Two-frequency multiphoton ionization of hydrogen atoms

C. Leone, S. Bivona, and R. Burlon
Dipartimento di Energetica e Applicazioni di Fisica, viale delle Scienze, Parco D'Orleans, 90128 Palermo, Italy

G. Ferrante
Istituto di Fisica Teorica dell'Università, P.O. Box SO, 98166 S. Agata di Messina, Italy
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The theory of two-frequency multiphoton ionization of hydrogen is reconsidered with particular emphasis on gauge aspects. The analysis is specialized to the case when a relatively weak high-frequency radiation field causes the ionization, while an intense low-frequency radiation field produces structured continuum states for the ionized electrons. Previous results, available in the literature, are recovered as limiting cases of the expressions derived in the present paper. Angular distributions, total cross sections as a function of the low-frequency field intensities, and photoelectron spectra are calculated. Several new features are found and compared with the available information.

I. INTRODUCTION

Ionization of atoms and molecules by very strong lasers is presently attracting considerable attention, thanks to the wealth of experimental observations available, its relevance for important applications, and the challenge it poses to the theory of radiation-matter interaction.[1] Among the cases of physical interest considered experimentally, attention has been paid also to the case when ionization is caused by two laser fields simultaneously.

In a recent experiment, Muller *et al.*[2] have reported on electron spectra resulting from multiphoton ionization of Xe due to two radiation fields of different frequency. A relatively low-intensity uv field causes ionization, while an intense ir field causes a redistribution of all the final states. They have observed sideband structures in the continuum, due to the stimulated absorption and emission of a series of infrared photons. An interesting feature is that the sidebands due to the emission of infrared photons are stronger than those corresponding to the absorption. In the preceding experiment two ionization pathways are simultaneously open: one is that of direct bound-free transition; the other passes through an intermediate resonance. At the moment there is no theory encompassing both mechanisms, thus there is no reliable theoretical interpretation to the observations of Ref. 2. By the way, the two-color experiments of Ref. 2 are largely in the same spirit as the soft x-ray photoeffect in the presence of a low-frequency laser field, which theoretically has been considered by different authors in the past,[3–8] and very recently revisited in Refs. 9 and 10, emphasizing gauge-consistency aspects.

In another experiment by Feldman *et al.*,[11] the resonant multiphoton ionization of sodium atoms by two equally intense lasers was investigated. The first laser had the wavelength in the range between 547 and 580 nm, while the other laser had fixed wavelength $\lambda_2=532$ nm. Along with the processes frequently observed previously (two-photon resonant three-photon ionization) the observed also resonantly enhanced five-photon ionization, in which emission of a photon with wavelength λ_2 takes place. Hence in this experiment too two different ionization pathways are simultaneously open.

Considering the present state of the theory of multiphoton ionization by very strong fields, it is likely that a comprehensive treatment of multicolor multichannel ionization, including the resonant one, will take same time; and that a useful preliminary step may be that of isolating and investigating single, important channels.

In this context, it is the aim of this paper to contribute to the theory of nonresonant two-color multiphoton ionization, with the further restriction that one of the two radiation fields is of low intensity and (relatively) high frequency, while the other is an intense low-frequency field. Hydrogen atoms are chosen as targets. A short account of this work has been presented in Ref. 10. As stated in Ref. 9, also, the problem of the two-color ionization has been addressed and a formal gauge invariant treatment developed.

In our paper we derive the forms of the S matrix in both the E and A gauges and show which chains of simplifications need to be performed to arrive at the ionization transition amplitudes currently encountered in the past literature. No effort is made to arrive at a factored matrix element, in the desire to maintain the gauge consistency as much as possible.

The paper is organized as follows. In Sec. II we give the formal theory of two-color multiphoton ionization of hydrogen atoms with special emphasis on gauge-consistency aspects. It is felt important because hybrid procedures, in which operators and wave functions are not simultaneously transformed, may yield results departing significantly from the correct ones, especially in the strong-field context, such as those of present days experiments. In Sec. III we discuss briefly the wave function used to describe the electron motion in the final state, in the presence of the low-frequency radiation field and the

Coulomb potential. In Sec. IV we work out explicit expressions for transition probabilities and cross sections to base on them a selected set of calculations, reported and commented on Sec. V. Section VI concludes our paper.

We observe finally that below it is tacitly assumed that the S-matrix formalism used here is valid for the problem at hand. The interested reader may find a discussion of this aspect of the problem in Ref. 12.

II. S-MATRIX FORMALISM FOR THE TWO-COLOR IONIZATION AND GAUGE CONSIDERATIONS

In this section we consider the S-matrix theory of two-color ionization within the radiation and electric fields gauges (A and E gauges). As particular cases we recover most of the results previously derived in the literature and establish the contacting points between them. Ionization of hydrogen atoms is taken as the study case.

For the sake of definiteness, below we will refer to the two fields of the problem as being, respectively, of low intensity and high frequency, and of high intensity and low frequency. However, most of the formal derivation is equally well suited for two fields of arbitrary frequency and intensity.

A. E gauge

In the gauge of the electric field the Schrödinger equation of an electron bound by a spherical symmetric potential in the presence of a low-intensity field of high frequency ω_H and of an intense laser field of low frequency ω_L is

$$\left[i\hbar\frac{\partial}{\partial t}-H_0-W^E\right]\Psi_E(\mathbf{r},t)=0\ , \tag{2.1}$$

with

$$W^E=W_H^E+W_L^E\ , \tag{2.2}$$

the interaction of the electron with the two fields

$$W_j^E=e\mathbf{E}_j(t)\cdot\mathbf{r}\quad(j=H,L)\ . \tag{2.3}$$

In (2.3) $\mathbf{E}_H(t)$ and $\mathbf{E}_L(t)$ are, respectively, the high- and low-frequency electric fields, both taken in dipole approximation and spatially homogeneous; e is the absolute value of the electron charge ($e=|e|$); and

$$H_0=\frac{\hat{\mathbf{p}}^2}{2m}+V(r)\ , \tag{2.4}$$

the energy operator of the unperturbed atom. The complete retarded Green function of Eq. (2.1) is

$$\left[i\hbar\frac{\partial}{\partial t}-H_0-W^E\right]G_E^+(\mathbf{r},t;r',t')=i\hbar\delta(\mathbf{r}-\mathbf{r}')\delta(t-t')\ . \tag{2.5}$$

Further, let us write

$$H_0\varphi_\alpha^0=\varepsilon_\alpha^0\varphi_\alpha^0\ , \tag{2.6}$$

$$\left[i\hbar\frac{\partial}{\partial t}-H_0-W_L^E\right]\Phi_E(\mathbf{r},t)=0 \tag{2.7}$$

and

$$\left[i\hbar\frac{\partial}{\partial t}-H_0-W_L^E\right]g_E^+(\mathbf{r},t;\mathbf{r}',t')=i\hbar S(\mathbf{r}-\mathbf{r}')\delta(t-t')\ , \tag{2.8}$$

where $\Phi_E(\mathbf{r},t)$ and $g_E^+(r,t;r',t')$ are the electron wave function and the complete retarded Green function in the presence of the low-frequency intense laser field only. φ_α^0 and ε_α^0 are, obviously, the eigenfunctions and energy eigenvalues of atom unperturbed Hamiltonian (2.4).

