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Use of the Coulomb Green's function in atomic calculations*

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Analytical and numerical results are presented which are related to the following higher-order atomic processes: retardation effects in two-photon bound-bound transitions, evaluation of the Bethe logarithm in excited states, and multiphoton ionization. These calculations are carried out within the framework of the conventional perturbation theory by using different representations of the Coulomb Green's function, each representation offering its own particular advantages.

I. INTRODUCTION

Recent experimental refinements observed in the field of atomic physics have given rise to a great interest in the calculation of higher-order terms of the perturbation expansion. This general trend is particularly important in calculations related to the study of multiphoton processes, as a matter of fact most of the features of these processes may be interpreted within the framework of time-dependent perturbation theory.

The main difficulty encountered in computation of amplitudes of order $N \geqslant 2$, comes from the presence of infinite sums extended to the whole atomic spectrum. Such sums are usually evaluated by using some adequate approximation techniques, but it is well known that if the atomic system under consideration is a hydrogenic one, the calculation can be carried out "exactly," i.e., without approximation.

To date two methods have been proposed in order to perform such exact calculations. Chronologically, the first one was given by Dalgarno and Lewis[1] in 1955. These authors reduced the calculation of a second-order amplitude to the numerical integration of an inhomogeneous differential equation. The method has been successfully applied to various two-photon problems such as Lamb-shift calculations by Schwartz and Tiemann,[2] elastic scattering of photons by Mittleman and Wolf,[3] and two-photon ionization by Zernik.[4] Later on Gontier and Trahin[5] succeeded in generalizing it to the cases where the number of photons involved in the process is greater than 2. Now this method has proven its reliability, but we must note that it is not so easy to use since it involves elaborate numerical methods, especially for higher-order calculations ($N > 2$).

The other method proposed, based on the use of compact representations of the Coulomb Green's function (CGF), does not suffer such limitation since it permits us to obtain, in theory, analytical expressions of the desired amplitudes. The advantages of this method have been recognized by Vetchinkin and Khristenko[53] and by Gavrila[6] in 1967, who used it in the calculation of second-order amplitudes in hydrogen atoms. Later on, as we shall explain, a number of authors have improved the technique and applied it to the study of many higher-order processes.

We used this latter method, and we shall present here our results concerning various multiphoton processes. Moreover, since in the course of this work we were induced to use different representations of the CGF, we have been able to achieve two distinct purposes. On the one hand, we got some interesting results on the retardation effect in two-photon bound-bound transitions, Lamb-shift calculations, and multiphoton ionization. On the other hand, we compared the respective advantages of the various representations of the CGF.

In Sec. II we recall some general properties of Green's functions and give a brief survey of the most useful representations of the CGF. Section III is dedicated to the study of the retardation effect in two-photon bound-bound transitions. An exact analytical evaluation of the Bethe sums for Lamb-shift calculations in higher excited states is given in Sec. IV, and Sec. V deals with the calculation of multiphoton ionization transition rates.

II. REPRESENTATIONS OF THE COULOMB GREEN'S FUNCTION

A. Generalities

The Green's function or resolvant for an operator H is the operator

$$G(E) = (E - H)^{-1}. \qquad (1)$$

Equation (1) is equivalent to the relation

$$(E - H)\,G(E) = I. \qquad (1')$$

On multiplying Eq. (1) by the projector $P_\nu = |\nu\rangle\langle\nu|$

associated with an eigenvalue E_ν of the operator H, which satisfies the equation

$$HP_\nu = E_\nu P_\nu \,, \tag{2}$$

one gets

$$G(E)P_\nu = (E-H)^{-1}P_\nu = (E-E_\nu)^{-1}P_\nu \,, \tag{3}$$

and by using the closure relation we have

$$G(E) = \sum_\nu \frac{|\nu\rangle\langle\nu|}{E-E_\nu} \,, \tag{4}$$

where the sum is extended to the whole spectrum of the operator H.

The representation of $G(E)$ in the position space $G(\vec{r},\vec{r}\,';E) = \langle\vec{r}\,|\,G(E)|\,\vec{r}\,'\rangle$ can be defined either from Eq. (4):

$$G(\vec{r},\vec{r}\,';E) = \sum_\nu \frac{\langle\vec{r}\,|\,\nu\rangle\langle\nu\,|\,\vec{r}\,'\rangle}{E-E_\nu} \tag{5}$$

or from Eq. (1′), as the solution of an inhomogenous differential equation:

$$(E-H)\,G(\vec{r},\vec{r}\,';E) = \delta(\vec{r}-\vec{r}\,') \,. \tag{6}$$

Note that the momentum-space representation of the Green's function is the solution of an integral equation, the form of which may be obtained from Eq. (6) by taking its Fourier transform.

The Green's function is introduced in the perturbation expansion in the form (4), but its properties are generally derived from the differential equation (6). The compact representations of the CGF used in our work have been obtained in this way—we recall it briefly in the following paragraphs.

B. Schwinger's representation (Ref. 7)

Schwinger[7] has obtained a representation of the CGF in momentum space. In his derivation, which we summarize below, he used the $O(4)$ symmetry properties of the hydrogen atom revealed by the Fock transformation.[8] The Schrödinger equation for a nonrelativistic hydrogenic atom in momentum space reads (throughout this paper we will use atomic units):

$$\tfrac{1}{2}(p^2+x^2)\,\Phi(\vec{p}) - \frac{\alpha}{2\pi^2}\int d^3\vec{p}\,'\,\Phi(\vec{p}\,')\,|\,\vec{p}-\vec{p}\,'|^{-2} = 0 \,, \tag{7}$$

where $x = (-2E)^{1/2}$ $(E<0)$, and $\alpha = Z$ corresponds to the coupling of the Coulomb potential.

