Phase dependence of $(N+1)$-color $(N>1)$ ir-uv photoionization of atoms with higher harmonics

Valérie Véniard, Richard Taïeb, and Alfred Maquet
Laboratoire de Chimie Physique-Matière et Rayonnement, Université Pierre et Marie Curie, 11, Rue Pierre et Marie Curie, 75231 Paris Cedex 05, France
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We discuss, via a numerical calculation, the main properties of atomic photoelectron spectra, as they would be obtained by using a radiation pulse containing $N+1$ frequencies associated with a ''Dirac comb'' of N higher harmonics together with the laser which has been used to generate them. We address more precisely the physically relevant situation in which the harmonics have much weaker intensities than the one of the laser. In such $(N+1)$-color photoionization processes, the atom can simultaneously absorb harmonic uv photons and exchange, i.e., absorb and/or emit, (via stimulated emission) laser ir photons. We have simulated the photoelectron spectra by numerically solving the time-dependent Schrödinger equation for a three-dimensional hydrogen atom in the presence of the radiation pulse. Our results show that, everything else being kept fixed, the magnitudes of the photoelectron peaks are strongly dependent on the difference of phase between successive harmonics. This strong dependence results from interference effects taking place between competing quantum paths leading to a given final state. An interesting feature is that these interferences involve transitions in the continuum states of the atom and do not depend on resonances in the discrete spectrum. Another interesting outcome of our study is to show that such effects should be observable with currently developed harmonic sources. [S1050-2947(96)01007-4]

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I. INTRODUCTION

The main motivation of the present paper is to address the possibility to perform a class of $(N+1)$-color $(N>1)$ photoionization experiments, which fully exploit the unique characteristics of high-order harmonic radiation emitted by atoms in the presence of a strong laser field [1]. Amongst these characteristics is the fact that the harmonic emission spectrum is constituted of coherent uv (and even soft x-ray) radiation lines with equally spaced frequencies and comparable intensities, the spacing being twice the frequency ω_L of the ir laser which has been used to generate the harmonics. We shall outline the main features of the multicolor photoelectron spectra obtained with such sources and shall consider more particularly the possibility to determine the relative phases of the harmonics themselves, these phases being one of the key parameters governing the shape of the spectra.

In a previous work, we had considered two-color ir-uv multiphoton ionization processes using simultaneously the fundamental of a strong ir laser and one of its higher uv harmonics, the latter having a frequency high enough to ionize the atom, through single-photon absorption [2,3]. In such conditions, ionization can result from the absorption of the high-frequency photon together with the exchange of one or several laser photons. The simulations were carried out via an *ab initio* numerical resolution of the time-dependent Schrödinger equation (TDSE) for a three-dimensional (3D) hydrogen atom, and our results confirmed the feasibility of observing such processes with currently developed harmonic sources. Experiments, designed to explore this class of processes are currently under way [4], see also Ref. [5].

Our original idea was to investigate the main features of such laser-assisted single-photon ionization (LASPI) processes and to discuss the modifications of the photoelectron spectra which may arise as a result of interferences with the above-threshold ionization (ATI), which take place whenever an intense enough laser field is present. One of our motivations was to explore the possibility to achieve a, at least partial, coherent control of the photoelectron current. Note that in such a scheme, one uses uv harmonics with an intensity which is much lower than the one of the ir laser. This contrasts somewhat with other currently explored scenarios relying on the use of lower harmonics with intensities comparable to the laser's [6].

One of the main outcomes of our simulations was to evidence the prominent role of the harmonic phase. Indeed, the phase difference between the laser and its harmonic is one of the key parameters which govern the relative intensities of the photoelectron peaks in the two-color spectra. More precisely, it appears that the possibility to achieve a partial coherent control of the ionization process, strongly depends on the harmonic phase. Note also that another most important instance, in which the phases of the harmonic components of the atomic dipole play a determining role, is related to the propagation of the harmonics in the macroscopic medium in which they are created [7]. These phase-matching properties are essential, in fact, to design new harmonic-based, vuv or soft-x-ray, coherent sources.

Although it has recently been shown, through a semiclassical description [8] of harmonic generation by a single atom on using a short-range potential [9], that the harmonic phases vary monotonously with the intensity of the driving laser field [7,9,10], their actual determination is still an open problem. One of the objectives of the present paper is to show that a detailed study of multicolor photoionization spectra, involving high harmonics and the (attenuated) laser field which has been used to generate them, can provide interesting information on the phase differences between successive harmonics. With this end in view, we have taken advantage of the fact that the harmonic spectra, emitted by laser-driven

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atoms, display a typical plateau in which successive (odd) harmonics have comparable intensities [1,10]. An idealized representation of these spectra is that of a Dirac comb, with equally spaced frequencies separated by $2\omega_L$, where ω_L is the laser frequency. Then, if one simulates an $(N+1)$-color ionization experiment with a Dirac comb constituted of N harmonics, in the presence of the (attenuated, this point is important) laser field, the resulting energy-resolved photoelectron spectrum is extremely sensitive to the phase differences between the harmonics. This feature, which is typical of such $(N+1)$-color spectra, will be illustrated in the following sections.

