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Harmonic generation by atoms in circularly polarized laser fields: far-off and near resonance regimes

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ABSTRACT The generation of harmonics by atoms interacting with two laser fields having coplanar circular polarizations and an integral frequency ratio is addressed through ab initio numerical simulations. A detailed characterization of a few specific harmonics is given. In particular, the two different cases where the total energy absorbed through photons is far off or close to the energy gap between different atomic states are investigated. It is found that the conversion efficiency in the harmonic generation is strongly dependent on the inner atomic structure and in certain specific cases it can be significantly enhanced within a small frequency range.

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1 Introduction

The interaction between an atom and two coplanar laser fields with circular polarizations and integral frequency ratio is certainly an intriguing topic. In fact, it is worth recalling that when a single circularly polarized laser field interacts with an atom no harmonic generation is possible.

In the case where the two laser fields have opposite polarizations the emitted harmonics are of order

$$n = k(\eta + 1) \pm 1, \tag{1}$$

where η is the frequency ratio and $k \in \mathcal{N}_+$. Hence, contrary to linear polarization, for even η even harmonics are also allowed. Selection rules of this type are characteristic of all systems whose Hamiltonian is invariant under a dynamical symmetry operation [1], i.e. a transformation in both space and time. A further example of such systems is a circular molecule, e.g. benzene, interacting with a single circularly polarized laser field [1–4]. The derivation of such selection rules can be achieved by means of different arguments: angular momentum conservation, group theory, and change of reference frame [4]. The configuration we investigate here is of particular interest because of the filtering effect due to the selection rules (1), i.e. the higher the value of η, the fewer the number of harmonics in a certain frequency range and the smaller the circular polarization of the emitted harmonics. This last point becomes particularly appealing for harmonics in the soft X-ray regime. Experimental [5] as well as theoretical investigations [6–8] of such configurations have been carried out during recent years. Harmonic generation from sources other than atoms and circular molecules, like linear molecules [9] and nanotubes [10], has been also addressed. The present paper is organized as follows: after a short summary of the theoretical frame within which our system lies, a full description of the model and of the numerical simulations performed is presented. A conclusion is then given. Atomic units (a.u.) are used throughout the paper.

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2 Theoretical frame

In order to fully understand the nature of the harmonics we investigate, let us firstly summarize the derivation of the selection rules (1) using angular momentum conservation. Let z be the propagation direction of the two lasers and σ^+ and σ^- the polarizations of the lasers of frequencies ω and $\eta\,\omega$, respectively. If a harmonic σ^+ is emitted, the sum of all the components along z of the angular momenta carried by the absorbed photons has to be $+1$. If the atom for example absorbs p photons from the low-frequency laser, it must absorb $p-1$ photons from the other laser. Hence, the total absorbed energy is $p\omega + (p-1)\eta\omega = \omega[p(1+\eta) - \eta] = \omega[(p-1)(1+\eta)+1]$. Since p is arbitrary, with $k = p-1$ and $N = \eta + 1$ we see that the harmonic of order $kN+1$ is emitted. With the same argument, starting from a harmonic of polarization σ^-, we obtain $kN-1$. The key point that should be stressed is the following: harmonic generation is possible if, and only if, the absorption of p photons from one laser is accompanied by the absorption of $p \pm 1$ photons from the other laser. Moreover, it is worth noticing that the emitted harmonics have circular polarization.

Angular momentum considerations, as well as symmetry invariances, provide information on the allowed orders, but they cannot be used to derive any details concerning relative intensities and structures in the harmonic spectra like plateaus and cut-offs. Above all, stating that a certain order is allowed does not mean that the corresponding harmonic is really emitted. In fact, the previous arguments do not include case-specific features such as the atomic structure and the laser intensities.