Assuming that at the time t' the atom is in an eigenstate of H_0, the wave function for $t\geq t'$ in the presence of two fields is written as [$x\equiv(\mathbf{r},t)$]

$$\Psi_E^+(x)=\int d^3r'G_E^+(x;x')\varphi^0(x')\quad(t\geq t') \tag{2.9}$$

or, expanding G_E^+ in terms of g_E^+,

$$G_E^+(x;x')=g_E^+(x;x')-(i/\hbar)\int d^4x''g_E^+(x;x'')W_H^E(x'')G_E^+(x'';x') \tag{2.10}$$

or, in more detail, as

$$\Psi_E^+(x)=\int d^3\mathbf{r}'g_E^+(x;x')\varphi^0(x')-(i/\hbar)\int d^4x''\int d\mathbf{r}'g_E^+(x;x'')W_H^E(x'')G_E^+(x'';x')\varphi^0(x')$$
$$=\Phi_E(x)-(i/\hbar)\int d^4x'g_E^+(x;x')W_H^E(x')\Psi_E^+(x')\ , \tag{2.11}$$

with

$$\Phi_E(x)=\int d\mathbf{r}'g_E^+(x;x')\varphi^0(x')\ . \tag{2.12}$$

For the S matrix one then has

$$S_{fi}^E=\lim_{\substack{t\to+\infty\\ t'\to-\infty}}\langle\varphi_f^0|\Psi_{E,i}^+\rangle=\langle\varphi_f^0|\Phi_{E,i}\rangle-(i/\hbar)\int_{-\infty}^{+\infty}dt\langle\Phi_{E,f}|W_H^E|\Psi_{E,i}^+\rangle\ , \tag{2.13}$$

with

$$\Phi_{E,i}(x)=\lim_{t'\to-\infty}\int d\mathbf{r}'g_E^+(x;x')\varphi_i^0(x') \tag{2.14}$$

and

$$\Phi_{E,f}(x)=\lim_{t\to+\infty}\int d\mathbf{r}'g_E^+(x;x')\varphi_f^0(x')\ , \tag{2.15}$$

$\Phi_{E,i}(x)$ and $\Phi_{E,f}(x)$ are the exact electron wave functions, in the presence of the low-frequency laser only, which tend to φ_i^0 for $t'\to-\infty$ and to φ_f^0 for $t\to+\infty$, re-

spectively. It is easy to see that Eq. (2.13) with the due identifications is in agreement with Eq. (6) of Ref. 9. An approximate expression of (2.13) is obtained assuming

$$\Phi_{E,i} \approx \Psi^{+}_{E,i} \approx \varphi^{0}_{i} , \tag{2.16}$$

i.e., neglecting the dressing effects of the two fields on the initial-state wave function. This approximation may be used when the interaction energy of the initial atomic state with the two fields is a very small fraction of a rydberg. For instance, we may require that

$$e\langle r\rangle E_j/\Delta\varepsilon \ll 1 \quad (j=H,L) , \tag{2.17}$$

where $\langle r\rangle$ is some average value of the electron radius of the initial state $\langle r\rangle \approx a_0$ in the hydrogen ground state and $\Delta\varepsilon$ the distance of the nearest atomic energy level from the initial one. Inserting (2.16) into (2.13) one obtains the approximate expression

$$S^{E}_{fi} \approx \delta_{fi} - (i/\hbar)\int_{-\infty}^{+\infty} dt \langle \Phi_{E,f} | W^{E}_{H} | \varphi^{0}_{i} \rangle . \tag{2.18}$$

B. A gauge

Paralleling now the derivation in the radiation gauge, we have instead

$$\left[i\hbar\frac{\partial}{\partial t} - H_0 - W^{A} \right] \Psi_A(x) = 0 , \tag{2.19}$$

$$W^{A} = W^{A}_{H} + W^{A}_{L} + W^{A}_{HL} , \tag{2.20}$$

$$W_j = (e/mc)\,\mathbf{A}_j\cdot\hat{\mathbf{p}} + (e^2/2mc^2)A_j^2 \quad (j=H,L) , \tag{2.20a}$$

$$W^{A}_{HL} = (e^2/mc^2)\,\mathbf{A}_H\cdot\mathbf{A}_L , \tag{2.20b}$$

$$\mathbf{A}_j = -c\int dt\,\mathbf{E}_j(t) . \tag{2.20c}$$

The appearance of an interesting term linking the two fields is to be noted also as the form taken by the energy operators,

$$\tilde{H}_0 = U^{-1}H_0U = \frac{1}{2m}\left[\hat{\mathbf{p}} + \frac{e}{c}(\mathbf{A}_H + \mathbf{A}_L)\right]^2 + V(r) , \tag{2.21}$$

with

$$U = U_H U_L \tag{2.22}$$

and

$$U_j = \exp\left[i\frac{e}{\hbar c}\mathbf{A}_j(t)\cdot\mathbf{r} \right] \quad (j=H,L) . \tag{2.23}$$

The unitary operator (2.23) serves to accomplish the transformation from the E gauge to the A gauge, and it is sometimes called "the Goeppert-Mayer operator."