Under the Fock transformation, corresponding to the variable change

$$\begin{gathered} \xi = (\xi_0, \vec{\xi}\,), \quad \vec{\xi} = \frac{2x\vec{p}}{p^2+x^2} \,, \\ \xi_0 = \frac{p^2-x^2}{p^2+x^2} \,, \quad \xi_0^2 + \vec{\xi}^{\,2} = 1 \,, \end{gathered} \tag{8}$$

Eq. (7) becomes

$$Y(\xi) = \frac{\nu}{2\pi^2}\int_{S^3} d\xi'\,Y(\xi')\,|\,\xi-\xi'|^2 \,, \tag{9}$$

where

$$\nu = \alpha x^{-1}, \quad Y(\xi) = \tfrac{1}{4}(x^2+p^2)^{-2}x^{-5/2}\,\Phi(\vec{p}) \,,$$

and S^3 is the unit hypersphere in $\mathfrak{R}^4$.

The corresponding momentum-space CGF is the solution of the equation

$$\tfrac{1}{2}(p^2+x^2)\,G(\vec{p},\vec{p}\,';E) - \frac{\alpha}{2\pi^2}\int d^3\vec{p}\,''\,G(\vec{p}\,'',\vec{p}\,';E)\,|\,\vec{p}\,''-\vec{p}\,|^{-2} = -\delta(\vec{p}-\vec{p}\,') \,. \tag{10}$$

In the new variables Eq. (10) reads

$$\Gamma(\xi,\xi') = \delta(\xi-\xi') + \frac{\nu}{2\pi^2}\int_{S^3} d\xi''\,\Gamma(\xi'',\xi')\,|\,\xi-\xi''|^{-2} \,, \tag{11}$$

with

$$\Gamma(\xi,\xi') = -\tfrac{1}{2}(2x)^{-3}[(x^2+p^2)(x^2+p'^2)]^2\,G(\vec{p},\vec{p}\,';E) \,. \tag{12}$$

Schwinger has shown that Eq. (11) is more easily solved than the original one in ordinary momentum space, and he got the result

$$\Gamma(\xi,\xi') = \sum_{nlm}\left(1-\frac{\nu}{n}\right)^{-1} Y_{nlm}(\xi)\,Y^*_{nlm}(\xi') \,, \tag{13}$$

where the functions $Y_{nlm}(\xi)$ are surface hyperspherical harmonics.

One of the most remarkable properties of this expression is that the infinite sum is extended only to a discrete spectrum. This feature is closely related to the character of the Fock transformation as noted by Decoster.[9] Moreover, this representation may be considered as a four-dimensional counterpart[10,11] of the so-called Sturmian representation introduced later on (see Sec. ID).

From Eq. (13) one gets: (i) integral representations of $\Gamma(\xi,\xi')$ by using the relations

$$\left(1-\frac{\nu}{n}\right)^{-1} = 1 + \frac{\nu}{n} + \frac{\nu^2}{n(n-\nu)} \,,$$

$$(n-\nu)^{-1} = \int_0^1 dz\,z^{-\nu}z^{n-1}, \quad \nu<1 \,;$$

(ii) corresponding expressions for $G(\vec{p},\vec{p}\,';E)$.

Such a momentum-space integral representation was used first by Gavrila,[6] who studied the elastic scattering of photons on a hydrogenic atom in the ground state. Later on he extended these calculations in order to take into account the retardation effect in elastic[12] and inelastic[13] (Compton) scattering on a K-shell electron. More recently, a similar technique has been used by Douglas[14] who applied it to the calculation of hyperfine-structure corrections.

We used a representation of this type, but in order to take full advantage of the $O(4)$ symmetry of the H atom, we performed the complete calculation in the four-dimensional space introduced by Fock's transformation (see Sec. III). A similar technique has been used by Lieber[15] in a Lamb-shift calculation.

C. Hostler's integral representation (Ref. 16)

Hostler[16] has given an integral representation of the CGF in position space. He used an approach, different from Schwinger's, based on a known theorem which states that a one-dimensional Green's function may be expressed as a product of two independent solutions of the corresponding homogeneous differential equation.[17] Thus if we consider the partial-wave component of the CGF, which is the solution of the inhomogeneous differential equation

$$(H_l - E)\,G_l(r,r';E) = -\delta(r-r')\,, \tag{14}$$

where

$$H_l = -\left(\frac{1}{2}\frac{d^2}{dr^2} + \frac{1}{r}\frac{d}{dr} + \frac{1}{r} - \frac{l(l+1)}{r^2}\right), \tag{15}$$

and we apply the above-mentioned theorem, one gets

$$G_l(r,r';E) = -\frac{\Gamma(l+1-x^{-1})}{xrr'}M_{1/x,\,l+1/2}(2xr_<)\times W_{1/x,\,l+1/2}(2xr_>)\,, \tag{16}$$

where $r_> \equiv \sup(r,r')$ and $r_< \equiv \inf(r,r')$ and the functions M and W are the known Whittaker's functions,[18] solutions of the radial Schrödinger equation

$$(H_l - E)R_{n,l}(r) = 0\,. \tag{17}$$

This form of the CGF was known for a long time, but is not very useful for practical computations since it does not factor the dependences on r and r' of $G_l(r,r';E)$, which obliges one to resort to numerical quadrature techniques. Moreover, owing to the oscillatory behavior of the functions W for large r, the numerical treatment of these functions is very cumbersome. As a consequence, this representation of the CGF has not been used very often. We can mention, however, the recent work by Laplanche *et al.*[19] on multiphoton ionization.

