Two-Color Multiphoton Ionization of Atoms Using High-Order Harmonic Radiation

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Laser-assisted single-photon ionization of an atomic system can be observed in the simultaneous presence of a strong low-frequency laser field and of one of its (weaker) high-order harmonics. Typical photoelectron spectra associated to such two-color processes are simulated via the numerical solution of the time-dependent Schrödinger equation, for a hydrogen atom in the presence of both fields. We address the question of the interplay between this laser-assisted process and above-threshold ionization. Our results show that new sets of two-color experiments can be performed at moderate laser intensities easily attainable by currently operated sources.

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The recent demonstration of the feasibility of using higher-order harmonics of the frequency of an infrared laser, as a coherent and pulsed (soft-)x-ray source, opens new perspectives in relation with its characteristics, which for some applications can compare favorably with those of existing ones [1]. Among these new applications, we wish to address the possibility of performing two-color experiments involving atomic states lying in the continuum, namely, laser-assisted single-photon ionization. It implies that the ionization process results from the absorption of one high-frequency photon together with the simultaneous exchange of laser photons. The observation of a similar process, i.e., laser-assisted Auger transitions, has been recently reported [2].

These processes are, in essence, very different from stepwise excitation schemes, which can involve several laser sources tuned in resonance with real atomic transitions and which are currently used to populate excited atomic states. Indeed, a much simplified picture of laser-assisted single-photon ionization is that of an atom being first brought in a continuum state via the absorption of one single high-frequency photon $\omega_H > E_I$, where E_I is the ionization energy of the atom, followed by the exchange of one or several photons from the laser field. Within this approximate model, which has been much discussed in the literature [3], free-free transitions (FFT's) would take place between atomic continuum states, thus giving rise to a photoelectron spectrum similar, in some respects, to the ones obtained in laser-assisted scattering experiments [4]. We wish to stress, however, that the *ab initio* treatment we present here does not rely on such crude approximations. On the other hand, the two-color ionization process discussed here, using two sources with commensurable frequencies, one of them being an (odd) high-order harmonic ($N \geq 13$) of the other, adds new features to the physics of the process.

Only a few two-color ionization experiments have been reported so far and, besides the pioneering work by Muller, van Linden van den Heuvell, and van der Wiel [5], in which two different lasers operated in the UV and IR ranges were used, no phenomenon similar to the one considered here has been observed yet. Let us mention, however, two-color experiments which have been realized with low-order harmonics, typically second or third order with comparable intensities, in atoms [6] and molecules for the coherent control of chemical reactions [7]. Another set of experiments has been performed in negative ions [8], one of the main motivations being to address the then ongoing discussion of the role of the ponderomotive energy $U_p = F_0/4\omega_L^2$, where F_0 is the laser field amplitude and ω_L its frequency (atomic units are used, unless otherwise stated).

As already mentioned, the possible observation of such two-color effects has been predicted and discussed in several instances, either on general grounds or for simplified atomic models [3]. It is only recently that calculations taking into account the whole complexity of the atomic structure have been reported, based either on the Floquet approach [9,10] or on a numerical solution of the time-dependent Schrödinger equation [11]. These calculations were restricted to low harmonic orders (see, however, Ref. [10]) and did not address the same issues as the ones considered here. See also Ref. [12] for a perturbative discussion in which no restriction was imposed on the frequencies.

We present here the results of a numerical simulation for a (3D) hydrogen atom in the presence of two fields. The calculation is conducted along a method similar to the one used by Schafer and Kulander in Ref. [11] and described in more detail in Ref. [13]. We turn now to the discussion of several interesting features of this class of processes, as they result from our calculations.