Recently, the characteristics and the field dependence of the harmonics emitted in our configuration have been addressed analytically through a calculation [8] based on the Lewenstein model [11]. With this analytical tool it is possible to derive a generalized cut-off for the harmonic spectra, an integral expression for the harmonic dipole strength, and to obtain again, independently, the selection rules (1). Concerning the harmonic intensities, such a model treatment holds as long as the total absorbed energy by the electron is not 'too close' to a transition [8].

3 Numerical simulations

The numerical simulations were performed through an ab initio integration of the time-dependent Schrödinger equation on a two-dimensional (2D) grid. Choosing a 2D grid permits us to run simulations quickly on any modern PC and it is not expected to introduce qualitative modifications to the phenomena we aim to investigate. In particular, this holds for the specific configuration we are addressing here. In fact, both the electric field vectors of the lasers as well as the electron's trajectories lie in the plane that is represented by the 2D numerical grid. In the direction perpendicular to that plane the electron wave packet merely spreads. This spreading is not expected to strongly affect the main features of the harmonic generation spectra such as the cut-off, for instance. Using a polar grid rather than a Cartesian one ensures that no artificial symmetry violation due to the numerics is introduced.

In polar coordinates (ϱ, φ), dipole approximation, and length gauge the time-dependent Schrödinger equation of our system reads

$$\mathrm{i}\frac{\partial}{\partial t}\Psi(\varrho,\varphi,t) = \Big[-\frac{1}{2\varrho}\frac{\partial}{\partial\varrho} - \frac{1}{2\varrho^2}\frac{\partial^2}{\partial\varphi^2} - \frac{\partial^2}{\partial z^2} + V_{\mathrm{at}}(\varrho) + \sin^2(\Theta t)\,(E_1\varrho\cos(\varphi-\omega t) + E_2\varrho\cos(\varphi+\eta\omega t))\Big]\Psi(\varrho,\varphi,t), \quad (2)$$

where the two laser pulses have a duration $T = \pi/\Theta$, with $\Theta = \omega/84$, and a sine-square shape. E_1 and E_2 are the two electric fields and $V_{\mathrm{at}}(\varrho)$ is a 'soft-core' 2D potential given by

$$V_{\mathrm{at}}(\varrho) = -\frac{\alpha}{\sqrt{\varrho^2+\beta^2}}. \quad (3)$$

The parameters α and β can be tuned in order to adjust the ionization energy. In our simulations we used $\alpha = 2.44$ and $\beta = 0.20$. These values provide an ionization potential of $I_{\mathrm{p}} = 2.0$, i.e. that of real He^+. As we aim to address the role played by resonances in the conversion efficiency, it is important to know the level scheme of the model potential we use. With the chosen parameters the lowest four excited states have energies $\Omega_1 = 0.985$, $\Omega_2 = 1.375$, $\Omega_3 = 1.548$, and $\Omega_4 = 1.592$. These energies are measured with respect to the ground state. The laser frequencies have been chosen in accordance with this level scheme. Concerning the frequency ratio η, all the results presented hereafter have been obtained taking $\eta = 4$. In Fig. 1 an example of a two-color spectrum obtained with laser fields of equal intensity is shown. As expected, only orders allowed by the selection rules are present. Substructures and broadening of certain harmonics can be present when bound states other than the ground state come into play [4].

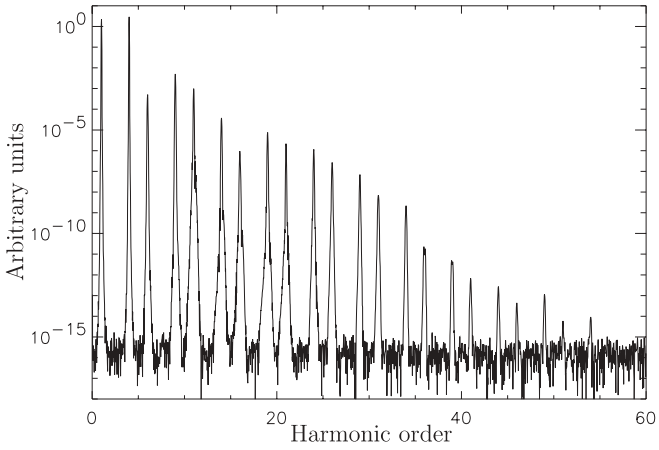

FIGURE 1 Example of a typical harmonics spectrum obtained with two circularly polarized laser fields. The electric field intensities are $E_1 = E_2 = 0.13$. The frequency ratio is $\eta = 4$ and $\omega = 0.0285\pi$