In 1964 Hostler[16] made a further step by using an integral representation[18] for the product of two Whittaker functions [Eq. (16)]. He obtained in this way the following formula:

$$G_l(r,r';E) = -2(rr')^{-1/2}\times\int_0^\infty dt\, e^{-(r+r')x\cosh t}\times(\coth\tfrac{1}{2}t)^{2/x}\, I_{2l+1}(2x(rr')^{1/2}\sinh t)\,, \tag{18}$$

where $I_\lambda(z)$ is a Bessel function of imaginary argument.

This partial-wave representation of the CGF has been found to be very useful for calculations made within the dipole approximation, and a number of authors have used it for calculations related to various two-photon processes. As far as we are concerned, we used it in the evaluation of the Bethe logarithm in excited states (see Sec. IV).

Before going further, we can point out that Hostler[16] has also given a representation of the complete CGF, $G(\vec{r},\vec{r}\,';E)$, by summing the partial-wave expansion

$$G(\vec{r},\vec{r}\,';E) = \frac{1}{4\pi}\sum_{l=0}^{\infty}(2l+1)P_l(\cos\theta)\,G_l(r,r';E)\,,\qquad \theta = (\vec{r},\vec{r}\,')\,, \tag{19}$$

after replacing G_l by its integral representation [Eq. (18)]. This last part of Hostler's results has been used in calculations taking into account the retardation effect (see for instance, the papers by Klarsfeld[20]).

Eventually we can mention that, as shown by Hostler[21] by taking the Fourier transform of $G(\vec{r},\vec{r}\,';E)$ one gets the corresponding representation in momentum space.

D. Sturmian representation (Ref. 22)

As we already said, the above-mentioned representations are very useful for calculations of second-order amplitudes related to various radiative processes. Nevertheless, they have not been used in higher-order calculations, owing to the presence of intricate multiple integrals.

In such cases it may be convenient to use a different type of representation of the CGF, which was first proposed by Hostler in 1970.[22] We will refer to this representation as "Sturmian" since it is given as an expansion over the complete set of the so-called Coulomb "Sturmian" functions introduced by Rotenberg.[23] Before rederiving explicitly this expression of the CGF, it seems of interest to recall some of the general properties of these functions.

Let the radial Schrödinger equation for a given angular momentum l be

$$[H_l' + \alpha V(r)]R_l(r) = ER_l(r)\,, \tag{20}$$

where

$$H_l' = -\left(\frac{1}{2}\frac{d^2}{dr^2} + \frac{1}{r}\frac{d}{dr} - \frac{l(l+1)}{r^2}\right), \tag{21}$$

and α is the coupling constant.

We define the corresponding conjugate or "Sturmian" equation as

$$[H_l' - E]S_{l,E}(r) = -\alpha_{l,E}V(r)S_{l,E}(r)\,, \tag{22}$$

where the Sturmian functions $S_{l,E}(r)$ verify the following boundary conditions:

$$S_{l,E}(0) \to 0, \quad S_{l,E}(\infty) \to 0 \tag{22'}$$

and $\alpha_{l,E}$ is an eigenvalue of the coupling constant.

The problem of finding eigenvalues of energy in the Schrödinger equation for a given value of the coupling constant α is transformed into a problem of finding eigenvalues of the coupling constant for a given value of energy and angular momentum. One can easily show that the Sturmian function $S_{nlE}(r) = \langle r | nlE\rangle$ [solution of Eq. (22)] belongs to a complete set with the orthogonality property

$$\langle n'lE | V | nlE\rangle = -\delta_{n',n} \tag{23}$$

(the negative sign corresponds to the case of an attractive potential V) and the closure relation

$$\sum_{n'} | n'lE\rangle\langle nlE | V = -1\,. \tag{24}$$

The Coulomb Sturmian functions are obtained by solving Eq. (22) after replacing $V(r) = -1/r$. Taking into account the boundary conditions [Eq. (22′)], one gets (i) the quantization of the coupling constant:

$$\alpha_{n,l,E} = nx, \quad x = (-2E)^{1/2}, \quad n = 1, 2, 3, \ldots\,. \tag{25}$$

(ii) The explicit form of the Coulomb Sturmian function $S^c_{n,l,E}(r)$:

$$S^c_{n,l,E}(r) = N_{n,l,E}\,e^{-xr}r^l\,{}_1F_1(l+1-n; 2l+2; 2xr)\,. \tag{26}$$

The normalization constant $N_{n,l,E}$ is obtained from the orthogonality relation [Eq. (23)] and reads

$$N_{n,l,E} = \frac{(2x)^{l+1}}{(2l+1)!}\left(\frac{(n+l)!}{(n-l-1)!}\right)^{1/2}. \tag{26'}$$

Returning to the CGF one can give an expansion of its partial-wave component $G_l(r,r';E)$ over the complete set of the Coulomb Sturmian functions for the energy $E<0$ and angular momentum l:

$$G_l(E) = \left(E - H_l' + \frac{1}{r}\right)^{-1} = \sum_{n=l+1}^{\infty} a(n,E)\,| nlE\rangle\langle nlE|\,. \tag{27}$$

From Eq. (22) one has

$$(H_l' - E)\,| nlE\rangle = \alpha_{n,l,E}(1/r)\,| nlE\rangle \tag{28}$$

and from Eq. (27)

$$\left(E - H_l' + \frac{1}{r}\right)\sum_{n=lH}^{\infty} a(n,E)\,| nlE\rangle\langle nlE | = I\,. \tag{29}$$

