, the structured spectrum becomes lost in a dominant background generated by the ionizing section, the only line surviving being the one at the laser frequency, corresponding to the light scattered by the asymptotically free electron (Thomson scattering) [Fig. 6(c)]. This simple analysis shows that when ionization takes place, harmonics are no longer generated. We mention that this result has been confirmed by a wavelet analysis of such trajectories, which allows us to follow the time dependence of the harmonic emission [14]. It is important to note, however, that this conclusion holds only for single-electron atoms, since otherwise the remaining bounded electrons would be still available for harmonic generation, though presumably in another (higher) intensity range.

The presence of an important background in the Fourier spectrum of the ionizing part of a trajectory [Fig. 6(c)] is a spurious effect which originates in the drift of the ejected electron in the field, which makes the atomic dipole grow in time, as shown in Eq. (5). The prescription to remedy to such an unphysical behavior is to consider the acceleration, which remains bounded in time, instead of the dipole (see the discussion in Ref. [23]).

Before closing this section, we mention that we have not found any important modifications of the power spectra associated with single trajectories differing only in the turn-on of the field, provided we perform the Fourier analysis far enough from the end of the rise time. We have investigated various rise times of the linear ramp, comprised between $\frac{1}{4}T_L \approx 18$ a.u. and $12T_L \approx 900$ a.u., without observing significant changes in the spectra. We have checked also that, although it can be of some importance for ionization, the detailed shape of the ramp ($\sin^2$, sech, etc.) has no noticeable influence on the spectra. We turn now to the study of an ensemble of trajectories which allows a better comparison with the case of real, spherically symmetric, atoms.

## IV. ENSEMBLE AVERAGES

In order to remove the unphysical even harmonics present in the single-trajectory spectra, it is necessary to reconstruct the spherical symmetry, or more precisely, the presence of an inversion center, existing in real atoms. To this end, one can consider an ensemble of classical atoms, initially in a microcanonical distribution:

$$\rho(E)=\delta(E-E_0), \quad E_0=-\tfrac{1}{2}\text{ a.u.}\ , \tag{6}$$

which amounts to choosing, at random, Kepler orbits with different eccentricities or angular momenta and orientations in space, compatible with the given energy. This is conveniently achieved by using the Monte Carlo classical trajectory method proposed by Abrines and Percival [20], who have shown that to construct such a microcanonical distribution, one has to specify the values of five parameters characterizing the elliptic orbits, within the following ranges:

$$0\le\beta\le 1, \quad \beta=2|E_0|L^2\ , \tag{7a}$$

$$0\le\varphi\le 2\pi\ , \tag{7b}$$

$$-1\le \cos\theta \le 1\ , \tag{7c}$$

$$0\le\psi\le 2\pi\ , \tag{7d}$$

$$0\le\alpha\le 2\pi\ . \tag{7e}$$

Here $L$ is the angular momentum, the triad of Euler angles $(\varphi,\theta,\psi)$ gives the orientations of the semimajor axis and of the plane of the ellipse and the mean anomaly $\alpha$ allows one to determine the position of the electron on the ellipse by (numerically) solving the Kepler equation which relates $\alpha$ to the eccentricity $e$ $[=(1+2E_0L^2)^{1/2}=(1-\beta)^{1/2}]$ and eccentric angle $\xi$:

$$\alpha=\xi-e\sin\xi\ . \tag{8}$$

From a set of these five parameters one defines the initial conditions for one trajectory, i.e., the position and momentum of the electron [see Eqs. (8)–(10) in Ref. [21],