We shall briefly present the model and the techniques used to perform the calculations in Sec. II. Then, in Sec. III, we shall specialize the calculation to treat a $(3+1)$-color scenario which is relevant to discuss recent experimental attempts to observe such processes. The more general case of an harmonic source constituted of a ''Dirac comb,'' is treated in Sec. IV and a simplified discussion of the phase-dependent interference patterns in the photoelectron spectra, will be given in Sec. V. The paper ends with a brief conclusion.

II. MULTICOLOR PHOTOELECTRON SPECTRA WITH HIGH-ORDER HARMONIC RADIATION

Harmonic spectra generated by atoms submitted to an intense laser pulse can be schematically represented as shown in Fig. 1(a). The harmonics within the plateau have approximately equal intensities and a subset of them can be idealized as a Dirac comb, see Fig. 1(b) [10]. One can use such a finite subset to perform single-photon ionization experiments in atoms, provided the frequencies are larger than the atomic ionization frequency. Then the photoelectron spectrum obtained from the above Dirac comb, would exhibit the typical shape shown in Fig. 1(c). See Ref. [11], for an experimental realization of such spectra. Note that in experiments one can use multilayer mirrors in order to refocus and select some harmonics. In a recent implementation of such a scheme [4] three harmonic frequencies were present after reflection, as a result of the finite bandwidth of the response (in the uv range considered) of the mirror used.

If, in addition to the harmonic light, the ir laser radiation is also present, the photoelectron spectra can be strongly modified [2,3]. One expects, and one has indeed already observed [4], the presence of equally spaced new lines, separated from the preceding ones by ω_L. At relatively low laser intensities, namely, if the laser radiation does not contribute to the ground-state depletion via the ATI, there is a global conservation of the total ionization probability, and the role of the laser is reduced to redistribute the photoelectrons amongst the newly accessible continuum states. Accordingly, the photoelectron peaks resulting from the absorption of the uv photons will be depleted, the difference with the laser-free spectrum being transferred to the new satellite peaks. As we shall show below, the relative magnitudes of the laser-assisted ir-uv photoelectron peaks strongly depend on the relative phases of the harmonics.

The method we have used to simulate the photoelectron spectra has been previously described in Refs. [2,3]. Here we only briefly mention that it is based on a numerical resolution of the TDSE for a 3D hydrogen atom in an external radiation field, which is much similar to the one implemented by Kulander, Schafer, and Krause [12]. We have modeled the time dependence of the multicolor pulse with a trapezoidal envelope of a total duration of eight laser cycles including one-cycle turn on and turn off. For instance, for the laser pulse one has

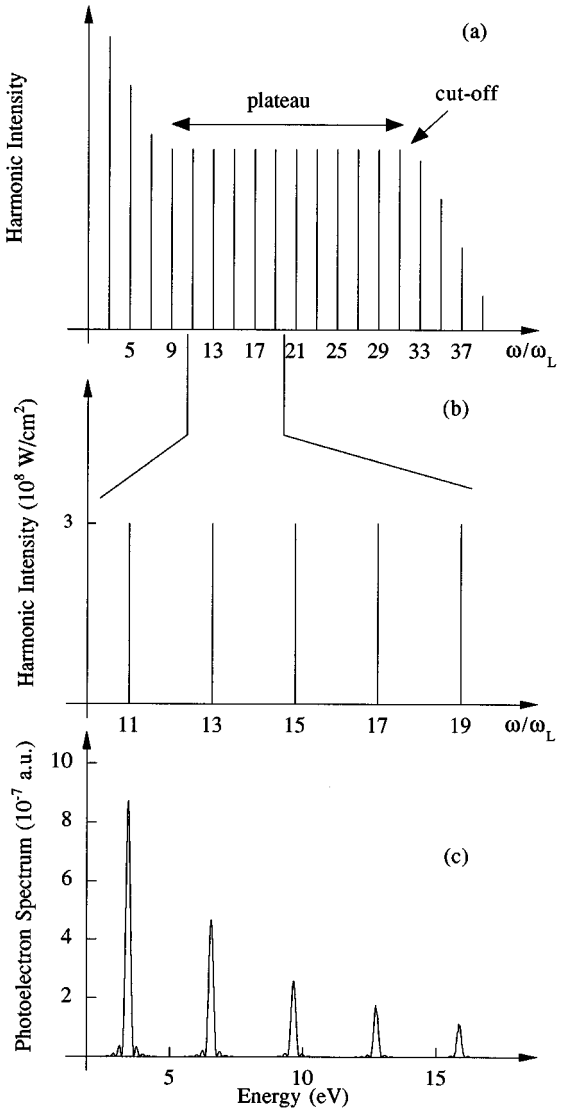

FIG. 1. (a) Schematic representation of an harmonic spectrum; (b) enlargement of the spectrum showing the ''Dirac comb'' of harmonic frequencies used in the calculation; (c) Photoelectron spectrum obtained from hydrogen 1s with a radiation pulse containing the harmonics shown above: $\omega_L = 1.55$ eV, $I_H = 3\times10^8$ W/cm^2 (see text).

$$F_1(t) = (t/T_L)F_{1,\max}\sin(\omega_L t), \quad 0 \leqslant t < T_L$$

$$F_1(t) = F_{1,\max}\sin(\omega_L t), \quad T_L \leqslant t < 7T_L$$

$$F_1(t) = [8-(t/T_L)]F_{1,\max}\sin(\omega_L t), \quad 7T_L \leqslant t \leqslant 8T_L . \tag{1}$$