In the presence of an intense low-frequency ω_L laser pulse, above-threshold ionization (ATI) is the dominant ionization process. In ultrashort laser pulses, the corresponding photoelectron spectrum consists of lines, the positions of which are given by conservation of energy, provided the ac Stark shift $\approx U_p$ of the continuum limit is properly taken into account. Their respective magnitudes and structures, as obtained in experiments, can

be recovered through nonperturbative calculations, even in complex atoms [14]. For the sake of illustration, we present in Fig. 1 the ATI spectrum we have obtained for a ground state hydrogen atom in the presence of a short pulse with a frequency equal to that of a Ti:sapphire laser operated at $\omega_L = 1.55$ eV. The time dependence of the pulse envelope is trapezoidal; its duration is eight cycles ($T_L = 110$ a.u.), with one-cycle linear turn-on and turn-off and an intensity $I_L = 1.75 \times 10^{13}$ W/cm^2. As shown in Fig. 1, the distribution of the photoelectron lines is typical of ATI spectra.

On the other hand, in the presence of a pulse with the same duration, containing the sole harmonic radiation with a frequency $\omega_H = 13\omega_L$ with a much lower intensity (here $I_H/I_L \approx 10^{-5}$), single-photon ionization takes place. This gives rise to a single photoelectron peak, as also shown in Fig. 1.

Interesting new physical effects are expected if the atom experiences the two fields simultaneously. Indeed, ionization can now result from the simultaneous absorption of the harmonic photon and of the exchange of several additional photons from the laser field, via absorption or stimulated emission. The signature of such processes is the presence in the photoelectron spectra of equally spaced satellite peaks, located on each side of the one resulting from the single-photon absorption. We note that, as the high-frequency field is a (odd) multiple of the lasers, the satellites are located at the same energies as those of the ATI peaks, as shown below.

To illustrate the main features of the process, we first consider the case of a ground state hydrogen atom, in the presence of a radiation pulse containing the fundamental frequency $\omega_L = 1.55$ eV of a Ti:sapphire laser and its 13th harmonic $\omega_H = 20.15$ eV. The pulse time dependence is the same as before. The harmonic intensity is kept fixed at $I_H = 3 \times 10^8$ W/cm^2, which is within reach of currently developed harmonic sources, while the laser peak intensity is varied among $I_L = 5 \times 10^{11}$ W/cm^2 [Fig. 2(a)], $I_L = 3 \times 10^{12}$ W/cm^2 [Fig. 2(b)], and $I_L = 1.75 \times 10^{13}$ W/cm^2 [Fig. 2(c)].

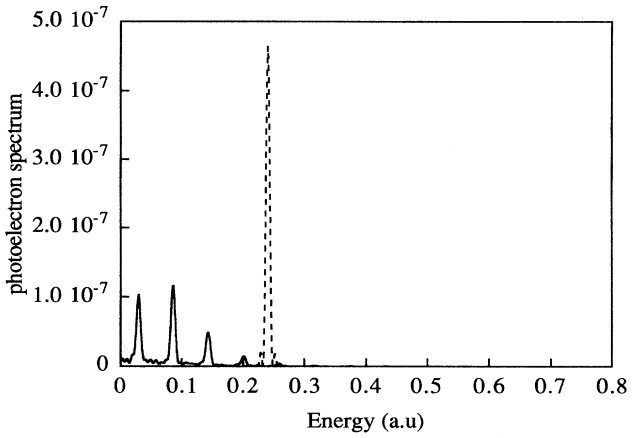

FIG. 1. Typical ATI spectrum (full line) as obtained from the occupation density of the positive energy atomic states, after a radiation pulse containing the fundamental frequency of a Ti:sapphire laser operated at $\omega_L = 1.55$ eV, with intensity $I_L = 1.75 \times 10^{13}$ W/cm^2. The dashed line represents the single-photon spectrum one would obtain with the harmonic field alone, $\omega_H = 13\omega_L$, with intensity $I_H = 3.10^8$ W/cm^2; see text.