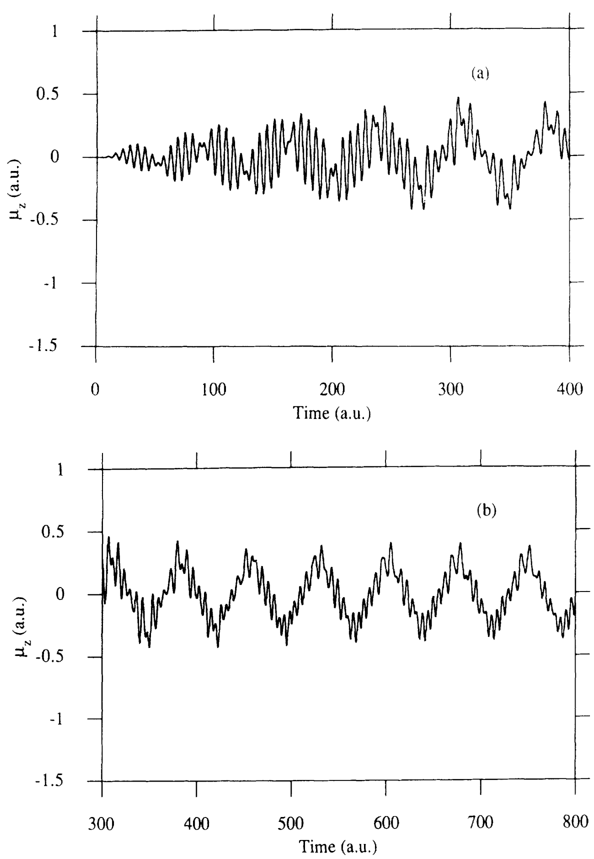


FIG. 7. Temporal variations of the averaged atomic dipole moment $\mu_z$ of an ensemble of trajectories along the laser polarization direction: $\omega_L=0.086$ a.u., $F_0=0.12$ a.u. 1000 Monte Carlo trajectories were used. (a) corresponds to the turn-on of the laser field, while for (b) the field amplitude remains constant.

but note the change of notation] at $t=0$, which allows one to start the integration of the equations of motion, Eqs. (2). One then determines the $z$ component of the atomic dipole and, by averaging over the number of trajectories chosen, one can follow the time evolution of the ensemble of atomic dipoles.

In Fig. 7(a) we show the early time evolution of the averaged atomic dipole, evolving from a microcanonical ensemble of 1000 initial Kepler trajectories, randomly chosen as described above, in the presence of a laser field with the same characteristics as in the preceding section, namely with a linear ramp up to 300 a.u. $\approx 4T_L$, followed by a constant amplitude $F_0=0.12$ a.u. Figure 7(b) displays the time dependence of the averaged dipole at a later time, i.e., when the field amplitude is constant. When compared with the evolution of the atomic dipole from a single trajectory, for the same field and time ranges shown in Fig. 1, one observes several notable differences. One is that, as a result of ensemble averaging, the atomic dipole is zero at $t=0$. Another feature of interest is that the oscillations of the averaged dipole are smoother and have smaller amplitudes than in the single-trajectory case. Indeed, the larger the number of trajectories, the smoother the averaged dipole, which globally follows the laser oscillations.

Shown in Figs. 8(a)–8(d) are the Fourier (FFT) spectra derived from such time-dependent dipoles, averaged over more than 2400 trajectories, for different values of the laser field strength comprised between $0.11 \leq F_0 \leq 0.14$ a.u. We have chosen to investigate this range of intensities to illustrate the strong variations of the harmonic spectra when one goes from the (relatively) low-intensity to the high-intensity regime. As a result of the averaging process, even harmonics no longer appear in the spectra: This was expected from the microcanonical sampling of our trajectories.