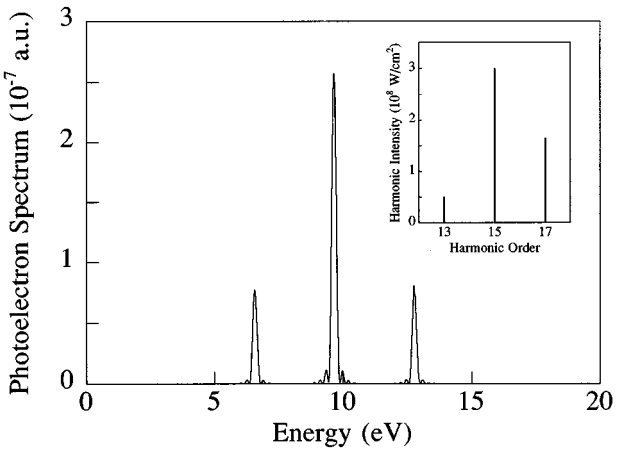

FIG. 2. Three-color photoelectron spectrum, for a radiation pulse containing the 13th, 15th, and 17th harmonics of a Ti:sapphire laser, $\omega_L = 1.55$ eV. The inset shows the corresponding harmonic intensities needed to reproduce the spectrum (see text).

Unless otherwise mentioned the laser frequency is $\omega_L = 1.55$ eV (Ti:sapphire laser). The photoelectron spectra are obtained from a spectral analysis of the atomic wave function after the end of the pulse [13]; see also Ref. [3], for more details regarding the technicalities of our computations. We turn now to a discussion of the $(3+1)$-color spectra, a scenario which is relevant to qualitatively discuss recent experiments.

III. (3+1)-COLOR PHOTOIONIZATION SPECTRA

We have first addressed the case in which three harmonics with unequal intensities are used. This situation approximately models preliminary experimental attempts to observe the process in He, using uv harmonics of a Ti:sapphire laser at the femtosecond laser facility in Palaiseau, in which a multilayer mirror was used to select the needed frequencies [4]. The photoelectron spectrum, obtained with the sole harmonics, then consists of three peaks with relative magnitudes as shown in Fig. 2.

Before proceeding, a few comments are in order. The spectrum shown in Fig. 2 has been simulated in hydrogen, with the harmonics 13, 15, and 17 of the laser frequency $\omega_L = 1.55$ eV. We have chosen these frequencies in order to reproduce a photoelectron spectrum with the same characteristics as the one obtained in the above-mentioned experiment. Note that we have checked, by comparing with a time-dependent Hartree-Fock calculation for He, that the main conclusions which can be drawn from our study in hydrogen are not altered when considering a more sophisticated model, at least in the low-intensity regime considered here. We have kept with the calculation in hydrogen for the sake of computational convenience. Then, in order to reproduce the spectrum in Fig. 2, the intensities of each harmonic component must be as shown in the inset. In fact, any departure from these relative intensities would notably modify the photoelectron spectrum, a feature which provides a quite stringent test on the relative intensities of the harmonics and, consequently, on the frequency dependence of the reflectivity of the mirrors. Note that in the course of the reflection process the phases of the harmonics are changed. The corresponding changes can be estimated and we shall not discuss this point here.

Another feature in our treatment, which deserves some comments, is that we have assumed that the harmonic pulses have the same duration, namely, the one of the laser which has been used to generate them, i.e., eight laser cycles as explained above. This may seem *a priori* unrealistic as it is known that the actual duration of the harmonic emission, not only varies with the harmonic order, but can be also much shorter than the laser pulse [14,15]. One should keep in mind, however, that the relevant time length is given by the overlap between the pulses, provided the laser itself does not significantly contribute to the ionization process, a condition which is fulfilled in the present discussion. It appears also that, at the relatively low intensities we shall consider here, the magnitudes of the peaks are directly proportional to both the intensity and duration of the pulse, as expected in a lowest-order perturbative regime. One can then play with these two parameters in order to reproduce a given spectrum, at least within an overall multiplicative factor. For instance, for the chosen duration of the trapezoidal harmonic pulses, the photoelectron spectrum shown in Fig. 2 has been obtained for the following maximum harmonic intensities shown in the inset: $I_{13} = 5\times10^7$ W/cm^2, $I_{15} = 3\times10^8$ W/cm^2, $I_{17} = 1.65\times10^8$ W/cm^2. Once these relative intensities are determined, one can consider the situation in which one adds a fourth color, namely, the laser radiation with a moderate intensity $I_1 = 8\times10^{11}$ W/cm^2. This corresponds to an intensity regime in which lowest-order perturbative estimates are expected to account fairly for the processes. Then, as shown in Figs. 3, some new photoelectron lines do appear resulting predominantly of two-photon processes involving the exchange of one laser photon in addition to the absorption of the uv photons [16]. In fact, in order to investigate the role of the relative phases of the harmonics, we have considered the following time dependences of the fields in the time interval $T_L \leqslant t \leqslant 7T_L$: $F_1(t) = F_{1,\max}\sin(\omega_L t)$ for the laser and $F_{2N+1}(t) = F_{2N+1,\max}\sin[(2N+1)\omega_L t + \phi_{2N+1}]$ for the harmonics, with $2N+1 = 13,15,17$ and where the phases ϕ_{2N+1} are the phases differences between the harmonics and the laser at $t = 0$.