By comparing these last two relations and by applying the closure theorem, Eq. (24) one gets eventually

$$a(n,E) = (1 - \alpha_{n,l,E})^{-1} \tag{30}$$

and then

$$G_l(E) = \sum_{n=l+1}^{\infty} \frac{| nlE\rangle\langle nlE |}{1 - nx} \tag{31}$$

or explicitly

$$\begin{aligned} G_l(r,r';E) = {} & -\frac{2^{2l+2}x^{2l+1}}{[(2l+1)!]^2}\,e^{-x(r+r')}(rr')^l \\ & \times \sum_{n=l+1}^{\infty}\left(n - \frac{1}{x}\right)^{-1}\frac{(n+l)!}{(n-l-1)!} \\ & \times {}_1F_1(l+1-n; 2l+2; 2xr) \\ & \times {}_1F_1(l+1-n; 2l+2; 2xr')\,. \end{aligned} \tag{32}$$

The main interest in this representation is that the finite summation is extended only over a discrete spectrum, as in Schwinger's Eq. (13).

To date, in spite of its interest, this representation has not been used very often in higher-order calculations. However, we can mention the work by Karule on multiphoton ionization[24] and by Mizuno on the third harmonic generation in H atoms.[25] We shall show its usefulness in Sec. V by applying it to the calculation of cross sections for multiphoton ionization.

Before going further we have to point out that there exists important literature on the properties of the CGF, related to the study of the scattering Coulomb T matrix. In this respect we mention the review paper of Chen and Chen,[10] which gives a good account of the analyticity properties of the Coulomb T matrix and discusses the connection between the various representations of the CGF.

III. RETARDATION EFFECT IN TWO-PHOTON BOUND-BOUND TRANSITIONS

Since 1969, several authors have considered this problem besides the previously quoted papers by Gavrila[12,13] and Klarsfeld[20], we also mention the works by Gorshkov and Polikanov,[26] who used an integral representation of the CGF similar to that of Hostler, and by Fronsdal,[27] who used an algebraic technique.

Although explicit forms may be obtained for specialized transitions,[13] an unpleasant characteristic of all these methods is that the final expression of the amplitude generally contains differential operators which act on hypergeometric functions of several variables. Obviously, this does not facilitate a direct numerical exploitation of the result.

In this work we tried to overcome this obstacle and to derive an explicit analytical expression for the transition amplitude in the general case. With this purpose in mind the calculation was performed in the four-dimensional space $\mathcal{R}^4$ introduced by the Fock transformation, in order to take full advantage of the symmetry properties of the hydrogen atom. In the following, we will first express the transition amplitude in $\mathcal{R}^4$ and then, after introducing a convenient quaternionian formalism,[28] we will derive the explicit expressions for the $1S$-$1S$ and $1S$-$2S$ transition amplitudes.

The amplitude associated with the second-order bound-bound transition between states $|n_1, l_1, m_1\rangle$ and $|n_2, l_2, m_2\rangle$ and corresponding to Fig. 1(a), reads in momentum space

$$\begin{aligned} T^{(2)}_{n_2/n_1}(\omega_1,\omega_2) = \iint d\vec{p}_1 d\vec{p}_2 \Phi^*_{n_2 l_2 m_2}(\vec{p}_2-\vec{k}_2)\vec{p}_2\cdot\vec{\epsilon}_2 \\ \times G(\vec{p}_2,\vec{p}_1;E_{n_1}+\omega_1)\vec{p}_1\cdot\vec{\epsilon}_1 \\ \times\Phi_{n_1 l_1 m_1}(\vec{p}_1-\vec{k}_1)\,, \end{aligned} \tag{33}$$

where the two photons involved are characterized by their energies ω_i, momentum $\vec{k}_i$, and unit polarization vector $\vec{\epsilon}_i$ $(i=1,2)$.

By applying the variable change [Eq. (8)] the amplitude becomes

$$\begin{aligned} T^{(2)}_{n_2/n_1}(\omega_1,\omega_2) = A \iint_{S^3\times S^3} d\xi_1 d\xi_2 Y^*_{n_2 l_2 m_2}(\eta_2)\vec{\xi}_2\cdot\vec{\epsilon}_2 \\ \times[x^2_{n_2}+(\vec{p}_2-\vec{k}_2)^2]^{-2}\Gamma(\xi_2,\xi_1) \\ \times[x^2_{n_1}+(\vec{p}_1-\vec{k}_1)^2]^{-2}\vec{\xi}_1\cdot\vec{\epsilon}_1 \\ \times Y_{n_1 l_1 m_1}(\eta_1)\,, \end{aligned} \tag{34}$$

where

$$A=(x^2+p_1^2)^2(x^2+p_2^2)^2\left(\frac{x_{n_1}x_{n_2}}{x^2}\right)^{5/2},$$

$$\xi_i=(\xi_{i0},\vec{\xi}_i)=\left(\frac{x^2-p_i^2}{x^2+p_i^2},\ \frac{2x\vec{p}_i}{x^2+p_i^2}\right),$$

$$d\xi_i=\left(\frac{2x}{x^2+p_i^2}\right)^3 d\vec{p}_i\,,\quad \vec{\xi}_i\cdot\vec{\epsilon}_i=\frac{2x}{x^2+p_i^2}\vec{p}_i\cdot\vec{\epsilon}_i\,,$$

$$Y_{n_i l_i m_i}(\eta_i)=\frac{[x^2_{n_i}+(\vec{p}_i-\vec{k}_i)^2]^2}{4x^{5/2}_{n_i}}\Phi_{n_i l_i m_i}(\vec{p}_i-\vec{k}_i)\,,$$