Another quite striking observation is the presence, especially in the lower-intensity range, of an atomic Kepler component with its satellites. The presence of these (ac Stark shifted) atomic frequencies was *a priori* unexpected since, without the laser, the averaging process from a microcanonical distribution results in an atomic dipole which stays zero in time, implying the disappearance of the atomic lines from the Fourier spectrum of an ensemble of field-free atoms. Note that this is verified here within an excellent approximation, as we have considered more than 2000 trajectories to perform our ensemble averages. On the contrary, in the presence of the laser, one observes a revival of the atomic Kepler frequencies which show up in the spectra obtained from en-

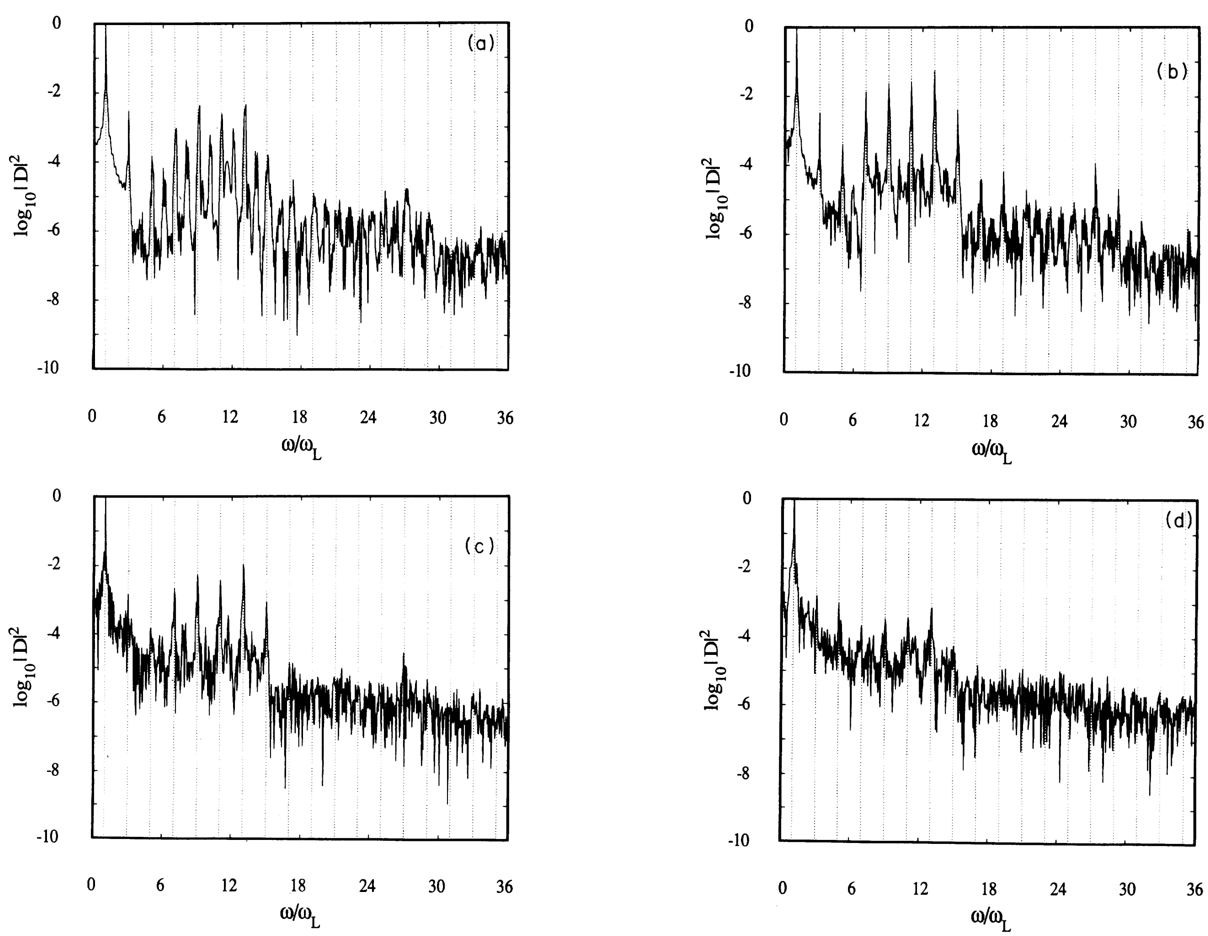


FIG. 8. Harmonic spectrum of a ground-state hydrogen atom in a laser field: $\omega_L=0.086$ a.u., 2400 Monte Carlo trajectories were used. (a) $F_0=0.11$ a.u. (b) $F_0=0.12$ a.u. (c) $F_0=0.13$ a.u. (d) $F_0=0.14$ a.u.