Shown in Fig. 3(a) is the photoelectron spectrum for the (oversimplified) test case in which the three harmonics have the same phase as the one of the laser, i.e., $\phi_{13} = \phi_{15} = \phi_{17} = 0$. One observes the presence of new, relatively small, satellite peaks. Comparing with the laser-free spectrum displayed in Fig. 2, one notes also that the new peaks have grown at the expense of the original ones which are slightly depleted. This was expected since the overall ionization probability is unchanged, the laser itself being not intense enough to contribute to the ground-state depletion, for such a short pulse.

Changing the phases can dramatically modify the photoelectron spectrum. For instance, if the phases differences are: $\Delta\phi_{15,13} = \phi_{15} - \phi_{13} = \pi$, and $\Delta\phi_{17,15} = \phi_{17} - \phi_{15} = \pi$, the spectrum changes as shown in Fig. 3(b) (note the change of scale). We have introduced the phase differences between successive harmonics because it appears that they are the relevant parameters for discussing these effects. This point will be addressed in more detail in Sec. V below. For this

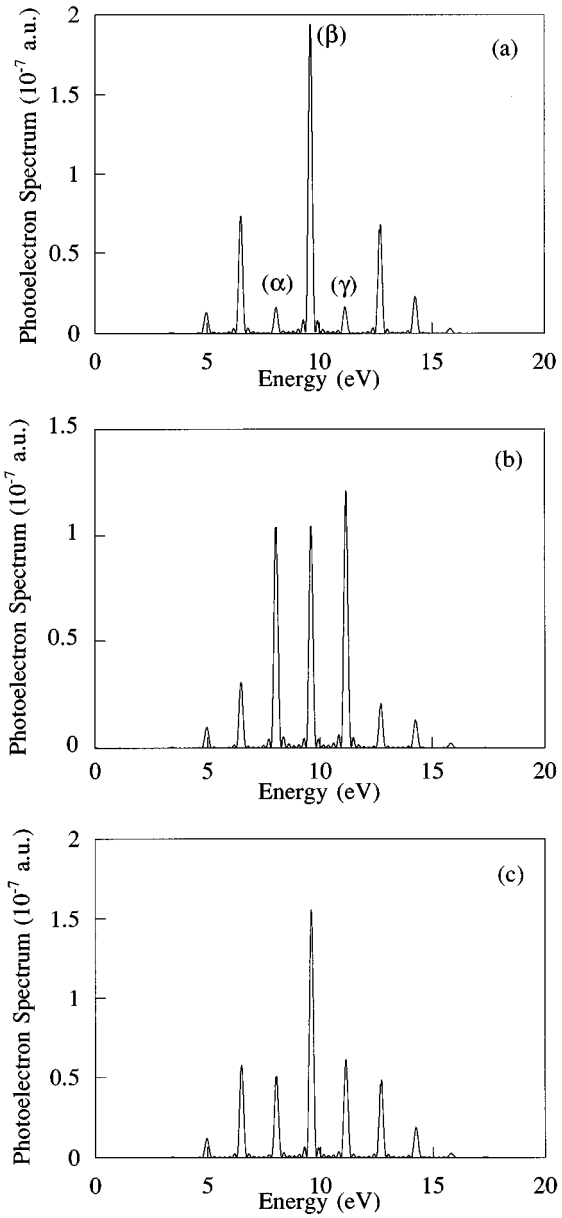

FIG. 3. Effect of the harmonic phases on the $(3+1)$-photoelectron spectrum, with $\omega_L = 1.55$ eV, $I_L = 8\times10^{11}$ W/cm^2, the three-color spectrum being shown in Fig. 2; (a) $\Delta\phi_{15,13} = \Delta\phi_{17,15} = 0$; (b) $\Delta\phi_{15,13} = \Delta\phi_{17,15} = \pi$; (c) $\Delta\phi_{15,13} = \Delta\phi_{17,15} = \pi/2$.

choice of phases, one observes that the satellites are much bigger: their magnitudes are now comparable to the ones of the original peaks which, in turn, are significantly depleted. The strong dependence of the sizes of the satellite peaks on these phases results from the fact that each of them can be reached via, (at least two) distinct quantum paths. It indicates also that the amplitudes associated with each of these paths have comparable magnitudes, so that strongly destructive interferences can occur. We shall also address this point in more detail in Sec. V.

In Fig. 3(c), we have presented a $(3+1)$-color photoelectron spectrum in which the relative heights of the peaks are similar to the ones observed in the above-mentioned experiment [4]. It is shown here as an illustrative example of the feasibility of determining the relative phases of the harmonics, starting from an experimental spectrum. For this spectrum, the respective phases differences are $\Delta\phi_{15,13} = 90°$ and $\Delta\phi_{17,15} = 90°$, respectively. We turn now to to a brief description of the method used to determine these phases.

The main idea is to monitor the variations of the relative heights of the peaks in terms of the phase differences between the harmonics and then to find the optimal combination needed to fit the spectrum. Here, more precisely, for the above set of fields intensities, we have monitored the heights of the peaks labeled (α), (β), and (γ) in Fig. 3(a). Note that these peaks can be associated with the following net absorption of energies: $E_\alpha = 14\omega_L$ $E_\beta = 15\omega_L$ $E_\gamma = 16\omega_L$, and that the peaks (α) and (γ) are reached through the exchange of at least one laser photon, in addition to the absorption of one uv (odd) harmonics. These peaks are obviously the most sensitive to interference effects. In Fig. 4(a) are reported the variations of the height of the peak (α), in terms of the phase difference $\Delta\phi_{15,13}$, the other difference $\Delta\phi_{17,15}$ *being kept fixed at the indicated values*. Parallely, are shown the variations of the heights of the peaks (β) Fig. 4(b) and (γ) Fig. 4(c).