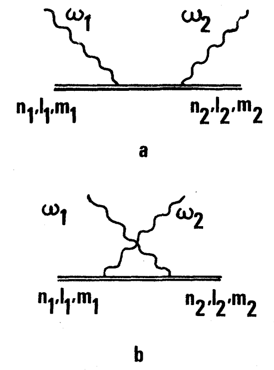

FIG. 1. Diagrams associated to the second-order bound-bound radiative transition between the states $|n_1, l_1, m_1\rangle$ and $|n_2, l_2, m_2\rangle$.

$$x_{n_i}=(-2E_{n_i})^{1/2}=\frac{1}{n_i}\,,$$

$$\eta_i=(\eta_{i0},\vec{\eta}_i)=\left(\frac{x^2_{n_i}-(\vec{p}_i-\vec{k}_i)^2}{x^2_{n_i}+(\vec{p}_i-\vec{k}_i)^2}\,,\ \frac{2x_{n_i}(\vec{p}_i-\vec{k}_i)}{x^2_{n_i}+(\vec{p}_i-\vec{k}_i)^2}\right).$$

In order to perform the integration over the hypersphere S^3, we have to express the propagator $[x^2_{n_i}+(\vec{p}_i-\vec{k}_i)^2]^{-2}$ and the four-vector η_i in terms of the integration variable ξ_i. As pointed out by Gazeau,[28] this task may be conveniently achieved by using a quaternionian formalism. As a matter of fact, the pair $P=(x,\vec{p})$ may be considered as an element of the noncommutative field Q of the quaternions with real coefficients. The Fock transformation corresponds to a stereographic projection Ξ_+ of the elements of Q on the multiplicative subgroup Ω of the quaternions of unit modulus, homeomorphic to S^3 in $\mathcal{R}^4$.

$$P=(x,\vec{p})\xrightarrow{\Xi_+}\xi=PP^{-1}=P^2|P|^{-2}. \tag{35}$$

By using the rules of the algebrā of the quaternions, one gets the four-dimensional version of the propagator

$$[x^2_{n_i}+(\vec{p}_i-\vec{k}_i)^2]^{-1}=\frac{4x^2}{|P_i|^2|K_{n_i}|^2}|\rho_{n_i}\kappa_{n_i}-\xi_i|^{-2}, \tag{36}$$

where

$$P_i=(x,\vec{p}_i),\quad |P_i|^2=P_i\overline{P}_i=x^2+p_i^2,$$

$$\overline{P}_i=(x,-\vec{p}_i);\quad K_{n_i}=(x+x_{n_i},\vec{k}_i),$$

$$\begin{aligned}\rho_{n_i}\kappa_{n_i}&=2xK_{n_i}|K_{n_i}|^{-2}-I\\ &=\left(\frac{x^2-x^2_{n_i}-k_i^2}{(x+x_{n_i})^2+k_i^2}\,,\ \frac{2x\vec{k}_i}{(x+x_{n_i})^2+k_i^2}\right),\end{aligned}$$

the modulus of which being

$$\rho_{n_i} = \left(\frac{(x - x_{n_i})^2 + k_i^2}{(x + x_{n_i})^2 + k_i^2} \right)^{1/2} .$$

Then, the amplitude reads

$$T^{(2)}_{n_2/n_1}(\omega_1, \omega_2) = C_{n_1 n_2} \iint_{S^3 \times S^3} d\xi_1 d\xi_2 Y_{n_2 l_2 m_2}(\eta_2) \vec{\xi}_2 \cdot \vec{\epsilon}_2 \times | \rho_{n_2} \kappa_{n_2} - \xi_2 |^{-4} \Gamma(\xi_2, \xi_1) | \rho_{n_1} \kappa_{n_1} - \xi_1 |^{-4} \vec{\xi}_1 \cdot \vec{\epsilon}_1 Y_{n_1 l_1 m_1}(\eta_1) , \qquad (37)$$

with

$$C_{n_1 n_2} = 2^8 x^3 | K_{n_1} K_{n_2} |^{-4} (x_{n_1} x_{n_2})^{5/2} .$$

On the other hand, the expression of the variable η_i in terms of ξ_i can be obtained by noting that

$$\eta_i = \Xi_+(x_{n_i}, \vec{p}_i - \vec{k}_i) = \Xi_+(\overline{K}_{n_i} - \overline{P}_i) ,$$

which gives

$$\overline{\eta}_i = 2(n_i x)^{-1} | \rho_{n_i} \kappa_{n_i} - \xi_i |^{-2} \times \left((1 + \xi_{i0})(1 + \rho_{n_i} \kappa_{n_i}) - \frac{n_i x}{1 + n_i x} [1 + (\rho_{n_i} \kappa_{n_i})_0](1 + \xi_i) \right) - I . \qquad (38)$$

This result shows that η_i is expressed as a linear combination of the following functions of the integration variable ξ_i:

$$| \rho_{n_i} \kappa_{n_i} - \xi_i |^{-2} \text{ and } \xi_i | \rho_{n_i} \kappa_{n_i} - \xi_i |^{-2} .$$