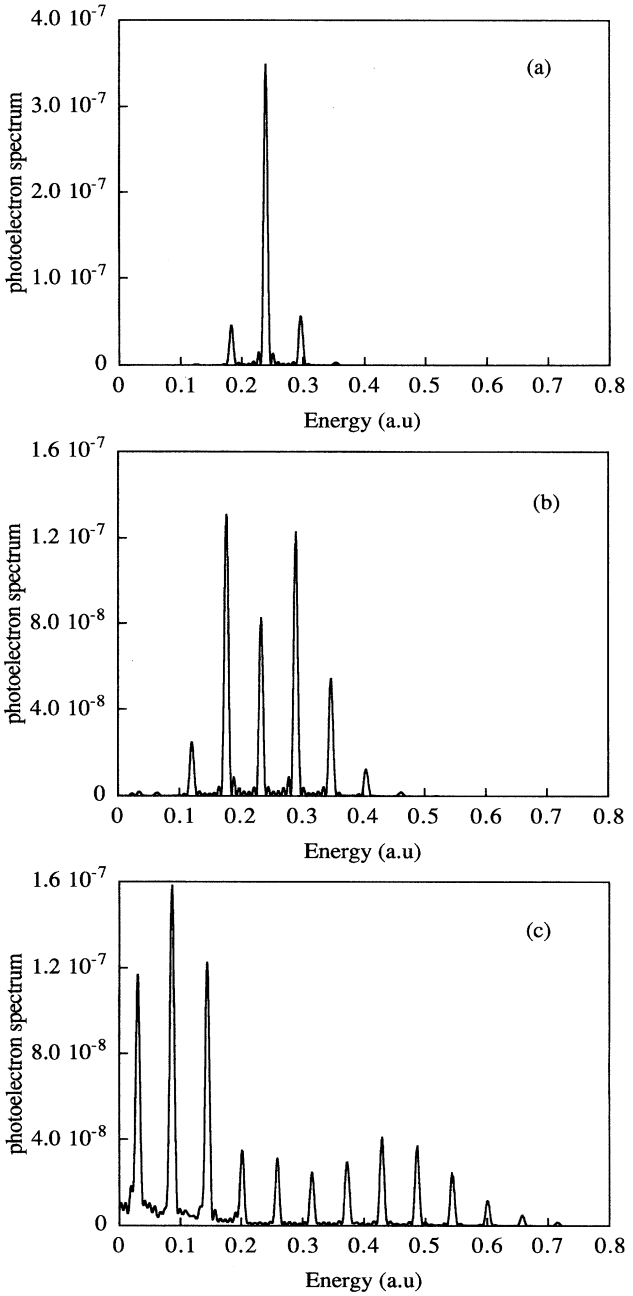

FIG. 2. Effect of the laser intensity on two-color photoelectron spectra, for radiation pulses containing the fundamental frequency of a Ti:sapphire laser, $\omega_L = 1.55$ eV and its 13th harmonic with a fixed intensity $I_H = 3 \times 10^8$ W/cm^2. (a) $I_L = 5 \times 10^{11}$ W/cm^2; (b) $I_L = 3 \times 10^{12}$ W/cm^2; (c) $I_L = 1.75 \times 10^{13}$ W/cm^2 (see text). Note the changes of scales.

Note that the computations have been performed with the $\mathbf{A} \cdot \mathbf{p}$ representation of the atom-field interaction, the size of the radial box has been chosen with $r_{max} = 1250$ a.u., and we have included up to $l_{max} = 20$ angular momenta in the partial wave expansions. We have checked that our results were converged and that they were similar to the ones obtained using the $\mathbf{E} \cdot \mathbf{r}$ gauge.

At low laser intensity $I_L = 5 \times 10^{11}$ W/cm^2 the two-color photoelectron spectrum displays only two satellite lines located symmetrically on each side of the one associated to single-photon ionization, as shown in Fig. 2(a). As expected, if the laser intensity is higher ($I_L = 3 \times 10^{12}$ W/cm^2), the number of satellite lines increases; see Fig. 2(b). Let us emphasize that for the short pulse length considered here, the ATI peaks are not visible on the linear scale used for the figures [in fact, they are barely visible at very low energy in Fig. 2(b)].