Several interesting conclusions can be drawn from the perusal of this set of figures. First, the peak (α) is the one which is the most sensitive to the phase difference $\Delta\phi_{15,13}$. This results from the fact that it can be reached via two dominant quantum paths involving either the absorptions of the 13th harmonic and of one laser photon, or the absorption of the 15th harmonic and the (stimulated) emission of one laser photon. In such a case, the resulting probability amplitude depends strongly on the phase difference between the two harmonics, as will be shown in Sec. V. One observes that the maximum height for (α) is reached for values of $\Delta\phi_{15,13}$ slightly larger than π [compare with Fig. 3(b), in which $\Delta\phi_{15,13} = \pi$]. The interesting point is that the variations of this peak are linked to the ones of the peak (β), see Fig. 4(b), in the sense that any growth of (α) occurs at the expense of (β). This is a direct consequence of the overall conservation of the ionization probability, as already mentioned. Things are, however, slightly more complicated as a result of the fact that the peak (β) is depleted also when transitions leading to the peak (γ) take place. These are, however, less affected by changes in $\Delta\phi_{15,13}$, as can be checked on Fig. 4(c).

Performing a similar analysis for the influence of $\Delta\phi_{17,15}$, one can univocally find the set of phases differences which permit to fit the spectrum, Fig. 3(c). As already mentioned the values found to fit this almost symetrical spectrum are $\Delta\phi_{15,17} = \Delta\phi_{15,13} = 90°$. Note that this result seems to indicate that in the above-mentioned experiment the harmonic phases vary monotonously, not only with the laser intensity but also with their order. As we shall show next the same analysis can be straightforwardly extended to the cases in which more than three harmonics are present.

IV. $(N+1)$-COLOR $(N>3)$ PHOTOIONIZATION SPECTRA

It is enough to consider the case $N=5$ to illustrate the main features of such spectra. If the uv harmonics have equal intensities, the five-color photoelectron spectrum obtained

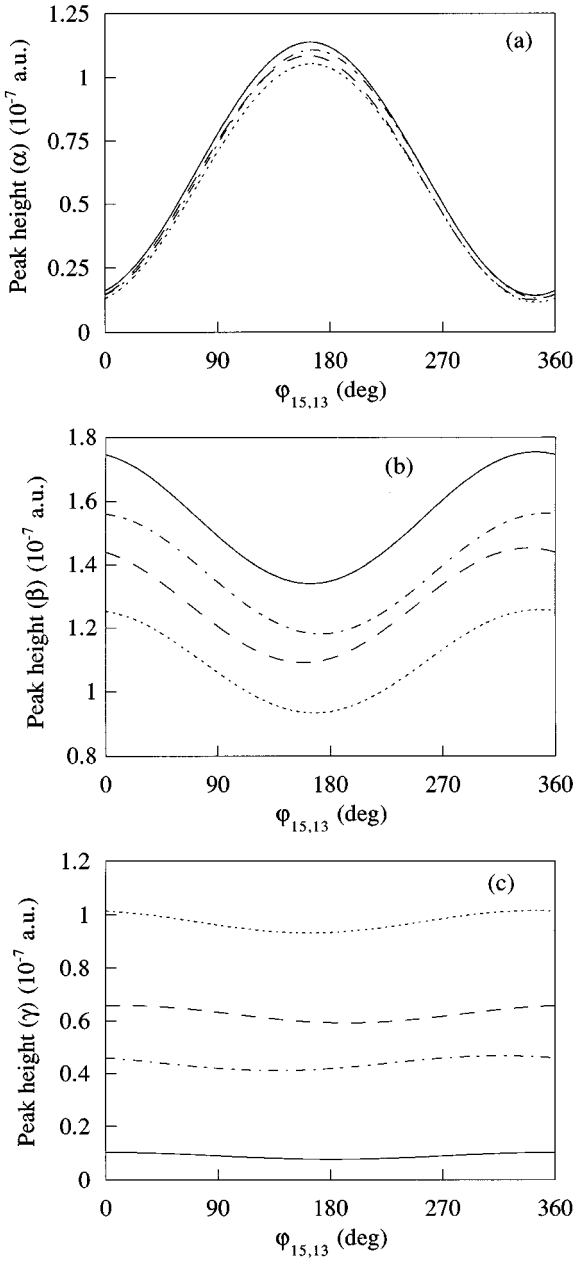

FIG. 4. Peak heights of the photoelectron spectrum as a function of the phase difference $\Delta\phi_{15,13}$, $\Delta\phi_{17,15}$ being kept fixed at the indicated values: full line, $\Delta\phi_{17,15}=0$; dashed line, $\Delta\phi_{17,15}=\pi/2$; dotted line, $\Delta\phi_{17,15}=\pi$; dashed-dotted line, $\Delta\phi_{17,15}=3\pi/2$. The graphs (a), (b), and (c) correspond to the peaks labeled (α), (β), and (γ) in Fig. 3(a), the frequencies and intensities being unchanged.