semble averages. As for the single-trajectory spectra, the atomic lines are shifted and exhibit equally spaced satellites separated from each other by the laser frequency. More precisely, at relatively low intensities [Fig. 8(a)] the atomic lines compete with the harmonics and it is only at higher intensities that these latter dominate. In fact, one observes [Fig. 8(b)] that the harmonic spectrum has the best signal-to-noise ratio at $F_0=0.12$ a.u., i.e., when the (ac Stark) shift of the atomic lines is such that they can be brought in close coincidence with a laser harmonic. At higher field strengths, the harmonics are still dominant, although their relative intensities decrease significantly with respect to the background; see Figs. 8(c)–8(d). One notes also that the atomic lines almost disappear into the rising background as the intensity grows.

These results raise several questions, among which one is why is there a "revival" of the atomic lines in the spectra obtained from ensemble-averaged dipoles. It is difficult to demonstrate that these frequencies should be present in the average motion of a set of laser-driven trajectories evolving from an initial microcanonical distribution, for the three-dimensional (3D) Kepler problem. We have verified, however, this point in the simpler case of a one-dimensional anharmonic oscillator and in the perturbative regime, i.e., at lower field intensity. One observes indeed for a given trajectory that the shift of the Kepler frequency comprises two distinct parts: one being dependent on the initial conditions (coordinates and phase of the field) while the other is independent. We note that the latter, which survives when performing the ensemble average, would in fact correspond to the ac Stark shift of the state. According to this picture the harmonic spectrum would be strongly enhanced when the shift of the Kepler frequency would be so that the atomic frequency would come into resonance with (odd) multiples of the field frequency. This seems to happen here at $F_0=0.12$; see Fig. 8(b). As a final remark concerning the presence of the atomic lines in the spectra, let us mention that by including a phenomenological damping term into the equations of motion one could eliminate these atomic lines. These features of the classical approach for harmonic spectra will be discussed in more detail in a future paper dealing with one-dimensional (1D) systems.

Another question related to these spectra is why is there an increase of the noise with the laser intensity. Part of this noise can be ascribed to the fact that at higher intensities, the proportion of ionizing trajectories increases significantly. The consequence of ionization is that the dipole associated with the corresponding trajectory becomes very large (in fact much too large), as compared to those which stay bounded; see Fig. 6(a). We have observed that, although ionizing trajectories can be oriented with almost equal probabilities towards positive or negative values of $z$, the presence of a few of them can spoil the statistics of our averaged dipole, even when we consider several thousands of trajectories. To remedy such an unpleasant feature we have accordingly excluded from the average the trajectories in which $r$ eventually becomes too large [24].

Another reason for the observed increase of the noise is the fact that, as the intensity increases, the amplitudes of the oscillations of the individual atomic dipoles grow significantly and become somewhat chaotic, even in the absence of ionization. This apparently chaotic behavior makes the averaged dipole more noisy, which, in turn, affects its Fourier spectrum; see Fig. 5(b). It should be noted that these two phenomena are likely to be closely connected as the onset of ionization seems to be related to the onset of chaos [16]. As already mentioned, a further indication on the possible role of chaos in the dynamics of the system is the observed sensitivity of the ionization time on the initial conditions, for an individual trajectory.