Such spectra, which are characteristic of laser-assisted single-photon ionization, display several interesting features. One is that, as the laser itself has a too low intensity to significantly contribute to the ionization process, the global ionization probability is conserved, which is confirmed by comparing the areas under the photoelectron peaks. Another feature of interest is that for the laser-assisted spectrum, the peaks are slightly shifted as a result of the ac Stark shifts of the energy levels. The analysis of the numerical results shows that the observed shift corresponds approximately to the ponderomotive energy U_p, as expected for photoelectrons within the laser field. In principle, the harmonic field induces also an ac Stark shift of the atomic levels. It is, however, negligible at the frequencies and intensities considered here. Let us eventually mention that we have verified that the results shown are quite different from estimates derived from simplified analysis using Coulomb-Volkov waves; see, for instance, Leone *et al.* in Ref. [3]. This point, as well as comparisons with another model introduced by Rzazewski and co-workers [3], will be discussed in more detail elsewhere.

We consider now the interesting case in which ATI and one-photon ionization have comparable probabilities. Satellites and ATI peaks are then degenerate and interference effects are expected. Note that such interference effects are phase dependent, see Refs. [3,6,7,9–11], and one expects notable changes in the spectra, depending on the relative phases of the two fields. A typical case, in which there is no phase shift between the two fields at $t = 0$, is illustrated in Fig. 2(c), for $I_L = 1.75 \times 10^{13}$ W/cm^2. An interesting feature of the results is the notable increase observed in the magnitudes of the photoelectron lines, as compared to the (single-color) ATI spectrum. These changes directly result from the fact that more quantum paths can lead to the same final energy states.

In spite of the fact that the question of the control of the relative phase between the laser field and one of its higher harmonics is still an open problem for the currently developed harmonic sources [1], we have illustrated this point in more detail in Fig. 3. In the figure are shown the variations of the intensities of two photoelectron peaks, which display a typical dependence on the relative phase at $t = 0$ between the laser field and its harmonic. As expected, one observes important changes in the magnitudes of the peaks when the phase is changed from 0 to $\pi/2$ and to π. Note that the changes are quite important in spite of the fact that, in contrast with most previous studies on phase effects where the fields had comparable intensities, the field strengths are very different, since the ratio $I_H/I_L \approx 10^{-5}$. Let us note also that, by studying the phase dependence of such two-color ionization yields, one should be able to get an estimate of the relative values of ATI and laser-assisted transition amplitudes.

In actual experiments involving strong laser fields, it might prove difficult to distinguish the satellites from ATI. A simple way to overcome this difficulty is to use the second harmonic of the laser as an intense low-frequency source, instead of the fundamental frequency itself. Then, if the high-frequency field is an odd harmonic of the lasers, the two sets of peaks can be unambiguously assigned as their energies differ by ω_L. More precisely, the ATI peaks are located at energies resulting from the absorption of $n(2\omega_L)$ by the atom, while the satellites will be located at energies corresponding to the absorption of $\omega_H \pm n'(2\omega_L)$, i.e., odd multiples of the laser frequency as ω_H is itself an odd harmonic of ω_L. This situation is illustrated in Fig. 4.

Shown in Fig. 4(a) is the ATI spectrum resulting from the ionization of the atom by a pulsed field with frequency $2\omega_L = 2.34$ eV (second harmonic of a Nd:YAG laser), a trapezoidal time dependence with two-cycle turn-on and turn-off, and a constant amplitude $I_L = 1 \times 10^{13}$ W/cm^2 duration of ten cycles. At such an intensity, which is still moderate, the magnitudes of the photoelectron peaks reg-