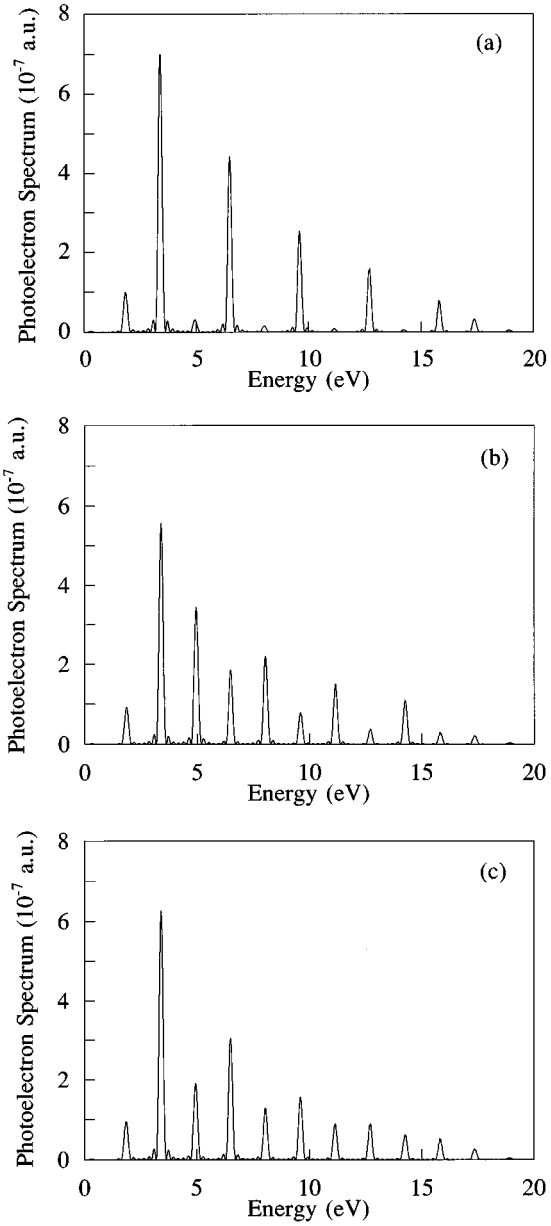

FIG. 5. Effect of the harmonic phases on the $(5+1)$-photoelectron spectrum, with $\omega_L=1.55$ eV, $I_L=8\times10^{11}$ W/cm^2, the five-color spectrum (''Dirac comb'') being shown in Fig. 1(c); (a) $(\phi_{11},\phi_{13},\phi_{15},\phi_{17},\phi_{19})=(0,0,0,0,0)$; (b) $(\phi_{11},\phi_{13},\phi_{15},\phi_{17},\phi_{19})=(0,\pi,0,\pi,0)$; (c) average over all possible phases.

without the laser has the shape already shown in Fig. 1(c). Adding a sixth color, namely, the ir laser radiation leads to photoelectron spectra such as the ones displayed in the Fig. 5. We will exemplify the influence of the harmonic phases in this set of figures, in which the respective intensities of the laser and of the harmonics are kept fixed as follows: $I_1=8\times10^{11}$ W/cm^2; $I_j=3\times10^8$ W/cm^2, where here $j=11,\ldots,19$.

First is shown in Fig. 5(a) the case in which the harmonics have all the same phase, i.e., $\Delta\phi_{i,j}=0$, where the indices i,j label consecutive (odd) harmonics. Quite surprisingly, no new satellite peaks show up between the original ones, at least with the linear scale chosen here. In fact they differ by almost two orders of magnitudes from the ones originating from the absorption of the harmonics. There is, however, a notable exception for the two small peaks located on each side of the original distribution. These peaks are the only ones which cannot be reached via interfering paths, at least in the low-intensity regime considered here. This indicates that if the harmonics had the same phase, the presence of the laser almost would not change the photoelectron spectrum. This results from strong destructive interferences which systematically take place for the peaks associated to laser-assisted transitions which can occurr via distinct quantum

paths. Moreover, the strong cancellations observed are the signature of the fact that the amplitudes associated to the interfering paths have almost equal magnitudes.

We turn now to another extreme case in which the successive harmonics have phases which differ from π, i.e., $\Delta\phi_{i,j}=\pi$, nothing else being changed with respect to the preceding case. The photoelectron spectrum is now dramatically changed, see Fig. 5(b). The satellite peaks become dominant, the original ones being notably depleted. This clearly indicates that constructive interferences have now taken place.

One can also simulate situations in which there would be no phase relations between the harmonics. This would correspond to a case in which the photoelectron spectrum would result from an average over several shots with different harmonic phases. Such a case is simulated in Fig. 5(c), in which one observes that the satellites have magnitudes comparable to the ones of the original peaks. Note that we have verified that a much similar spectrum could be reproduced by assuming that the sequence of phases differences is $(0;\pi/2;0;\pi/2;0)$. Comparing the set of Fig. 5 helps to evidence the strong influence of the harmonic phases and also hints at the possibility to determine them from the analysis of multicolor photoelectron spectra. As the role of quantum interferences is essential in the interpretation of these spectra, we turn now to a brief discussion of a typical (though simplified) case.