The laser frequency ω has to be chosen carefully. In fact, on one hand we want the lower harmonics not to be affected by the atomic levels; on the other hand we want to approach resonances for the higher harmonics.

The total absorbed energy which leads to harmonic generation is ωN^*, with $N^* = k + \eta(k \pm 1)$. Obviously, N^* is also the order of the emitted harmonic. Therefore, choosing $\omega = \Omega_i/N^*$ provides the 'virtual' resonance, i.e. the N^*th harmonic is resonant with the transition between the ground state and the ith excited state. This is a particular kind of resonance; in fact, neither of the two absorption processes is directly resonant, but their combination is. The ionization rate is almost unaffected by the closeness to such a kind of resonance and always remains very low. Choosing $N^* = 11$ and $i = 1$, the expected resonant frequency is $\omega = 0.0285\pi$. A scheme of such a configuration is presented in Fig. 2. While the harmonic no. 11 is exactly resonant with the first excited state, the harmonics no. 6 and no. 9 are not affected by any energy level.

Once the frequency value was fixed, a series of simulations for different laser field intensities have been performed. As we

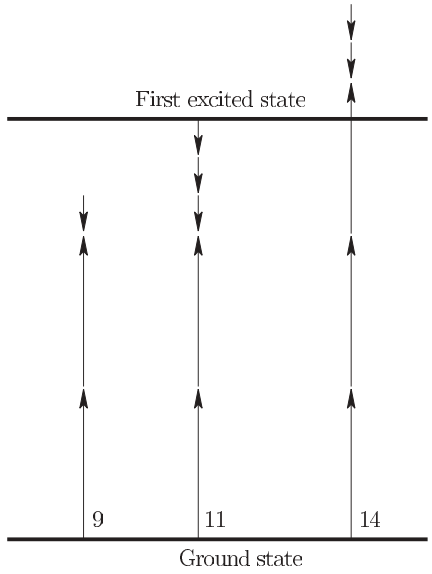

FIGURE 2 Energy scheme for the generation of the harmonics no. 9, no. 11, and no. 14

deal with two different absorption processes, in order to obtain efficient harmonic generation it is necessary that the two separate processes are 'likely', i.e. the probabilities of absorbing the required numbers of photons from each of the two lasers must be of the same order of magnitude.

In Fig. 3 the behavior of the harmonic no. 6 vs. the electric field E_1 for different values of E_2 is plotted. As expected, the intensity of the harmonic increases smoothly with increasing laser intensity. A very different behavior is observed for the harmonic no. 11 (Fig. 4). For relatively low intensities of E_1 the behavior is similar to that of the harmonic no. 6, but when the electric field E_1 becomes stronger a much more complex behavior appears. This is clearly due to the resonance with the first excited state. Actually, the frequency $\omega = \Omega_1/11$ is close to resonance, but not exactly resonant. In fact, one should take into account the level shifts due to the dynamical Stark effect: all levels are expected to be moved upward. Giving an analytical estimation of how much the lower-lying excited states are moved up is a very difficult task already when only a single, linearly polarized laser field is taken into account, let alone in our two-color configuration. In particular, it is difficult to distinguish between the different contributions of the two laser fields.