The complete retarded Green function is given by the solution to the equation

$$\left[i\hbar\frac{\partial}{\partial t} - H_0 - W^{A} \right] G^{+}_{A}(x;x') = i\hbar\delta^4(x-x') \tag{2.24}$$

and the unperturbed eigenfunctions of $\tilde{H}_0$ are given by

$$U^{-1}H_0U\tilde{\varphi}^{0}_{\alpha} = \varepsilon_\alpha\tilde{\varphi}^{0}_{\alpha} , \tag{2.25a}$$

with

$$\tilde{\varphi}^{0}_{\alpha} = U^{-1}\varphi^{0}_{\alpha} . \tag{2.25b}$$

Further, one has

$$\left[i\hbar\frac{\partial}{\partial t} - H_0 - W^{A}_{L} \right] \Phi_A(x) = 0 , \tag{2.26}$$

$$\left[i\hbar\frac{\partial}{\partial t} - H_0 - W^{A}_{L} \right] g^{+}_{A}(x;x') = i\hbar\delta^4(x-x') , \tag{2.27}$$

where $\Phi_A(x)$ is the electron wave function in the presence of the low frequency only, written in radiation gauge, and g^{+}_{A} the retarded Green operator giving the time evolution of

$$g^{+}_{A}(x;x') = U^{-1}_{L}(x)g^{+}_{E}(x;x')U_L(x') . \tag{2.28}$$

We remark here that H_0, Eq. (2.4), appearing in Eqs. (2.19)–(2.28), is no longer the energy operator, this property belonging instead to (2.21).

Assuming as before the system to be in the state $\tilde{\varphi}^{0}(x')$ at t', the wave function for $t \geq t'$ is written as

$$\Psi^{+}_{A}(x) = \int d\mathbf{r}'\,G^{+}_{A}(x,x')\tilde{\varphi}^{0}(x') \quad (t \geq t') . \tag{2.29}$$

Expanding now G^{+}_{A} in terms of g^{+}_{A}

$$G^{+}_{A}(x;x') = g^{+}_{A}(x;x') - (i/\hbar)\int d^4x''\,g^{+}_{A}(x;x'')\times[W^{A}_{H}(x'') + W^{A}_{HL}(x'')]\times G^{+}_{A}(x'';x') \tag{2.30}$$

and substituting into (2.29), one has

$$\Psi^{+}_{A}(x) = \int d\mathbf{r}'\,g^{+}_{A}(x;x')\tilde{\varphi}^{0}_{f}(x') - (i/\hbar)\int d^4x''\,g^{+}_{A}(x;x'')[W^{A}_{H}(x'') + W^{A}_{HL}(x'')]\times G^{+}_{A}(x'',x')\tilde{\varphi}^{0}(x') . \tag{2.31}$$

Finally, for the S matrix we have

$$S^{A}_{fi} = \lim_{\substack{t\to+\infty \\ t'\to-\infty}} \langle \tilde{\varphi}^{0}_{f} | \Psi^{+}_{A,i} \rangle = \langle \tilde{\varphi}^{0}_{f} | u_{A,i} \rangle - (i/\hbar)\int_{-\infty}^{+\infty} dt \langle u_{A,f} | W^{A}_{H} + W^{A}_{HL} | \Psi^{+}_{A,i} \rangle , \tag{2.32}$$

with

$$u_{A,i} = \lim_{t'\to-\infty} \int d^3x'\,g^{+}_{A}(x;x')\varphi^{0}_{i}(x') \tag{2.32a}$$

and

$$u_{A,f} = \lim_{t\to+\infty} \int d^3x'\,g(x;x')\varphi^{0}_{f}(x') . \tag{2.32b}$$

An approximate equivalent to that done in Eq. (2.16) is

$$u_{A,i} \approx \Psi^{+}_{A,i} \approx \tilde{\varphi}^{0}_{i} . \tag{2.33}$$

Substituting (2.33) into (2.32) one obtains

$$S_{fi}=\delta_{fi}-(i/\hbar)\int_{-\infty}^{+\infty}dt\langle u_{A,f}|W_H^A+W_{HL}^A|\tilde{\varphi}_i^0\rangle \ . \quad (2.34)$$

(2.34) should be considered the S-matrix element, equivalent in the **A** gauge to the approximate result (2.18).

C. Some limiting cases and contact with previous work

Now we show which manipulations are necessary to obtain from the exact result (2.32) some currently encountered expressions of the S matrix in the A gauge. For low intensity of the high-frequency laser, we approximate to unity the unitary transformation operator (2.23),

$$U_H(x)\approx 1 \ , \quad (2.35)$$

and neglect the A^2 term in the interaction operator (2.20a),

$$W_H^A\approx(e/mc)\,\mathbf{A}_H\cdot\hat{\mathbf{p}} \ . \quad (2.36)$$

In physical terms, the approximation (2.35) amounts to neglecting the oscillating part of the electron momentum, provided by the high-frequency radiation field. According to (2.35) one has the following approximate relations:

$$\tilde{\varphi}_n^0\approx U_L^{-1}\varphi_n^0 \ , \quad (2.37)$$

$$u_{A,i}\approx\Phi_{A,i}=U_L^{-1}\Phi_{E,i} \ , \quad (2.37a)$$

$$u_{A,f}\approx\Phi_{A,f}=U_L^{-1}\Phi_{E,f} \ . \quad (2.37b)$$

Using (2.35)–(2.37) we obtain an approximate expression for the S-matrix element, in the limit of very small intensities of the high-frequency radiation field,

$$S_{fi}\approx\langle\varphi_f^0 U_L|\Phi_{A,i}\rangle - (i/\hbar)\int_{-\infty}^{+\infty}dt\langle\Phi_{A,f}|(e/mc)\,\mathbf{A}_H\cdot\hat{\mathcal{P}}_L|\Psi_{A,i}^+\rangle \ , \quad (2.38)$$

with

$$\hat{\mathcal{P}}_L=\hat{\mathbf{p}}+(e/c)\,\mathbf{A}_L \ , \quad (2.39)$$

the resulting kinetic momentum in the radiation gauge. The S-matrix element (2.38) has two features to be noted: one is represented by the first term, which is not the δ function δ_{fi}; the other one is represented by the interaction formed by the scalar product of the high-frequency field $\mathbf{A}_H(t)$ with the electron kinetic momentum resulting from the presence of a strong low-frequency field. Of course, in (2.39) the field contribution to the kinetic momentum cannot be neglected in general since the low-frequency field may be of arbitrary intensity. The S-matrix element of Ref. 3 may be obtained by the following simplifications: (i) in the first term of Eq. (2.38), the dressing of the initial state of the function is neglected, i.e.,

$$\Phi_{A,i}\approx U_L^{-1}\varphi_i^0 \quad (2.40)$$

and (ii) in the second term the exact wave function Ψ_{Ai}^+ is approximated by the wave function in the presence of the low-frequency laser only, i.e.,

$$\Psi_{A,i}^+\approx\Phi_{A,i} \ . \quad (2.41)$$

By the approximations introduced in Eq. (2.40) and (2.41), the S-matrix element of Eq. (2.38) becomes

$$S_{fi}\approx S_{fi}-(i/\hbar)\int_{-\infty}^{+\infty}dt\langle\Phi_{A,f}|\,\mathbf{A}_H\cdot\hat{\mathcal{P}}_L(e/mc)|\Phi_{A,i}\rangle \ . \quad (2.42)$$

Finally, the S-matrix element, which is the starting point of Refs. 4–8, is obtained by neglecting in Eq. (2.42) the oscillatory term in the kinetic momentum, Eq. (2.39), i.e.,

$$\hat{\mathcal{P}}_L\approx\hat{\mathbf{p}} \ . \quad (2.43)$$

This approximation may be allowed only for weak fields.