Since the hyperspherical surface harmonics $Y_{n_i l_i m_i}(\eta_i)$ contained in Eq. (37) are merely finite polynomial expansions in terms of $| \rho_{n_i} \kappa_{n_i} - \xi_i |^{-2q}$ and $\xi_i | \rho_{n_i} \kappa_{n_i} - \xi_i |^{-2q}$, $q \geqslant 2$; it follows that we can consider these expressions as the result of the action of a differential operator with respect to $\rho_{n_i} \kappa_{n_i}$, on the quantity $| \rho_{n_i} \kappa_{n_i} - \xi_i |^{-4}$. Thus the transition amplitude may be written

$$T^{(2)}_{n_2/n_1}(\omega_1, \omega_2) = C_{n_1 n_2} \mathcal{Y}_{n_1 l_1 m_1}(\partial \rho_{n_1}, \partial \rho_{n_1} \kappa_{n_1}) \mathcal{Y}^*_{n_2 l_2 m_2}(\partial \rho_{n_2}, \partial \rho_{n_2} \kappa_{n_2}) \times \iint_{S^3 \times S^3} d\xi_1 d\xi_2 \vec{\xi}_2 \cdot \vec{\epsilon}_2 | \rho_{n_2} \kappa_{n_2} - \xi_2 |^{-4} \Gamma(\xi_2, \xi_1) | \rho_{n_1} \kappa_{n_1} - \xi_1 |^{-4} \vec{\xi}_2 \cdot \vec{\epsilon}_1 , \qquad (39)$$

where

$$\partial \rho_{n_i} \kappa_{n_i} = (\partial_{(\rho_{n_i} \kappa_{n_i})_0}, \nabla_{\rho_{n_i} \vec{\kappa}_{n_i}}) .$$

The calculation is then reduced to the evaluation of the remaining integral, to the determination of the explicit form of the operators $\mathcal{Y}$, and of their action on the result. Now, the integral has been calculated by Gazeau,[28] who obtained

$$\iint_{S^3 \times S^3} d\xi_1 d\xi_2 \vec{\xi}_2 \cdot \vec{\epsilon}_2 | \rho_{n_2} \kappa_{n_2} - \xi_2 |^{-4} \Gamma(\xi_2, \xi_1) | \rho_{n_1} \kappa_{n_1} - \xi_1 |^{-4} \vec{\xi}_1 \cdot \vec{\epsilon}_1 = 4\pi^2 \left(\frac{\vec{\epsilon}_1 \cdot \vec{\epsilon}_2}{4} \mathcal{J}_2^{2-\nu} + (\rho_{n_1} \vec{\kappa}_{n_1} \cdot \vec{\epsilon}_2)(\rho_{n_2} \vec{\kappa}_{n_2} \cdot \vec{\epsilon}_1) \mathcal{J}_3^{3-\nu} \right) . \qquad (40)$$

Here, the functions $\mathcal{J}_a^b$ are given by

$$\mathcal{J}_a^b = (\rho_{n_1 >} \rho_{n_2 >})^{-2a} \int_0^1 dz\, z^{b-1} | 1 - R z \kappa_{n_1} \kappa_{n_2} |^{-2a} , \qquad (41)$$

$$\mathcal{J}_a^b = (\rho_{n_1 >} \rho_{n_2 >})^{-2a} (1/b) F_1(b, a, a, b+1; R e^{i\omega}, R e^{-i\omega}) , \qquad (41')$$

where

$$R = (\rho_{n_1 <} \rho_{n_2 <} / \rho_{n_1 >} \rho_{n_2 >}) ,$$

$$\rho_{n_i >} = \sup(\rho_{n_i}, 1);$$

$$\rho_{n_i <} = \inf(\rho_{n_i}, 1);$$

$$b > 1, \quad \cos\omega = (\kappa_{n_1} \overline{\kappa}_{n_2})_0 ,$$

and $F_1(\alpha, \beta, \beta', \gamma; x, y)$ is an Appell hypergeometric function of two variables.[29] The operators $\mathcal{Y}$ which enter Eq. (39) are obtained from the explicit form of the corresponding functions $Y_{nlm}(\eta_i)$. For instance we have, respectively,

$$Y_{100}(\eta_i) = (2\pi^2)^{-1/2} ,$$
$$Y_{200}(\eta_i) = (2\pi^2)^{-1/2} 2\eta_{i0} , \qquad (42)$$

and thus

$$\mathcal{Y}_{100} = (2\pi^2)^{-1/2}, \tag{43}$$

$$\begin{aligned}\mathcal{Y}_{200}(\partial_{\rho_2}, \partial_{\rho_2\kappa_2}) &= (2\pi^2)^{-1/2}\, 2\{x_2^2 |K_2|^{-2} \\ &\quad\times[\partial_{(\rho_2\kappa_2)_0} + (2+x^{-1})x\rho_2^{-1}\partial_{\rho_2}\rho_2^2] - 1\}.\end{aligned} \tag{43$'$}$$

The last result is obtained by using the following derivation formulas:

$$\begin{aligned}|\rho\kappa - \xi|^{2q} &= \rho^{2-q}(q-1)^{-1}(1-\rho^2)^{-1}\partial_\rho \\ &\quad\times[\rho^{q-1}|\rho\kappa - \xi|^{-2(q-1)}],\end{aligned} \tag{44}$$

$$\begin{aligned}\xi\,|\rho\kappa - \xi|^{-2q} &= \rho\kappa\,|\rho\kappa - \xi|^{-2q} \\ &\quad + 2^{-1}(q-1)^{-1}\partial_{\rho\kappa}|\rho\kappa - \xi|^{-2(q-1)}.\end{aligned} \tag{44$'$}$$

From the above we can get easily the desired explicit expressions for transition amplitudes related to the following.

A. Elastic scattering on a hydrogenic atom in the fundamental state

By specializing the general formula (40) with the help of the transformation relations previously given, one gets

$$\begin{aligned}T^{(2)}_{1/1}(\omega,\omega) &= 2C_{1,1}\bigg(\frac{\vec{\epsilon}_1\cdot\vec{\epsilon}_2}{4}\,\frac{1}{2-\nu}F_1(2-\nu,2,2,3-\nu;z,\bar{z}) \\ &\quad + \frac{4x^2}{[(x+1)^2+k^2]^2}(\vec{k}_1\cdot\vec{\epsilon}_2)(\vec{k}_2\cdot\vec{\epsilon}_1) \\ &\quad \times\frac{1}{3-\nu}F_1(3-\nu,3,3,4-\nu;z,\bar{z})\bigg),\end{aligned} \tag{45}$$

where

$$z = \frac{(x-1)^2+k^2}{(x+1)^2+k^2}\exp\{i\arccos[(\kappa_1\kappa_2)_0]\},$$

$$\nu = Z/x\,.$$

This result coincides exactly with Gavrila's.[12] We can add, however, that here the structure of the variable z permits us to display the angle between two four-dimensional vectors κ_1 and κ_2.