A possible method for studying how a full resonance affects the harmonic generation is to perform a series of numerical simulations where all parameters but the laser frequency ω are kept constant. Increasing the energy of the photons compensates the shift due to the Stark effect and allows us to reach the shifted excited state again with 11 photons. As the amount of the shift is unknown different values of ω have been used. The results of these simulations are presented in Fig. 5. It appears that the harmonic intensity strongly depends on the exact value of the laser frequency, i.e. a small increase or decrease in the frequency value can change the harmonic intensity by several orders of magnitude. From Fig. 5 one realizes that the full resonance is achieved for $\omega = 0.030\pi$ and $E_1 = 0.07$, leading to an energy shift of about 0.052 a.u. All these values are obtained keeping constant $E_2 = 0.13$. If the value of E_2 is different the previous values for ω and E_1 do not hold any more, although the physics remains qualitatively the same. The massive interferences visible in Fig. 5 are

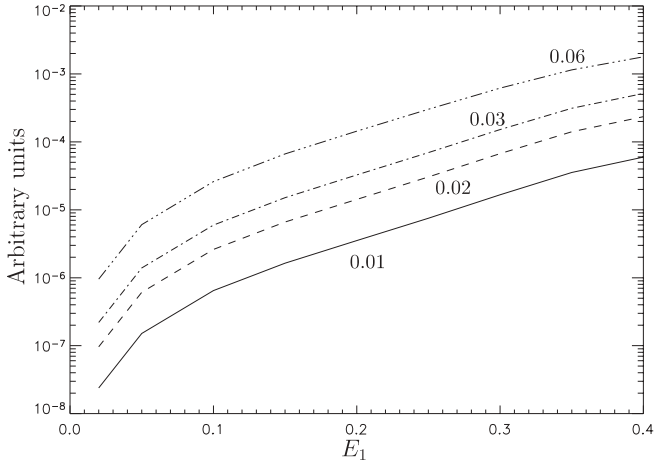

FIGURE 3 Dipole strength of the harmonic no. 6 for different values of E_2. The laser frequency is $\omega = 0.0285\pi$. The behavior is purely perturbative

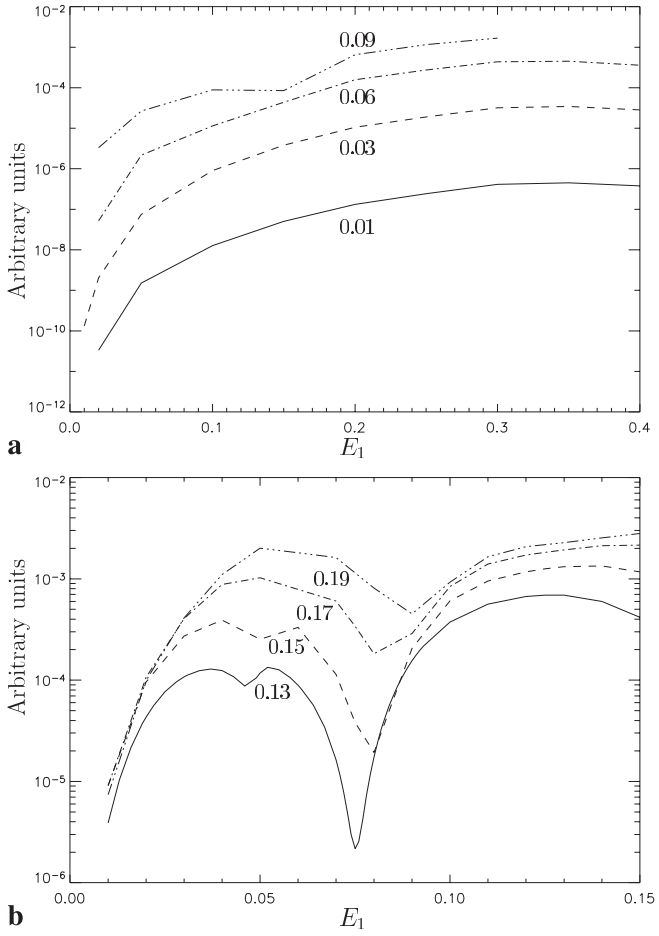

FIGURE 4 Dipole strength of the harmonic no. 11 for low (**a**) and high (**b**) values of E_2. The laser frequency is $\omega = 0.0285\pi$