Inserting (2.43) into (2.41) one obtains

$$S_{fi}\approx\delta_{fi}-(i/\hbar)\int_{-\infty}^{+\infty}dt\langle\Phi_{A,f}|(e/mc)\,\mathbf{A}_H\cdot\hat{\mathbf{p}}|\Phi_{A,i}\rangle \ . \quad (2.44)$$

It is to be noted that the simplification (2.43) implies small values of the low-frequency field. It in turn means that the unitary transformation operator U_L may be, accordingly, set equal to 1, and $\Phi_{A,i}\approx\varphi_i^0$. One has, thus, a variety of approximate expressions for the S-matrix element in **A** gauge, which have been used in Refs. 3–8. These expressions are expected to be largely equivalent and to have a range of validity restricted to low intensities of the low-frequency field; moreover, to be, except (2.42), also gauge dependent. A measure of the expected gauge dependence may be obtained by means of numerical calculations of the same quantities in the two gauges, considered in this section.

III. APPROXIMATE WAVE FUNCTION OF EJECTED ELECTRONS

In Sec. IV the starting point to calculate the differential and total cross section is Eq. (2.18). To evaluate this matrix element exactly it is necessary to know the wave function $\Phi_{E,f}$ of the electron moving under the joint action of the Coulomb potential and of the low-frequency laser field. Assuming an adiabatic switching on of the laser, asymptotically $\Phi_{E,f}$ must behave as the field-free incoming Coulomb wave $|\mathbf{q}\rangle$,

$$|\mathbf{q}\rangle=N(\mathbf{q})\exp(i\mathbf{q}\cdot\mathbf{r})F(-i\nu,1,-i(qr+\mathbf{q}\cdot\mathbf{r})) \ , \quad (3.1)$$

where

$$\nu=1/qa_0 \ , \quad (3.1a)$$

$$N(q)=\exp(\pi\nu/2)\Gamma(1+i\nu)(2\pi)^{-3/2} \ , \quad (3.1b)$$

$$\mathbf{p}=\hbar\mathbf{q} \ . \quad (3.1c)$$

Unfortunately, no exact solution is known for $\Phi_{E,f}$ when the two fields are simultaneously acting. If the influence of the Coulomb field could be neglected, then $\Phi_{E,f}$ would be exactly known, and would be given by the now familiar nonrelativistic Volkov plane wave

$$\Phi^{v}_{E,F}=\exp\left[-\frac{i}{2m\hbar}\int^{t}d\tau\left[\mathbf{q}+\frac{e}{mc}\mathbf{A}(\tau)\right]^2\right]\times\exp[i(e/mc)\mathbf{A}(t)\cdot\mathbf{r}]\exp(i\mathbf{q}\cdot\mathbf{r})\ . \tag{3.2}$$

As an approximate wave function we use then a Volkov-Coulomb wave function

$$\Phi^{c}_{E,f}=\exp\left[-\frac{i}{2m\hbar}\int^{t}d\tau\left[\mathbf{q}+\frac{e}{mc}\mathbf{A}(\tau)\right]^2\right]\times\exp[i(e/mc)\mathbf{A}(t)\cdot\mathbf{r}]|q\rangle\ , \tag{3.3}$$

i.e., a Coulomb function modulated in time by the laser in the same way as the Volkov plane waves.

For a laser linearly polarized, spatially and temporally homogeneous, taken in the dipole approximation, with vector potential given by

$$\mathbf{A}_L(t)=\mathbf{A}_{0L}\cos(\omega_L t+\varphi_L)\ , \tag{3.4}$$

the Volkov-Coulomb function may be written as

$$\Phi_{E_{if}}\approx\exp[i\cos(\omega_L t+\varphi_L)(e/mc)\mathbf{A}_{0L}\cdot\mathbf{r}]\times\exp\{-i[\alpha_q\sin(\omega_L t+\varphi_L)+\rho\sin(2\omega_L t+2\varphi_L)]\}\times\exp\left[-\frac{i}{\hbar}(\varepsilon^0_q+\Delta)t\right]\left|q\right\rangle\ , \tag{3.5}$$

with

$$\alpha_q=(e/mc\omega_L)\mathbf{A}_{0L}\cdot\mathbf{q}\ , \tag{3.6a}$$

$$\Delta=(e^2A^2_{0L}/4mc^2)\ , \tag{3.6b}$$

$$\rho=\Delta/2\hbar\omega_L\ . \tag{3.6c}$$

Equation (3.3) has been derived and used also previously by several authors in different contexts and under different restrictions.[13–16] It is generally considered that the ansatz (3.3) gives a satisfactory approximation to the exact solution insofar as high-intensity fields are concerned, and the discrete part of atomic spectrum does not play a role. It restricts the validity of (3.3) to one-step, far-nonresonant ionizing events.

IV. CROSS SECTION

In Sec. II we have developed an S-matrix formalism of two-color ionization in E gauge and obtained an approximate formula (2.18) which we will use below. Though approximate, formula (2.18) is physically clear: the perturbation W^E connects two states φ^0_i and $\Phi_{E,f}$ which are both eigenstates of the electron energy operator, and thus correspond to observables. Therefore the results which will be obtained below should be not affected by the limitation of previous works, concerning the gauge consistency. We give now the cross section of multiphoton ionization of hydrogen atoms in their ground state in the presence of two radiation fields.

