Why hyper-Raman lines are absent in high-order harmonic generation

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(Received 28 September 2000; published 29 May 2001)

The absence of hyper-Raman lines in the observed spectrum emitted by an atom in the presence of a strong laser field is explained by evaluating the spectrum emitted by a unidimensional atom. It is found that the lines are emitted during a short time interval by the atom and that they therefore are small when compared with harmonical lines that are instead emitted during the laser pulse.

DOI: 10.1103/PhysRevA.64.013802 PACS number(s): 42.65.Ky, 32.30.Jc

The atoms of a low-pressure gas driven by an intense laser pulse of frequency ω_L diffuse electromagnetic radiation [1,2]. The experimentally observed spectrum is formed from a wide plateau of odd harmonics of ω_L that can be as large as $300\omega_L$, followed by a rapid cutoff of the emission. The scattered radiation seems to inherit from the pump field some of the properties of coherence [3–5] which makes the effect interesting for the practical possibility to obtain radiation in the ultraviolet (UV) or extreme ultraviolet (XUV) band; furthermore, computer simulations and theoretical calculations have shown that a superposition of harmonic fields with suitable phase permits the construction of pulses with a duration of very few optical cycles [6,7]. This effect, called high-order harmonic generation, is a fascinating problem in modern atomic physics. From the theoretical point of view, it is interesting because it represents a good field to devise new nonperturbative approximations to the laser atom interaction theory that can be easily tested with the experiments. In fact, the intensity of the field used in order to have a relevant observable emission must be larger than 10^{13} W/cm^2, far beyond the intensity of 10^{10} W/cm^2 pointed as the extreme limit in the use of traditional perturbation theory [8].

An unsettled question in the description of harmonic generation consists of the fact that many theories predict, together with the harmonic lines, the presence of other frequencies called hyper-Raman lines, which are never observed during actual experiments of harmonic generation [9]. Their position is given by $\omega_{hR}=\tilde{\omega}_i-\tilde{\omega}_j\pm 2k\omega_L$ with the k integer; in the presence of a cw laser field the $\hbar\tilde{\omega}_n$ are the quasienergies of the dressed atom. Several explanations have been proposed for the failure to detect these lines. For example, it has been argued that they are emitted preferentially in a direction orthogonal to the incident laser field and easily escape detection; or that their intensity decreases by increasing the laser field, or that slow variations of the laser intensity keeps these lines low, or that slow atomic ionization abate their presence. For sure, as theory and simulations state, in the presence of a pulsed pump, their position is not fixed since it depends upon the value of the laser field [9,10].

The strong acceleration of the electrons driven by the electric field is the physical mechanism causing the emission. By considering a one-electron atom, a common way to obtain the spectrum is to use the Larmor formula:

$$\frac{\mathrm{d}S(\omega)}{\mathrm{d}\omega}=\frac{4e^2}{3c^3}|\mathbf{a}_F(\omega)|^2, \tag{1}$$

with $\mathrm{d}S(\omega)$ the energy emitted by the atom during the whole laser shot in the frequency range $\mathrm{d}\omega$ and $\mathbf{a}_F(\omega)$ the Fourier transform of the quantum averaged electron acceleration $\mathbf{a}(t)$ that can be calculated by means of the Ehrenfest theorem. For later use, we write the Fourier transform as

$$\mathbf{a}_F(\omega)=\int_{-\infty}^{\infty}\mathbf{a}(t)f_\omega(t)\mathrm{d}t; \quad f_\omega(t)=\frac{\exp(-i\omega t)}{\sqrt{2\pi}} \tag{2}$$

and $f_\omega(t)$ can be called the analyzing kernel of the Fourier transform.

The Fourier expansion is a rather sensitive technique and might require accurate and realistic models: however, few considerations can help in the choice of the approach to be used in the calculation. The observed and calculated charac-