V. (2+1) QUANTUM INTERFERENCES

For the sake of discussion, we consider the typical case of two distinct two-photon transition amplitudes leading to the same satellite line, denoted (α), in a $(2+1)$-color photoelectron spectrum, see Fig. 6. One assumes that the fields have frequencies ω_L for the laser and $\omega_p=(2p-1)\omega_L$ and $\omega_{p+1}=(2p+1)\omega_L$, for the harmonics. We assume, in addition, that we are in a low-intensity regime, so that lowest-order perturbation theory can provide sensible estimates of the transition probability. Within this framework, it is enough to assume that the fields have constant amplitudes, as follows:

$$\mathbf{F}_1(t)=iF_1\boldsymbol{\epsilon}_1[\exp(i\omega_L t)-\exp(-i\omega_L t)], \tag{2}$$

which can account for both emission and absorption of laser photons, and

$$\mathbf{F}_j(t)=F_j\boldsymbol{\epsilon}_j\exp[-i(\omega_j t+\phi_j)], \tag{3}$$

where the indices j label the harmonics and $\boldsymbol{\epsilon}_1=\boldsymbol{\epsilon}_j$ are the linear polarization vectors for the laser and its harmonics, which are assumed to be parallel. Here we have assumed positive harmonic frequencies to account for the absorption of one of either harmonic photons. Once the time integration is performed one is left with a transition probability amplitude to reach the state (α) which can be split as follows:

$$T_\alpha=i[F_1F_{p+1}\exp(-i\phi_{p+1})M_{p+1}-F_1F_p\exp(-i\phi_p)M_p], \tag{4}$$

where M_p and M_{p+1} are related to lowest-order (here second-order) perturbative amplitudes associated to the paths labeled (a) and (b) in Fig. 6. Note that there are other

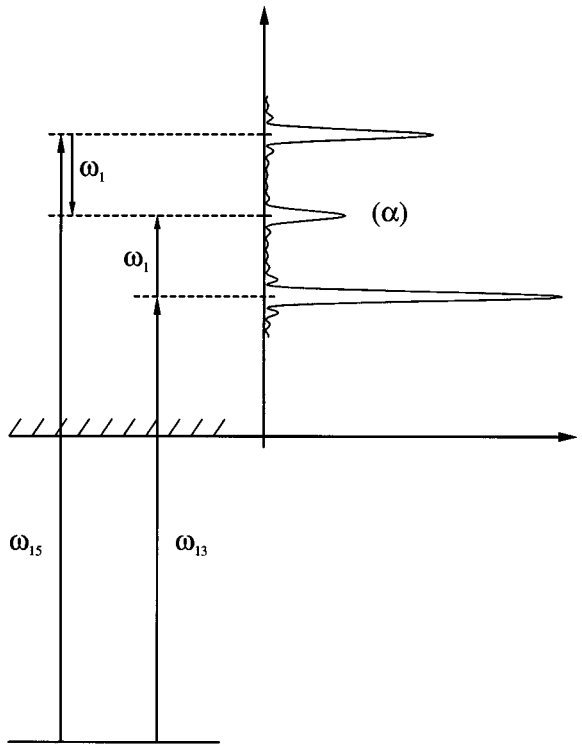

FIG. 6. Energy diagrams schematically representing photoionization processes involving, respectively: (a) absorption of the $(2p+1)$th harmonic followed by the emission of a laser photon; (b) absorption of the $(2p-1)$th harmonic followed by the absorption of a laser photon. A photoelectron spectrum corresponding to the process is shown on the right-hand side.

second-order transition amplitudes leading to the same final state, in which the laser photon is exchanged first. However, these amplitudes do not contribute to the interference effects discussed here and, moreover the contributions of the ones shown here are by far dominant. The quantities M_j read

$$\begin{aligned}M_j&=\langle\psi_\alpha|\boldsymbol{\epsilon}_1\cdot\mathbf{r}G(\epsilon_{1s}+\omega_j)\boldsymbol{\epsilon}_j\cdot\mathbf{r}|\psi_{1s}\rangle\\&=|M_j|\exp i\theta_j,\end{aligned} \tag{5}$$

where θ_j is a characteristic atomic phase and $G(\Omega)$ is the Coulomb Green's function. We note that the final continuum state (α) is, in this lowest-order approximation, a superposition of s and d states. Replacing in Eq. (4) for T_α and assuming that the harmonic fields have equal intensities, i.e., that $F_p=F_{p+1}=F_H$, one has

$$\begin{aligned}T_\alpha=iF_1F_H[&|M_{p+1}|\exp i(\theta_{p+1}-\phi_{p+1})\\&-|M_p|\exp i(\theta_p-\phi_p)].\end{aligned} \tag{6}$$

The transition probability is then proportional to

$$|T_\alpha|^2=F_1^2F_H^2(a-b\cos\Delta\phi-c\sin\Delta\phi), \tag{7}$$

where $\Delta\phi=\phi_{p+1}-\phi_p$, the coefficients a, b, and c being atomic quantities, expressed in terms of the reduced matrix elements M_p and M_{p+1}