The S-matrix element (2.18) is used, taking as final state the function $\Phi_{E,f}$, Eq. (3.5). It gives

$$S_{fi}=\sum_s e^{-is\varphi_L}\frac{(-i)^s}{2}\int_{-\pi}^{+\pi}d\alpha\, f_s(\alpha)K(\mathbf{q},E^L_g\hat{\mathbf{z}}\sin\alpha)[\delta(\varepsilon^0_q+I_0+\Delta-s\hbar\omega_L+\hbar\omega_H)-\delta(\varepsilon^0_q+I_0+\Delta-s\hbar\omega_L-\hbar\omega_L)]\ , \tag{4.1}$$

where

$$f_s(\alpha)=\exp\{is\alpha-i\alpha_q\cos\alpha-i\rho\sin2\alpha)\ , \tag{4.2}$$

$$E^L_g=(e/\hbar c)A_{0L}\ , \tag{4.2a}$$

$$K(\mathbf{q},E^L_g\hat{\mathbf{z}}\sin\alpha)=\langle\mathbf{q}|\exp(-iE^L_g\hat{\mathbf{z}}\cdot\mathbf{r}\sin\alpha)eE_H\hat{\mathbf{z}}\cdot\mathbf{r}|0\rangle\ , \tag{4.2b}$$

$$\varepsilon^0_q=\hbar^2q^2/2m\ . \tag{4.2c}$$

In (4.1)–(4.2) I_0 is the ionization energy, $|0\rangle$ the spatial wave function of the $1s$ state, and $\hat{\mathbf{z}}$ the unitary vector along the direction of $\mathbf{A}_{0L}$.

The integral (4.2b) after evaluation with the method of Ref. 17 becomes

$$K(\mathbf{q},E^L_g\hat{\mathbf{z}}\sin\alpha)=eE_HN^*(q)(\pi a^3_0)^{-1/2}(16\pi a^4_0)(T/S)^{i\nu}\nu(i\nu-1)a_0T^{-3}\times\{E^L_g\sin\alpha[(T/S)(2-iqa_0)+(T^2/S^2)(1+i\nu)(1-iqa_0)(1-i\nu)^{-1}-(\nu+2i)\nu^{-1}]+q_z[(T/S)(1-iqa_0-(\nu+2i)\nu^{-1}]\}\ , \tag{4.3}$$

with

$$T=1+a^2_0(\mathbf{q}+E^L_g\hat{\mathbf{z}}\sin\alpha)^2\ , \tag{4.4}$$

$$S=a^2_0(E^L_g)^2\sin^2\alpha+(1-iqa_0)^2\ . \tag{4.5}$$

In Eq. (4.1) the first term corresponds to the process in which s photons of the low-frequency field are absorbed and one photon or high frequency is emitted; instead, the second term corresponds to the process in which the high-frequency field causes the ionization, while the low-frequency one causes a redistribution of all the final states; while in the first case only absorption of low-frequency photons is energetically possible, in the second case both absorption and emission channels are open.

Thus the first term describes (nonresonant) processes similar to those observed by Feldman *et al.*,[11] while the second one describes processes of the type observed by Muller *et al.*[2]

It is worthwhile to note that the presence of the Goeppert-Mayer factor in Eq. (4.2b) is responsible for the coupling between the initial atomic states and all the partial waves with definite orbital quantum number 1 entering the expansions of the final Coulomb function.

Below we will be interested in the second process only. Proceeding in the usual way, the following multichannel multiphoton differential cross section is arrived at:

$$\frac{d\sigma(E_g^L)}{d\Omega}=\sum_{s=s_1}^{\infty}\frac{d\sigma(s,E_g^L)}{d\Omega}\ , \tag{4.6}$$

with

$$\frac{d\sigma(s,E_g^L)}{d\Omega}=2^6\omega_H a_0^3(\pi c)^{-1}(1-e^{-2\pi\nu_s})^{-1}|T_s(q,E_g^L)|^2\ , \tag{4.7}$$

$$T_s(\mathbf{q}_s,E_g^L)=\int_{-\pi}^{+\pi}d\alpha\, f_s(\alpha)B(\mathbf{q}_s,E_g^L\hat{\mathbf{z}}\sin\alpha)\ , \tag{4.8}$$

$$B(\mathbf{q}_s,E_g^L\hat{\mathbf{z}}\sin\alpha)=(\pi a_0^3)^{1/2}[16\pi a_0^4 eE_H N^*(q_s)]^{-1}\times K(\mathbf{q}_s,E_g^L\hat{\mathbf{z}}\sin\alpha)\ , \tag{4.9}$$

$$\hbar^2 q_s^2/2m=s\hbar\omega_L+\hbar\omega_H-I_0-\Delta\ . \tag{4.10}$$

s_i is the smallest negative integer for which (4.10) is still positive, and indicates the channel with the highest number of low-frequency photons emitted. The complete, angle-resolved cross section is given by

$$\sigma(E_g^L)=\sum_{s=s_1}^{\infty}\sigma_s(E_g^L)\ , \tag{4.11}$$

with

$$\sigma_s(E_g^L)=\int\frac{d\sigma(s,E_g^L)}{d\Omega}d\Omega\ . \tag{4.12}$$

V. CALCULATIONS AND COMMENTS

The treatment given above is now applied to the calculations of the differential cross section (DCS), Eq. (4.7), and total cross section (TCS), Eq. (4.12), for different numbers s of the exchanged low-frequency photons. The geometry is the parallel one, when the two fields are taken in the same direction. In Fig. 1 we show the DCS, Eq. (4.7), for $s=\pm1$, ±2, ±4, ±7, ±9, and ±10, taking $\hbar\omega_H=50$ eV, $\hbar\omega_L=1.17$ eV, and the laser intensity $I_L=5\times10^{12}$ W/cm^2.

For small $|s|$ the DCS of emission and absorption exhibit largely a similar behavior, with values of the same order of magnitude except at angles near 0° and 90°. When $|s|$ increases the patterns of two DCS become different and the DCS corresponding to the absorption tend to prevail over the emission ones. Besides, the DCS tend to shift towards small ejection angles.