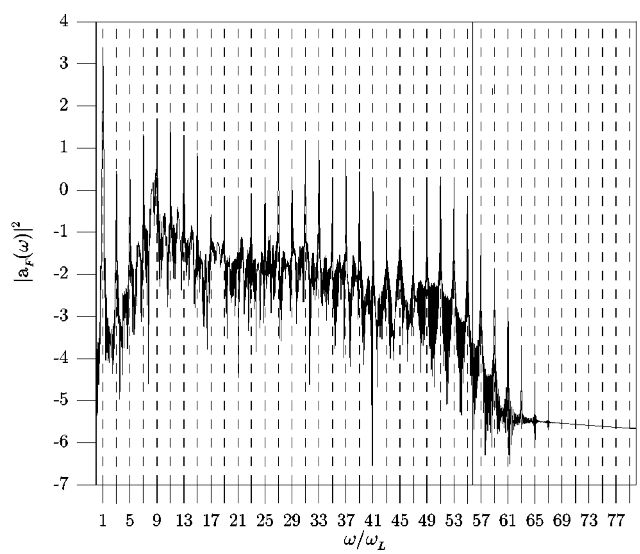

FIG. 1. Fourier power spectrum emitted by a one-dimensional atom with the short range potential $U(x)$. The relevant parameters entering the calculations are: $I_L=1.4\times10^{14}$ W/cm^2, $\omega_L=\omega_{10}/9$ corresponding to $\lambda_L=1096$ nm. The vertical line indicates the cutoff frequency as predicted by the recollision model.

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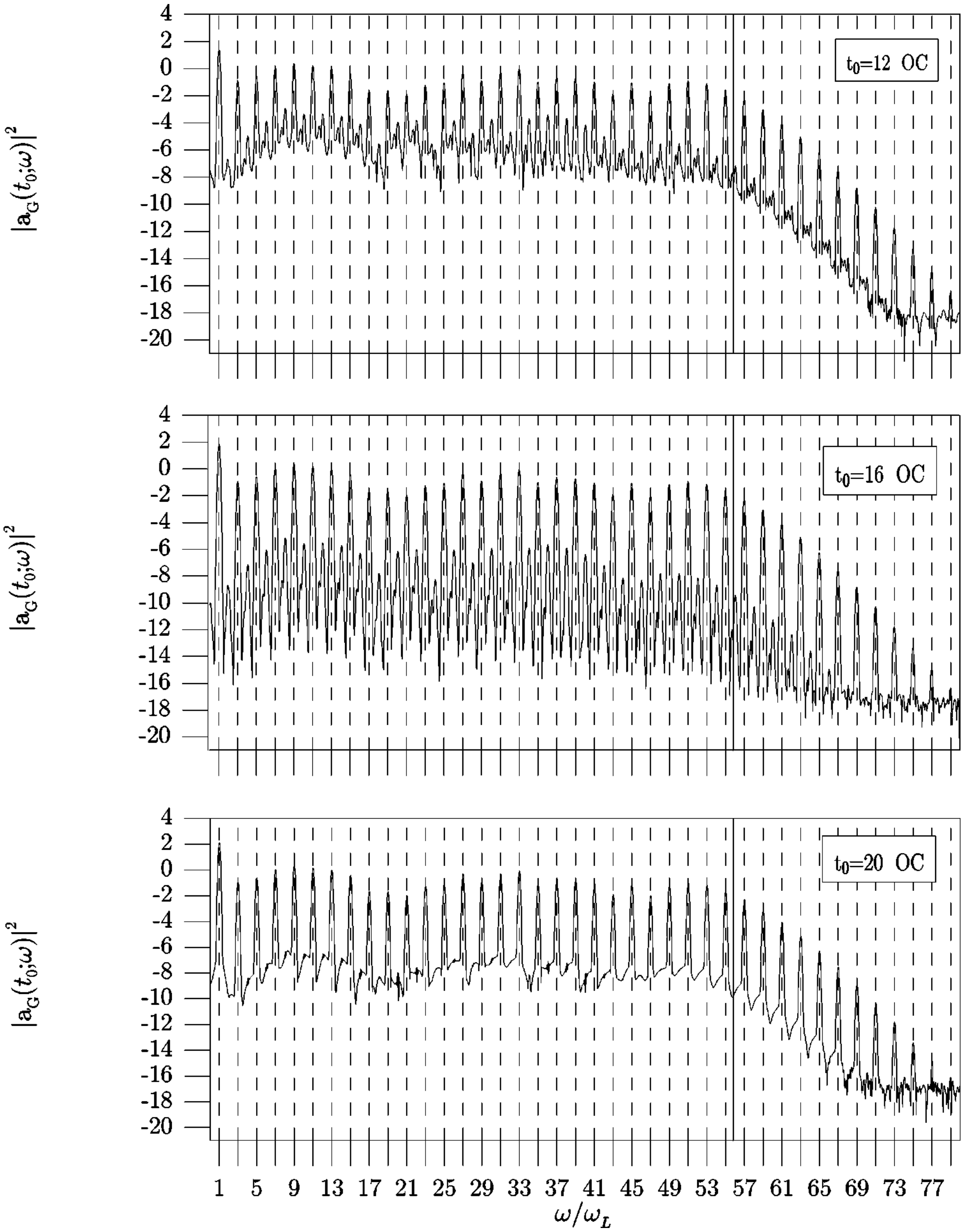