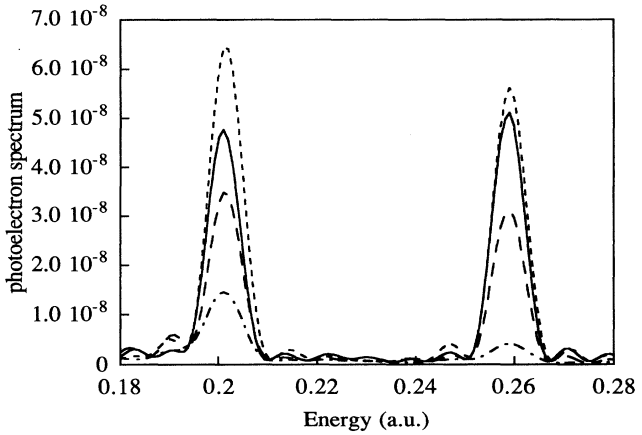

FIG. 3. Dependence of two peaks of the two-color photoelectron spectrum on the initial relative phase ϕ of the laser and of its 13th harmonic (Ti:sapphire laser $\omega_L = 1.55$ eV, see text). The dot-dashed line represents the ATI spectrum for the laser alone. Long-dashed line, $\phi = 0$; full line, $\phi = \pi/2$; dashed line, $\phi = \pi$.

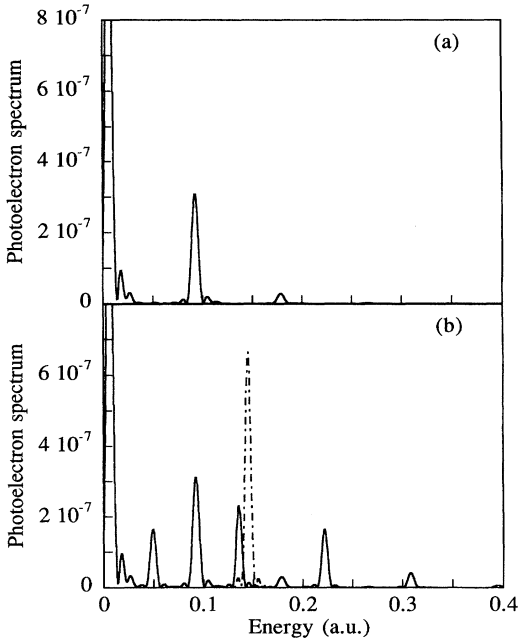

FIG. 4. Comparison of (a), ATI spectrum for the second harmonic of a Nd:YAG laser $2\omega_L = 2.34$ eV and (b), two-color ionization photoelectron spectrum for the same laser pulse containing, in addition, a weaker component at the 15th harmonic of the fundamental frequency (see text). The dot-dashed line represents the single-photon spectrum one would obtain with the harmonic field alone.

ularly decrease with their order, as shown in the figure. If the radiation pulse contains, in addition, the 15th harmonic of the Nd:YAG laser ($\omega_H = 17.55$ eV), with an intensity $I_H = 2 \times 10^8$ W/cm^2, the photoelectron spectrum is again strongly modified, as shown in Fig. 4(b). As the two processes can no longer interfere, unchanged ATI peaks are still visible. However, new peaks, corresponding to laser-assisted single-photon ionization, are also present. They are separated from the ATI peaks by 1.17 eV, i.e., the fundamental laser photon energy, and can be identified without ambiguity.

As a final comment, we wish to mention that our results demonstrate the feasibility to observe laser-assisted effects involving continuum states using high-order harmonics, even at quite moderate laser intensities, at least much weaker than the ones currently used in multiphoton physics. Important changes are predicted in the photoelectron spectra, as compared to the ones obtained with only one source, and interference effects between ATI and two-color ionization are shown to be observable when the intensity of the high-order harmonic is of the order of 10^{-5} times that of the laser with $I_L \approx 10^{13}$ W/cm^2. Among the potential applications, let us mention the possibility to reach continuum atomic states with angular momenta different from the ones reached through a single-photon ionization process. This opens the perspective to probe a variety of final atomic states, which are currently out of reach of conventional high-energy photoionization spectroscopies, in the vacuum ultraviolet and soft-x-ray ranges.

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