In Fig. 2 we report the DCS as in Fig. 1, when $E_g^L=0$, i.e., when the gauge transformation operator is set equal

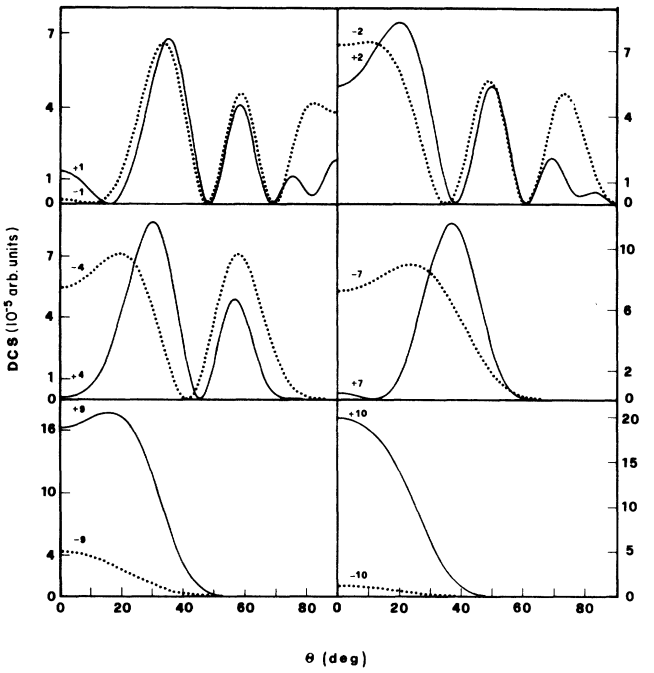

FIG. 1. Differential cross sections [DCS, Eq. (4.7)] for different numbers of exchanged low-frequency photons; θ is the angle between the momentum of the photoelectron and the electric fields. The energy of the high-frequency photon is $\hbar\omega_H=50$ eV. The intensity and the photon energy of the low-frequency laser are, respectively, $I_L=5\times10^{12}$ W/cm^2 and $\hbar\omega_L=1.17$ eV. The number of exchanged photons is indicated on the curve.

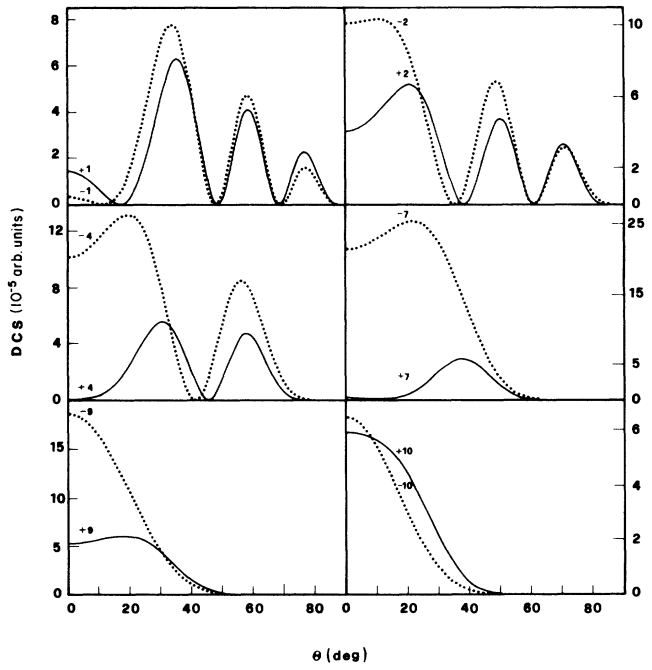

FIG. 2. Same as in Fig. 1 with the gauge-transformation operator $U_L=1$.

to 1. We observe that the emission DCS tends to prevail on the absorption DCS for any $|s|$. However, as in Fig. 1, for small $|s|$ the emission and absorption DCS exhibit a similar behavior, while again the patterns are different when $|s|$ increases.

In Fig. 3 we show the TCS, Eq. (4.12), versus the laser intensity I_L for $s=\pm1$, ±2, ±4, and ±7, $\hbar\omega_H=50$ eV, and $\hbar\omega_L=1.17$ eV. For any s the TCS exhibit a linear behavior for small intensities. The intensity domain of linear growth is then larger than higher the photon multiplicity s. The absorption cross sections are slightly larger than emission cross sections. After the region of linearity, the TCS exhibit a decreasing oscillatory behavior with an inversion between emission and absorption; in this oscillatory region generally emission TCS are larger than the absorption TCS.

This oscillatory behavior appears to yield, on the average, larger cross sections for emission rather than for absorption. Eventually each curve disappears abruptly, as the channel closes due to the ionization potential shift. The answer why both emission and absorption cross sections decreases after the linear growth is contained in Fig. 4.

In Fig. 4 we report the same quantities as in Fig. 3 grouping separately the TCS of absorption and emission. The slope of the curves for different s at small intensity increases with $|s|$ while at larger intensities the TCS tend to take values of the same order of magnitude. Figure 4 shows that by increasing the laser intensity an increasing number of channels opens, which eventually have comparable probabilities; it is responsible for the kind of saturation shown by Figs. 3 and 4.

In Figs. 5 and 6 we show the electron spectra obtained for different values of the high-frequency photon energy ($\hbar\omega_H=50$ and 100 eV) and several low-frequency laser intensities I_L. The results are normalized to the value of $s=0$. Figures 5 and 6 show that, for small values of the low-frequency laser intensity I_L, the sidebands corresponding to the absorption are stronger than those of emission. In practice, this occurs when the coupling between the ionized electron and the low-frequency laser α_q, Eq. (3.6a), is very small for all s. When the laser intensity I_L increases the sidebands corresponding to the emissions are larger than those of absorption, at least for small values of $|s|$. Increasing the value of $|s|$, the sidebands corresponding to the absorptions are again stronger than those of the emissions. An interpretation to these results is proposed below.

In a very simple picture (arising from the previous treatments[4-8]) the stimulated ionization process may be