B. Two-photon transition between 1*S* and 2*S* states (Ref. 30)

The formal result is slightly more complicated than the preceding one, owing to the structure of the operator $\mathcal{Y}_{200}$ [Eq. (43′)]. One eventually gets

$$\begin{aligned}T^{(2)}_{2/1}(\omega_1,\omega_2) &= C_{1,2}x_2^2|K_2|^{-2} \\ &\quad\times[\vec{\epsilon}_1\cdot\vec{\epsilon}_2\alpha_2 + 4(\rho_1\vec{\kappa}_1\cdot\vec{\epsilon}_2)(\rho_2\vec{\kappa}_2\circ\vec{\epsilon}_1)\alpha_3],\end{aligned} \tag{46}$$

where

$$\begin{aligned}\alpha_m &= 2m\rho_1(\kappa_{10}-\kappa_{20}\cos\omega)\,\mathcal{J}_{m+1}^{m+1-\nu} \\ &\quad + \left(2+\nu-|K_2|^2x_2^{-2} - \frac{(\rho_2\kappa_2)_0}{m-\nu}\right)\mathcal{J}_m^{m-\nu} \\ &\quad + \left(\frac{2+\nu}{\nu}+\rho_2^{-1}\kappa_{20}\right)|1-R\kappa_1\kappa_2|^{-2m},\end{aligned} \tag{46$'$}$$

with

$$(\rho_{n_i}\kappa_{n_i})_0 = \frac{x^2-x_{n_i}^2-k_i^2}{(x+x_{n_i})^2+k_i^2}\,, \quad \cos\omega = (\kappa_1\kappa_2)_0\,,$$

$$\rho_{n_i}^2 = \frac{(x-x_{n_i})^2+k_i^2}{(x+x_{n_i})^2+k_i^2} < 1\,,$$

$$R = \rho_1\rho_2\,.$$

By taking the limiting values $k_i \to 0$, which correspond to the dipole approximation, the amplitude reduces to

$$\begin{aligned}[T^{(2)}_{2/1}(\omega_1,\omega_2)]_{\text{dip}} &= \sqrt{2}\,\tfrac{64}{27}\nu^2(\nu+1)^{-1}(4-\nu^2)^{-1} \\ &\quad\times[\tfrac{1}{3}(1+\nu) - {}_2F_1(1,-1-\nu;3-\nu;R_{\text{dip}})],\end{aligned} \tag{47}$$

which is the known result given previously by Klarsfeld[20,31] and Rapoport and Zon.[32]

By working out the calculation in the four-dimensional space introduced by the Fock transformation, we have encountered several advantages. First, we think that after a simple assimilation of the quaternionian formalism, calculations are easier and more compact than in the ordinary momentum space. Second, it appears that this method is quite general and can be applied, in theory, to any second-order bound-bound transition.

However, in spite of the generality of this technique its application to problems concerning higher excited states becomes difficult on account of the increasing complexity of the differential operators $\mathcal{Y}$ involved. This fact has induced us to explore the possibilities of other representations of the CGF.

IV. BETHE SUMS FOR LAMB-SHIFT CALCULATIONS
(Ref. 33)

The Lamb-shift of an energy level (n,l,j) in a hydrogenic atom reads, to the first order in α,[34]

$$\Delta E_{n,l,j} = \frac{4\alpha mc^2}{3\pi n^3}\sum_{i\geq 4}\sum_{k\geq 0} A_{ik}(Z\alpha)^i\ln^k(Z\alpha)^{-2}. \tag{48}$$

If we assume a pointlike nucleus, with a charge Z and an infinite mass, the first terms of the expansion are, respectively,[34]

$$A_{41}=\delta_{l,0}\,,$$
$$A_{40}=\left(\tfrac{11}{24}-\tfrac{1}{5}\right)\delta_{l,0}+\frac{3}{8}\frac{C_{l,j}}{2l+1}-\ln k_0(n,l)\,, \tag{48$'$}$$

where

$$C_{l,j}=(l+1)^{-1}\delta_{j,\,l+1/2}-l^{-1}\delta_{j,\,l-1/2}$$

corresponds to the contribution of the anomalous magnetic moment of the electron; $-\frac{1}{5}$ comes from the vacuum polarization [see Fig. 2(a)] and $\ln k_0(n,l)$, corresponding to the nonrelativistic contribution of the self-energy [see Fig. 2(b)] is the so-called Bethe logarithm or logarithmic average excitation energy of the level (n,l).[35]

To date, the calculation of higher-order terms of this expansion was partially completed by Erickson in 1971,[36] and many authors are currently carrying on the calculation of the terms in α^2.

It is interesting to recall that the lowest-order terms A_{40} and A_{41} contribute 99.3% to the total shift $E_{2S_{1/2}}-E_{2P_{1/2}}$ in hydrogen and that the theory gives a fairly good account of the higher-order terms from which the global contribution amounts to 0.7%. Now, the components of the coefficients A_{40} and A_{41} are all exactly known with the exception of the Bethe logarithm, the evaluation of which involves the calculation of a second-order amplitude. Note that the contribution of the Bethe logarithm amounts to about −369.8 MHz as compared with a total positive shift $E_{2S_{1/2}}-E_{2P_{1/2}}\sim+1057.9$ MHz. This led us to perform a new accurate calculation which will permit a better comparison with the experimental results. We shall show below the interest in using the CGF in such a computation for the states $1S$ to $4P$.