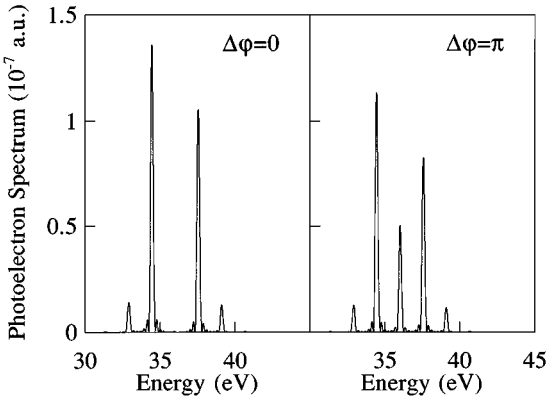

FIG. 7. Effect of the harmonic phases on the $(2+1)$-photoelectron spectrum, for a radiation pulse containing the fundamental frequency of a Ti:sapphire laser, $\omega_L=1.55$ eV, $I_L=10^{11}$ W/cm^2 and its 31st and 33rd harmonic with a fixed intensity $I_H=3\times10^9$ W/cm^2. Left panel: $\Delta\phi=0$; right panel: $\Delta\phi=\pi$.

$$a=|M_p|^2+|M_{p+1}|^2,$$

$$b=2|M_pM_{p+1}|\cos\Delta\theta,$$

$$c=2|M_pM_{p+1}|\sin\Delta\theta, \tag{8}$$

where $\Delta\theta=\theta_{p+1}-\theta_p$.

This simplified form of the transition probability helps to discuss the interference effects observed above. For instance, the magnitudes of the photoelectron peaks depend only on the harmonic phases differences and *do not depend* on the relative phases of the harmonics with respect to the one of the laser. Another point is that strong interferences occur if $|M_p|\approx|M_{p+1}|$. This happens in the limiting case of very high harmonics $(p>>1)$, where we have in addition $\theta_p\approx\theta_{p+1}$, which leads to strongly destructive (constructive) interferences for $\Delta\phi=0$ $(\Delta\phi=\pi)$. We have checked, by using the 31st and 33rd harmonics, that this is indeed the case, see Fig. 7. This confirms, in a somewhat amplified fashion, the tendency already observed in Figs. 5. Indeed, at lower harmonic frequencies, M_p and M_{p+1} are more different, therefore, the interferences are not so strong and the values of $\Delta\phi$ for which they occur are slightly changed.

This shows that multicolor photoionization spectra can be used as a sensitive probe of the phase differences between successive harmonics. As already mentioned, these phases are important intrinsic properties of the harmonics and their determination would present a great interest as it would provide fundamental informations on the basic mechanisms leading to the generation of this source of uv radiations. One notes that, to date, besides the results of semiclassical simulations [7,9,10], and of a quantum computation for a simplified two-level atomic model [17], there exists no systematic study of the dependence of the harmonic phases as a function of the parameters of the laser. Our results indicate that these phases could, at least in principle, be determined through $(2+1)$-color photoionization experiments. Note that this phase determination should be easier for high harmonics.

VI. CONCLUSION

We have presented a computation of $(N+1)$-color atomic photoionization spectra, as they should be observed when using a radiation pulse containing a Dirac comb of N harmonic uv frequencies, together with one of the ir laser which has been used to generate them. If the harmonic frequencies are high enough to lead to single-photon ionization, they are then very efficient to ionize the atomic target, even at the intensities attained by the currently developed harmonic sources. In the presence of the N harmonics the photoelectron spectrum consists of N equidistant lines, separated from each other by $2\omega_L$. When the laser is also present, additional satellite peaks do appear, which are located in between the preceding ones.

A remarkable result of our analysis is that the satellite peaks have notable magnitudes, even at moderate laser intensities. In fact, even in the regime in which lowest-order perturbation theory applies, the satellite peaks can have magnitudes comparable to the ones of those resulting from the absorption of harmonic photons. This results from the strong free-free radiative coupling induced by the ir laser field, between continuum states. One observes further that the satellite peaks grow at the expense of the original ones, the total ionization probability not being significantly changed by the presence of the laser in the short pulses considered here. Even more interesting is the strong sensitivity of the magnitudes of the photoelectron peaks on the phases of the harmonics. We have shown that extreme situations can be envisioned, such as the satellite peaks being almost negligible or, on the contrary, being dominant. The relevant parameter to control the relative heights of the peaks is the phase difference between successive harmonics.

Such strong variations result from the fact that the interfering amplitudes leading to the satellite final states have comparable magnitudes and opposed signs. A second-order calculation in hydrogen confirms the fully numerical approach. We believe that such a result should be transposable to other atomic systems as the physics of the process is dominated by the properties of free-free transition amplitudes in the continuum. We note that contrary to what occurs at higher laser intensities [2,3], the phase difference between the harmonics and the laser has no direct influence on the heights of the photoelectron peaks. As a consequence, we believe that the study of $(N+1)$-color photoionization spectra should allow one to determine the relative phases of successive harmonics, an intrinsic quantity of interest from both the experimental and theoretical points of views. One could argue that in realistic experimental conditions, the harmonic phases may vary during the pulse duration, as the phases are intensity dependent. However, recent simulations indicate that the phase differences between successive harmonics remain constant [7]. Therefore, we think that our method should work, even for an intense femtosecond pulse. Finally, as it is in principle possible to modify the phases of the harmonics, this opens the possibility to achieve the control of the magnitudes of given photoelectron peaks.

Note added in proof. Recently, an experimental work has been reported in which $(4+1)$-color photoelectron spectra are observed [18].

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