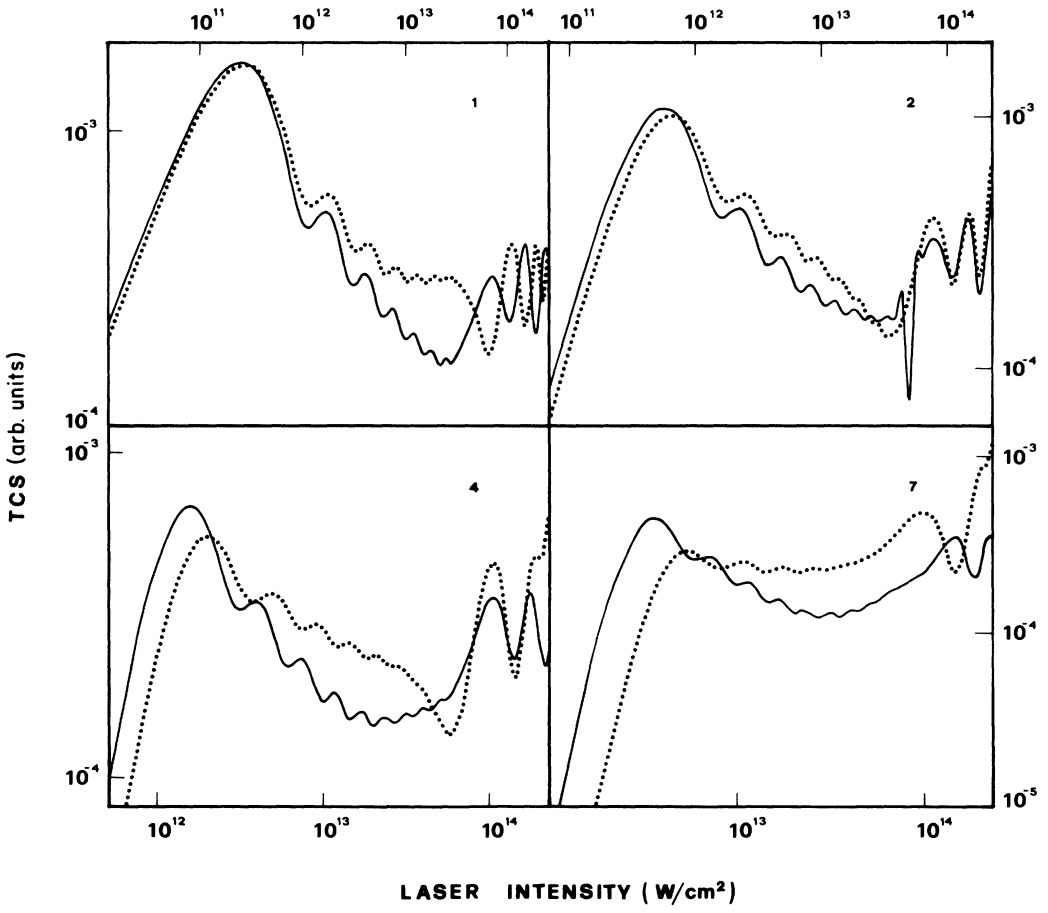

FIG. 3. Total cross sections (TCS) vs the low-frequency laser intensity I_L for different numbers of exchanged low-frequency photons. Emission, dotted curve; absorption, solid curve. The frequencies of the two fields are the same as in Fig. 1.

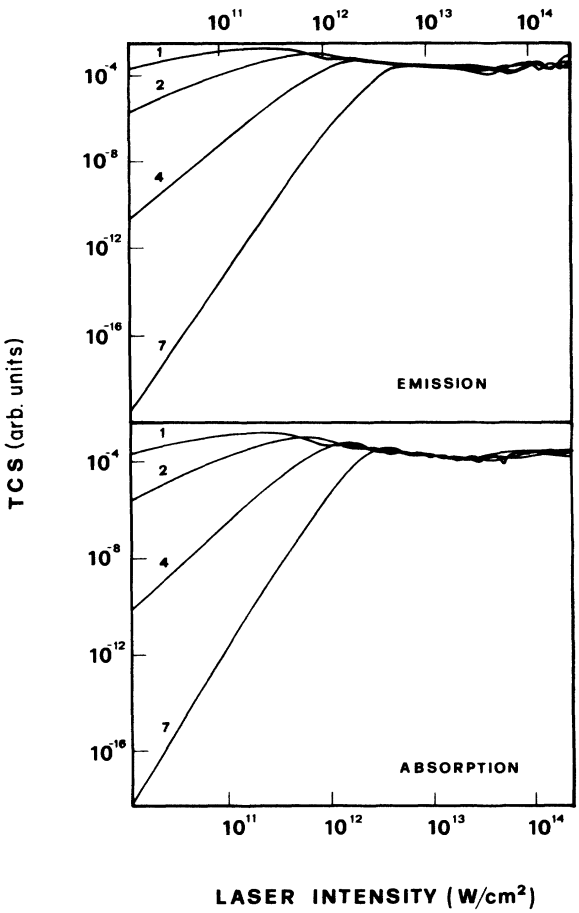

FIG. 4. TCS of emission and absorption. Same caption as in Fig. 1.

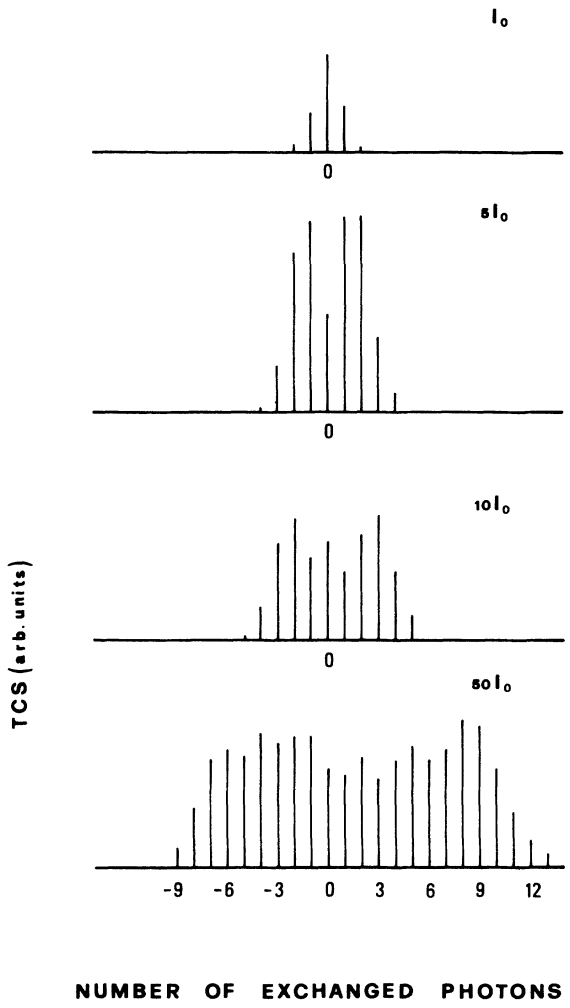

FIG. 5. TCS vs the number of the exchanged low-frequency photons for different values of the low-frequency laser intensity. $I_0 = 10^{11}$ W/cm^2. The energy of the high-frequency photons is $\hbar\omega_H = 50$ eV.

viewed in the following terms. The ionization dynamics have essentially the same probabilities as for the case of absence of a strong stimulating field, while the probabilities of additionally emitting or absorbing s low-frequency photons are governed by squared Bessel functions J_s^2 of order s and argument containing the coupling parameter α_q (a strong purely coherent field is understood). Thus, for instance, the probability of the electron being ionized with additional absorption of s strong-field photons, is just given by the product of the probabilities of the two separate events

$$W_S^+ = w^+(s\hbar\omega_L) W_i(\varepsilon_0 + s\hbar\omega_L) , \tag{5.1}$$

where

$$w^+(s\hbar\omega_L) \sim J_s^2[\alpha_q(s)]$$

is the probability of absorbing s low-frequency photons with $W_i(\varepsilon_0 + s\hbar\omega_L)$ is the laser-free ionization probability evaluated at the final energy $\varepsilon_0 + s\hbar\omega_L$ (ε_0 is the final electron energy in the absence of the strong low-frequency field).