One question our classical model cannot properly address is the possible role of resonances in harmonic generation. Indeed, it is known that in the low-frequency range investigated here, the model somewhat underestimates the ionization probabilities [15]. More precisely, it has been shown that at a scaled frequency $\omega_L \approx 0.086$, the presence of resonances enhances significantly the ionization cross section, as compared to the classical calculation, which is essentially nonresonant in this range. The exact positions and widths of these resonances strongly depend on the details of the (dressed-) atomic spectrum, the determination of which is still a very difficult problem, necessitating large-scale computations [6]. It appears, however, that as the main effect of resonances is to increase the cross sections for the concurrent process of ionization, their presence would lead to an overall de-

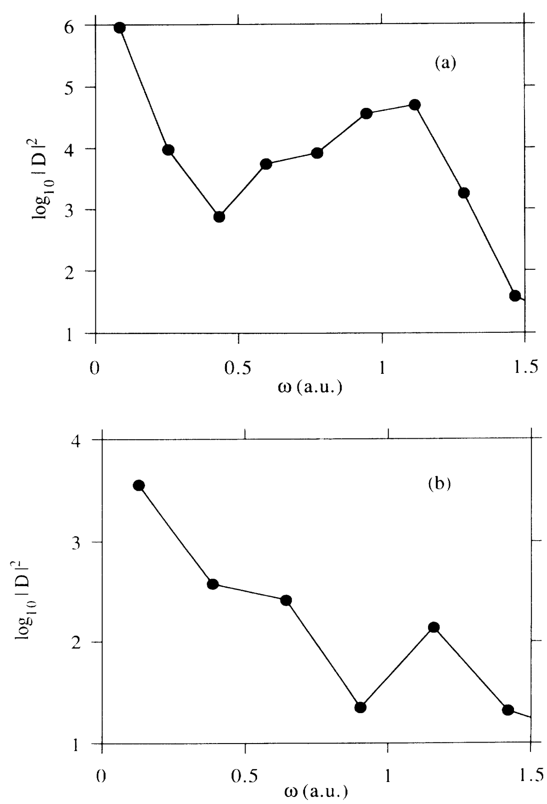


FIG. 9. Relative intensities of the harmonic lines as a function of their order, (a) $\omega_L=0.086$ a.u., $F_0=0.12$ a.u.; (b) $\omega_L=0.129$ a.u., $F_0=0.11$ a.u.

crease of the harmonic intensities. On the other hand, it is not clear to which extent they would modify the relative intensities of different harmonic orders. In order to partially answer this point, we have computed the harmonic spectrum obtained at a scaled frequency $\omega_L = 0.129$ (third harmonic of the Nd laser), for which the agreement between the classical model and the experiment for ionization is excellent [15]. The corresponding spectrum, shown in Fig. 9, displays the same features as the ones obtained at lower frequency, the plateau spanning, as expected, the same frequency range centered on the Kepler frequency.

Coming back to the ensemble-averaged spectra, one observes that at a given low frequency, generation of high-order harmonics takes place with a significant intensity in a quite narrow domain of field intensity, namely between $F_0 \approx 0.11$ and 0.13 a.u. at $\omega_L = 0.086$ a.u. In the lower intensity range, one stays in a perturbative regime, and the magnitudes of the successive harmonics decrease steadily with their order. At higher intensities, ionization becomes dominant, and higher-order harmonics are drowned in a growing noisy background. In fact, it seems that the onset of classical chaos, at such a low frequency and in the adiabatic regime, takes place precisely at $F_0 \approx 0.13$ a.u. [25]. We will come back to this point in the following short discussion.

## V. CONCLUSIONS

In this study we have verified that a classical model reproduces qualitatively the harmonic spectrum generated by atoms submitted to an intense laser field, in the so-called low-frequency regime. The validity of our model is obviously limited to the case of hydrogen in which our results should nevertheless correctly describe the harmonic spectrum generated in the presence of intense Nd laser light (first, second, and third harmonics).