FIG. 2. Gabor power spectrum emitted by a one-dimensional atom with the short-range potential $U(x)$ at different instants of time t_0 shown in the inset. The relevant parameters entering the calculations are: $I_L=1.4\times10^{14}$ W/cm^2, $\omega_L=\omega_{10}/9$ corresponding to $\lambda_L=1096$ nm. The vertical line indicates the cutoff frequency as predicted by the recollision model. The Fourier spectrum is given in Fig. 1.

teristics of the spectrum are quite independent from the specific gas or model used, and consequently, independent from the specific details of the bare Hamiltonian. The ensuing seminal idea is that in the presence of strong fields, the electron dynamics is determined more by the laser field than by the exact details of the electron atom interaction energy. Being assured by the previous considerations, two-level atoms or one-dimensional atoms have been extensively used as models, with success [11,12]. Since the one-dimensional approach reproduces with accuracy all the features of the experiments and still retains simplicity, we will use it in this paper.

If we call $H_0(x)=-(\hbar^2/2m)(\partial^2/\partial x^2)+U(x)$ the Hamiltonian of the one-dimensional atom in the absence of laser, the full Hamiltonian of the problem in the length gauge is: $H(x,t)=H_0(x)+ex\mathcal{E}_0 f(t)\sin\omega_L t$ with $\mathcal{E}_0$ the peak external electric field, $f(t)$ a function that describes the pulse envelope, and $U(x)$ the electron nucleus interaction energy.

In what follows, we will use the following expression for $U(x)$:

$$U(x)=-\frac{U_0}{\cosh^2\dfrac{x}{x_0}}, \tag{3}$$

describing a short-range potential; the time-independent Schrödinger equation $H_0(x)u_n(x)=E_n u_n(x)$ allows a finite number of discrete states and a continuum set of eigenstates and eigenvalues that can be written in closed form [13]. In the following, we will make the choice $U_0=\frac{3}{2}I_{\rm H}$ and $x_0=2a_0$ with $I_{\rm H}=me^4/(2\hbar^2)$ the ionization energy of the hydrogen. With such parameters the Hamiltonian $H_0(x)$ supports two bound states with definite and opposite parity and of energy $E_0=-I_{\rm H}$ and $E_1=-\frac{1}{4}I_{\rm H}$.

$U(x)$, being a short-range potential, allows us to focus the numerical effort in a limited region of the space where photon emission essentially occurs. $U(x)$ is by far academically important since it can be fruitfully used in problems of negative ions.

We have numerically integrated the time-dependent Schrödinger equation with a laser-pulse profile $f(t)$ that switches on and off for six laser optical cycles (OC) with a $\sin^2$ envelope and that is constant during 20 OC. The value of the flat laser intensity has been taken as 1.4×10^{14} W/cm^2. The photon energy is $\hbar\omega_L=\hbar\omega_{10}/9$ with $\hbar\omega_{10}=E_1-E_0$ and corresponds to a wavelength of $\lambda_L=1096$ nm. The cutoff predicted by the well-known recollision model [14] is $\hbar\omega_M\approx I_H+3U_p$, and will always be shown in the following reported spectra.

In Fig. 1 we show the Fourier power spectrum of the acceleration of the electron. The spectrum presents a full comb of odd lines and reproduces well the standard behavior of the experimental spectra, with a plateau and a clear cutoff at the position predicted by the recollision model. No impressive evidence of hyper-Raman lines is present in the plot.

A point of discussion when performing the Fourier analysis is that a specific transform coefficient $\mathbf{a}_F(\omega)$ is obtained from the full time-dependent wave function without referring to the difference from one temporal location to another in the data stream, so that it contains the information of a specific frequency but none about when it has been emitted. In other words, short-living transient phenomena and emission of high-frequency fields, often bound to last for a very short time interval, are liable to be drowned out by other long-lasting processes, leaving, if any, only a tiny feature in the Fourier spectrum that can easily be overlooked or hidden. In order to remediate to this particular flaw intrinsic to the Fourier transform, several different approaches have been proposed [15,10,16,17]. In what follows, we shall use a Gabor transform of the electron acceleration to obtain information on the temporal evolution of the spectrum emitted by the one-dimensional atom.