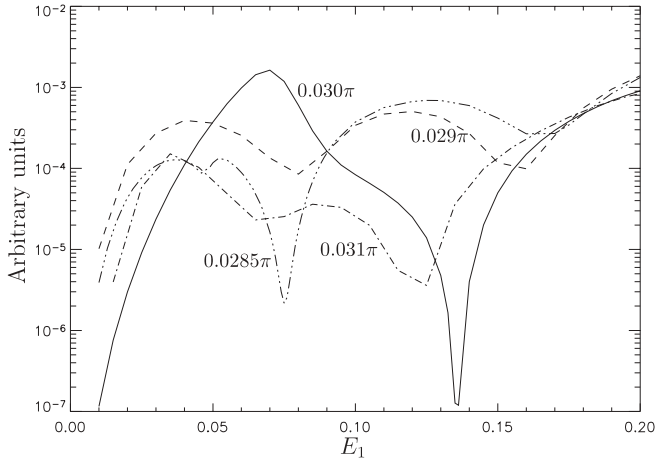

FIGURE 5 Dipole strength of the harmonic no. 11 vs. E_1 for different laser frequencies. $E_2 = 0.13$ is held constant

presumably due in part to the strong dependence of the generalized cross section on the laser frequency [12] and in part to the presence of different processes that may cancel each other or add up. In order to better estimate the dependence of the conversion efficiency with respect to the exact location of the excited state a different kind of study has been carried out. Taking the laser intensities that in Fig. 5 give the maximum harmonic intensity, namely $E_1 = 0.07$ and $E_2 = 0.13$,

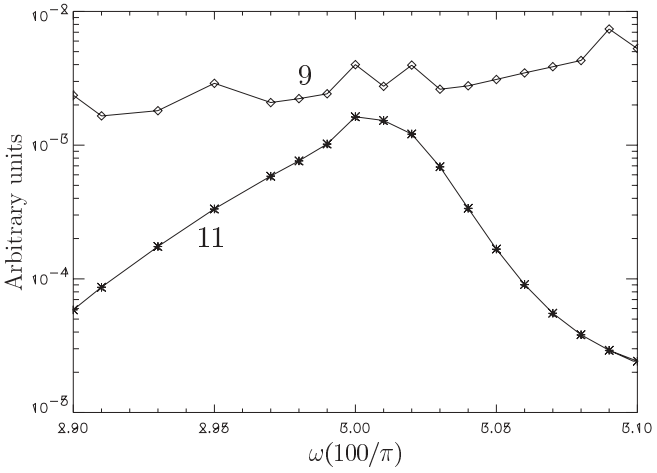

FIGURE 6 Dipole strength of the harmonics no. 9 and no. 11 vs. laser frequency

a series of simulations for different laser frequencies within a small frequency range has been performed. The results of such a study are presented in Fig. 6. While the intensity of the harmonic no. 9 varies slowly, the intensity of the harmonic no. 11 exhibits a strong enhancement due to the resonance. The efficiency of these resonances is comparable with that found in the case of a linearly polarized driving field [13]. Because of the high ionization potential, the enhancement in harmonic generation due to resonances does not lead to a significant increase of the ionization rate, which remains negligible for all frequencies presented.

4 Conclusions

In this work an extensive numerical study of harmonic generation in the two-color coplanar configuration has been presented. In particular, the cases of far-off and near resonance absorption have been addressed. It has been shown that the atomic levels can be used as an important tool in order to enhance significantly the intensity of a particular harmonic without increasing the ionization rate.

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