We have shown also that, beyond this somewhat expected success, the classical approach provides an interesting view on the origin and width of the plateau which has been observed in the harmonic spectra. One observes in fact that the plateau is approximately centered on the (shifted) Kepler frequency, characteristic of the atomic system. This indicates that the maximum order of the harmonics with a significant intensity, i.e., the high-frequency edge of the plateau, depends on the value of the Kepler frequency, as measured in units of the field's. For example, at $\omega_L = 0.043$ a.u. (Nd laser), one has $[\omega_0/\omega_L] = 23$, where $[x]$ is the integer part of $x$, and the plateau extends up to $N = 27$ (see Fig. 4 of Ref [8]); on the contrary, at $\omega_L = 0.086$ a.u. (second harmonic of the Nd laser), one has $[\omega_0/\omega_L] = 11$ and, although it spans the same frequency range around $\omega_0$, the plateau extends only up to the harmonic $N = 15$, i.e., about half of the preceding case; see Fig. 8(b). The implication of these results is that the higher the laser frequency, the more narrow the plateau and, accordingly, the smaller the number of harmonics. See also Fig. 9, where this point is clearly illustrated.

Although this analysis seems to be qualitatively correct (the plateau has been observed to be much less developed at high frequencies [1–3]), it is not clear to what extent they can be transposed to the case of real (quantal) atoms. Indeed, the question arises of the relevance of the classical approach, which is known to underestimate the ionization probability in the low-frequency range we have discussed here [15,16]. This can be ascribed to the fact that in the classical model, there is only one characteristic atomic frequency, namely Kepler's, which directly corresponds to the ground-state ionization energy as $\omega_0 = 1/n_0^3$, where $n_0$ is the principal quantum number of the corresponding Bohr orbit. Classical resonances are expected to occur at frequencies $\omega = \omega_0/2$, $\omega_0 = \omega_0/3, \ldots$ in order of decreasing importance. In fact, the analysis presented here indicates that the harmonic lines are notably more intense when the field intensity $F_0$ is such that the (shifted) Kepler frequency $\omega_0 = n\omega_L$ (here $n = 11$ at $\omega_L = 0.086$ a.u. and $F_0 = 0.12$ a.u.). It is clear, however, that no quantitative conclusion can be drawn from this model since it is very unlikely that this classical resonance will coincide with a real (quantum) one. Nevertheless, we believe the overall picture remains correct: The plateau is centered on the main atomic frequencies which can come into resonance, depending on the field intensity and frequency.

Another observation following this classical analysis is that harmonic generation and (multiphoton) ionization, which both occur in the same intensity range, are in fact competing processes. More precisely, high-order harmonic generation becomes important at intensities beyond the perturbative regime: This is a necessary condition to be able to observe high-order harmonics with significant intensities. Multiphoton ionization becomes important also in this intensity range and competes with harmonic generation in the sense that the ejected photoelectron no longer generates harmonics. Moreover, in the low-frequency regime we have considered here, the onset of ionization which coincides with the onset of the classical chaos renders the Fourier spectra much noisier, so that higher-order harmonics are lost in a growing background. Accordingly, a prediction of our model is that significant numbers of high-order harmonic photons can only be obtained in a quite narrow range of intensities, i.e., beyond the perturbative regime but before the chaotic regime which prevails at higher intensities. Note, however, that in multielectron atoms, in the case of a first ionization, the remaining ionic core will remain available (if produced in enough number) to continue to generate harmonics.

The presence of the (shifted) Kepler lines, accompanied by their satellites, in the spectrum of the light radiated by an ensemble of driven atomic dipoles, raises several questions. It should be noted first that, had we included a phenomenological damping term in the equations of motion, the Kepler lines would have disappeared from the Fourier spectra, provided these latter had sampled the trajectories for a long enough time. Their role remains nevertheless very important as they seem to determine the position and extension of the plateau, probably through the possible existence of (multiphoton) resonances which could enhance the harmonic intensity. This point cannot be clearly elucidated with the help of a quantitative analysis in the case of the classical 3D hydrogen atom, since it is extremely difficult to carry out an analytical calculation to determine the ac-shifted atomic frequencies, in the presence of the external field. This was one of our main motivations to consider simpler 1D anharmonic systems which lend themselves more easily to analysis and which will be considered in more detail in the future.

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