We recall that the Gabor transform is the most used short-time Fourier transform, occasionally (and wrongly) confused with a wavelet transform; it consists of substituting the Fourier analyzing kernel $f_\omega(t)$ with the new one

$$g_{t_0,\omega}(t)=\exp\left\{-\frac{1}{2}\left(\frac{t-t_0}{\sigma}\right)^2\right\}\mathrm{e}^{-i\omega t} \tag{4}$$

with constant σ. We define Gabor spectrum of a signal $A(t)$ as the function

$$A_G(t_0;\omega)=\int_{-\infty}^{\infty}A(t)g_{t_0,\omega}(t)\mathrm{d}t; \tag{5}$$

as apparent, the Gabor transform yields the Fourier transform of a selected clip of width 2σ around the central time t_0 from the full data stream. The correspondence $\lim_{\sigma\to\infty}g_{t_0,\omega}(t)=\sqrt{2\pi}f_\omega(t)$ shows that the Fourier transform can be seen as a particular Gabor transform with infinite temporal window (no time localization possibility).

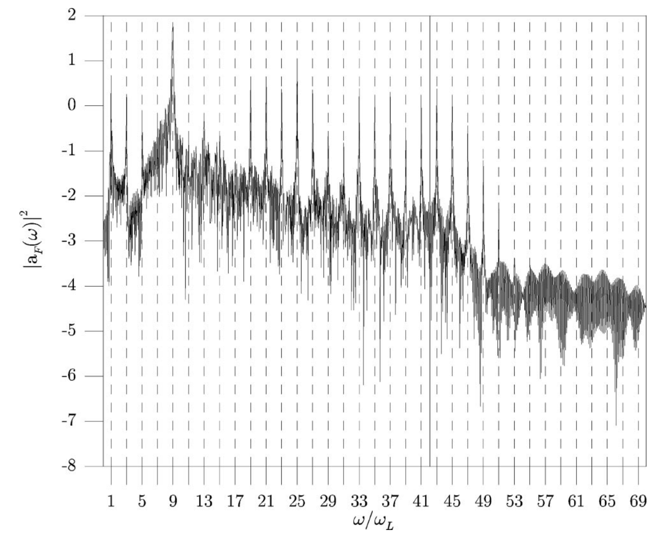

FIG. 3. Fourier power spectrum emitted by a one-dimensional atom with the soft core potential $W(x)$. The relevant parameters entering the calculations are: $I_L=1.0\times 10^{14}$ W/cm^2, $\omega_L=\omega_{10}/9$ corresponding to $\lambda_L=1040$ nm. The vertical line indicates the cutoff frequency as predicted by the recollision model.

By inserting the Gabor transform into the Larmor formula, we obtain a function of t_0 and ω that we define as spectrum emitted at time t_0. This interpretation is supported by the property that

$$\int_{-\infty}^{\infty}A_G(t_0;\omega)\mathrm{d}t_0=\sqrt{2\pi\sigma^2}A_F(\omega), \tag{6}$$

meaning that, apart from an irrelevant normalization factor, the sum over all the emission times gives the Fourier spectrum. As said, the Gabor spectrum gives information on the frequencies emitted in the time interval $t_0\pm\sigma$ with an uncertainty $\delta\omega=1/(2\sigma)$; from the Fourier point of view, the infinite width of the temporal window yields a perfect accuracy in the determination of the frequency.

In Fig. 2, we show the Gabor spectra taken at different instants of time t_0 with $2\sigma=4$; OC hyper-Raman lines appear only at the beginning of the pulse flat part, reach the maximum around the center of the pulse ($t_0=16$ OC) and disappear quickly. We argue that their short life makes their detection difficult during an actual experiment.

This behavior is independent of the atomic model used. In fact, we obtained similar results by using the well-known soft-core potential

$$W(x)=-\frac{Ze^2}{\sqrt{x_0^2+x^2}}, \tag{7}$$

with $Z=1$ and $x_0=1$ a.u. In this case the difference between the ground-state energy and the first excited-state energy is $\omega_{10}=0.39$ a.u. so $\omega_L=\omega_{10}/9$ corresponds to $\lambda_L=1040$ nm. We considered the same pulse envelope as in the previous case and the intensity of the laser is 1.0×10^{14} W/cm^2. Figure 3 shows that the Fourier spectrum