Within this picture, for open channels, starting from few eV above threshold as a rule $W_i(\varepsilon_0 - s\hbar\omega_L)$ is sufficiently larger than $W_i(\varepsilon_0 + s\hbar\omega_L)$ to yield the prevalence of emission over absorption, irrespective of the values of $w^+(s\hbar\omega_L)$ and $w^-(s\hbar\omega_L)$. However, this picture misses an important physical aspect (which manifests itself as gauge dependence); namely, it misses that in the final state the electrons have an oscillatory component of the energy, imparted by the strong field. It produces in some effective way the averaging of $W_i(\varepsilon_s)$ around $W_i(\varepsilon_0)$. To some extent, it is reflected in the structure of Eqs. (4.1), (4.2), (4.7), and (4.8).

For the sake of simplicity, we adopt here a crude approximation, which, however, is expected to be in accordance with the preceding equations. Namely, we take that the same $W_i(\varepsilon_0)$ will appear in both the absorption and emission channels,

$$W_s^+ \approx w^+(s\hbar\omega_L) W_i(\varepsilon_0) , \tag{5.2a}$$

$$W_s^- \approx w^-(s\hbar\omega_L) W_i(\varepsilon_0) . \tag{5.2b}$$

Now emissions and absorptions are controlled by w^- and

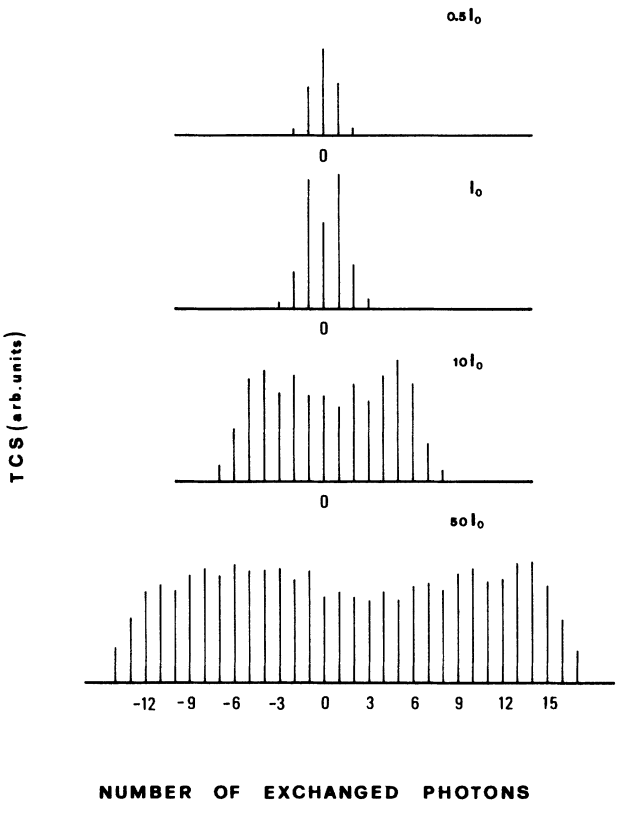

FIG. 6. Same as in Fig. 5 with $\hbar\omega_H = 100$ eV.

w^+ and a number of predictions may be easily anticipated which are fully supported by the numerical calculations of the exact formulas. In particular, the following.

(i) For weak assisting fields, the absorption and emission probabilities are proportional to the coupling parameter

$$w^{\pm}(s\hbar\omega_L) \sim [\alpha_q(\pm s)]^{2|s|} \sim c_0 E_{0L}^{2|s|} [2m(\hbar\omega_H \pm s\hbar\omega_L - I_0 - \Delta)]^{|s|}, \quad \alpha_q(s) < 1 \qquad (5.3)$$

with c_0 a constant, grouping several parameters. It is evident that in this domain $w^+(s\hbar\omega_L)$ is larger than $w^-(s\hbar\omega_L)$ (see Fig. 3).

(ii) Increasing the intensity of the assisting field, $w^{\pm}(s\hbar\omega_L)$ increases linearly up to some maximum values and then enters a nonincreasing nonlinear regime. As $w^+(s\hbar\omega_L)$ is larger than $w^-(s\hbar\omega_L)$ in the linear regime, it is obvious that $w^+(s\hbar\omega_L)$ reaches first its maximum and first enters the nonincreasing nonlinear regime. Accordingly, at higher intensities there should be an inversion domain in which emission processes should have higher probabilities than absorptions (see Fig. 3).

(iii) For sufficiently high s, irrespective of the intensity domain, the coupling parameter $\alpha_q(s)$ becomes very small for emission (negative s), and the absorption processes have larger probabilities than emission ones. It must be added that the absorption channels are open from 1 to ∞, while the emission ones only from -1 to $-s$ [see Eq. (4.6)].

VI. CONCLUDING REMARKS

We have revisited the theory of two-frequency multiphoton ionization of hydrogen, with particular emphasis to gauge aspects. The theory has been specialized to the case when a relatively weak high-frequency radiation field cases the ionization and a strong low-frequency field produces structured continuum states for the ejected electrons. We have also shown how to recover from the expression derived in the present paper, several previous results available in the literature.

Selected sets of calculations have been performed on angular distributions, on total cross sections as functions of the low-frequency field intensities, and on the photoelectron spectra. Several new features have been found, as compared with the available information, and detailed comments have been devoted to them.

As is typical of this kind of processes, the too many physical parameters of the problem make rather difficult a complete understanding of all its interesting aspects. Besides, we have left out of our consideration the more general case of two comparable fields, the influence of the radiation properties and many other aspects. On some of them, we hope to report in the future.

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