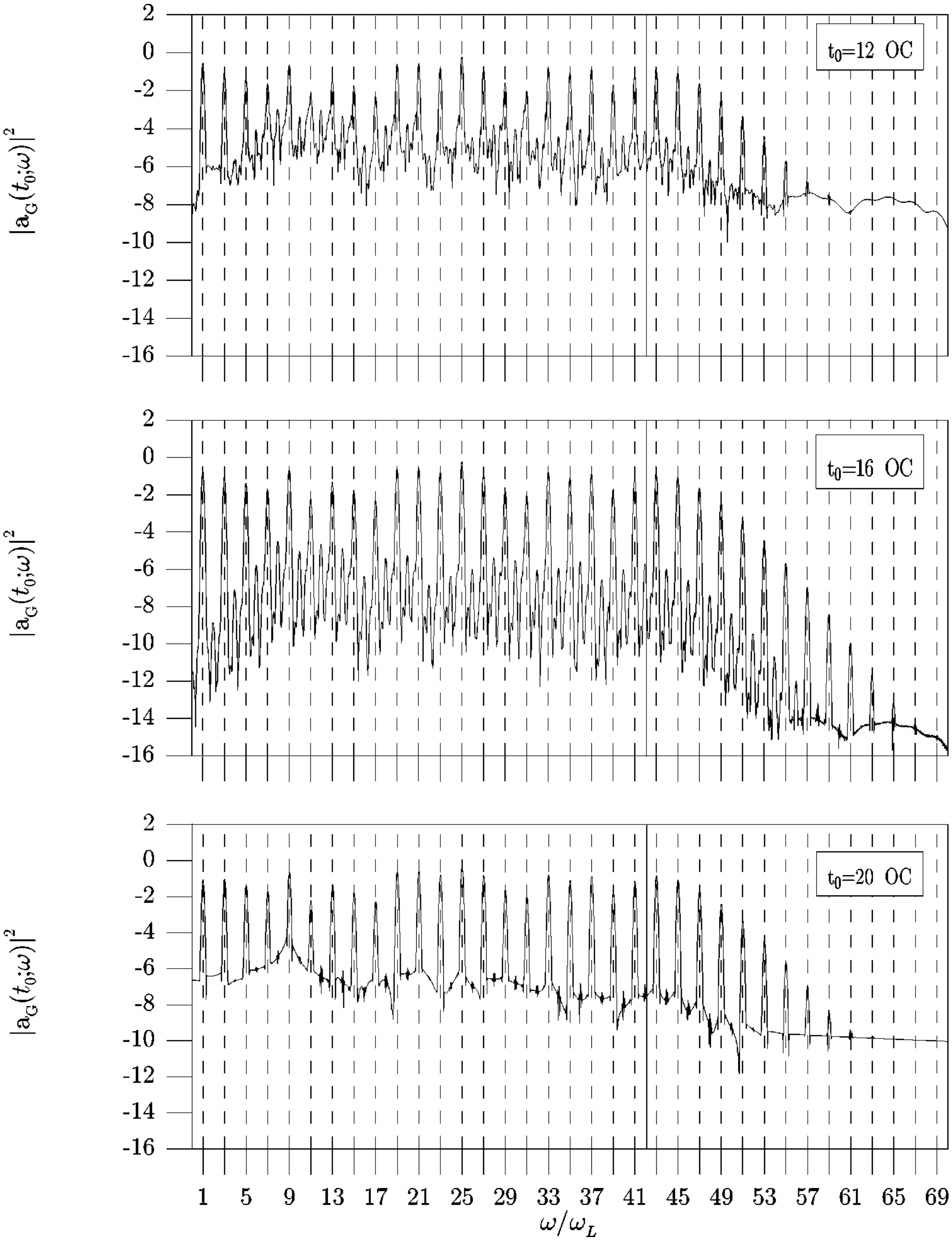

FIG. 4. Gabor power spectrum emitted by a one-dimensional atom with the soft core potential $W(x)$ at different instants of time t_0 shown in the inset. The relevant parameters entering the calculations are: $I_L = 1.0\times 10^{14}$ W/cm^2, $\omega_L = \omega_{10}/9$ corresponding to $\lambda_L = 1040$ nm. The vertical line indicates the cutoff frequency as predicted by the recollision model. The Fourier spectrum is given in Fig. 3.

does not contain hyper-Raman lines. But, in Fig. 4, we see that the atom emits hyper-Raman lines in the central part of the pulse.

Perhaps it is worth noting that the Gabor spectra have a background much lower than that of the corresponding Fourier spectrum. This allows us to see that the extension of the cutoff region is wider than that showed by the Fourier spectrum. From this point of view, the Gabor transform can be considered a very useful spectroscopic tool.

In conclusion, we studied the problem of hyper-Raman lines emission in high-order harmonic generation. We used a one-dimensional atom with a short-range potential and with a soft-core potential. Whereas the Fourier spectra of the acceleration do not show hyper-Raman lines, the Gabor spectra indicate that actually they are emitted by the atom. In particular, we showed that hyper-Raman lines are emitted only in the central part of the pulse and this could make their experimental detection very difficult.

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