The Bethe logarithm corresponds to the finite part of the nonrelativistic contribution of the self-energy [see Fig. 2(b)] to the Lamb shift. It is usually defined either as[35]

$$\ln k_0(n,l)=\frac{\sum_{n',l'}|\langle n,l|\vec{\mathrm{p}}|n',l'\rangle|^2(E_{n'}-E_n)\ln|E_{n'}-E_n|}{\sum_{n',l'}|\langle n,0|\vec{\mathrm{p}}|n',l'\rangle|^2(E_{n'}-E_n)}\,, \tag{49}$$

or as[2]

$$\ln k_0(n,l)=\lim_{K\to\infty}\left[\tfrac{1}{8}n^3\Lambda_{n,l}(K)-\tfrac{1}{4}nK+\delta_{l,0}\ln K\right], \tag{50}$$

with

$$\Lambda_{n,l}(K)=\mathrm{P}\int_0^K\omega\,d\omega\,\langle nl|\vec{\mathrm{p}}G(E_n-\omega)\vec{\mathrm{p}}|nl\rangle\,, \tag{51}$$

where P denotes Cauchy's principal value of the integral. Note that the calculation is made within the dipole approximation. Bethe[35] has demonstrated that the retardation effect may be neglected to a very good approximation. For a more recent discussion of this point, see the paper by Au and Feinberg.[54]

Calculations of $\ln k_0(n,l)$ were first performed, for the low-lying states, by Bethe, Brown, and Stehn[37] and improved later on by Harriman,[38] who extended the calculations up to the $4p$ state. These calculations were based on the direct evaluation of the definition formula [Eq. (49)] using term-by-term summation for the contribution of intermediate discrete states and numerical integration for that of intermediate continuum states.

More accurate values were obtained later on, with the help of methods which permit the exact implicit evaluation of the infinite sum appearing in the equivalent definition [Eq. (50)]. We must recall here the works by Schwartz and Tiemann,[2] who used Dalgarno's method, by Lieber,[15] who used the Schwinger's representation of the CGF [Eq. (13)], and by Huff,[39] who used Fronsdal's[27] algebraic approach.

These very accurate calculations were restricted to the lowest-lying states owing to the difficulty of the analytical computation. Now several recent experiments[40] have been performed for many higher excited states. Thus it seemed interesting to provide accurate theoretical values for these states. In order to achieve this purpose, we used Hostler's integral representation [Eq. (18)], which is well adapted for obtaining analytical expressions.

The main part of the calculation deals with the evaluation of the integral $\Lambda_{n,l}(K)$ [Eq. (51)], the computation of which may be split into two steps. First we have to obtain an analytical expression of the second-order matrix element $\langle nl|\vec{\mathrm{p}}G(E_n-\omega)\vec{\mathrm{p}}|nl\rangle$; then we must perform the integration over the photon energy ω.

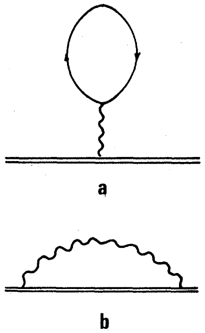

FIG. 2. Lowest-order contributions to the Lamb shift: (a) vacuum polarization; (b) self-energy.

A. Analytical expression of $\langle nl|\vec{\mathrm{p}}\,G(E_n-\omega)\,\vec{\mathrm{p}}\,|nl\rangle$

It is easily shown that after performing the angular calculation, the matrix element can be reduced as follows:

$$\langle nl\,|\,\vec{\mathrm{p}}\,G(E_n-\omega)\,\vec{\mathrm{p}}\,|\,nl\rangle = \frac{l+1}{2l+1}\int_0^\infty r^2dr\int_0^\infty r'^2dr'a(r)\,G_{l+1}(r,r';E_n-\omega)\,a(r')$$
$$+\frac{l}{2l+1}\int_0^\infty r^2dr\int_0^\infty r'^2dr'b(r)\,G_{l-1}(r,r';E_n-\omega)\,b(r')\,, \tag{52}$$

where

$$a(r)=\left(\frac{d}{dr}-\frac{l}{r}\right)R_{n,l}(r)\,,$$
$$b(r)=\left(\frac{d}{dr}+\frac{l+1}{r}\right)R_{n,l}(r) \tag{52$'$}$$

and $R_{n,l}(r)$ is the radial hydrogenic function

$$R_{n,l}(r)=C_{n,l}e^{-r/n}r^l\;{}_1F_1(-n+l+1;2l+2;2r/n)\,, \tag{52$''$}$$

with

$$C_{n,l}=2^{2l+1}n^{-l-2}\frac{1}{(2l+1)!}\left(\frac{(n+l)!}{(n-l-1)!}\right)^{1/2}.$$

Given the structure of the functions $R_{n,l}(r)$ one sees that the desired amplitude [Eq. (52)] may be expressed as a linear combination of integrals

$$J_{\lambda,p}^{\nu,\nu'}(W)=\int_0^\infty dr\,r^{\lambda+p}$$
$$\times\int_0^\infty dr'r'^{(\lambda+p)}e^{-(\nu r+\nu' r')}G_\lambda(r,r';W), \tag{53}$$

where p is an integer $\geqslant 1$.

Integrals of this type are easily evaluated in the particular case $p=1$, by using Hostler's integral representation [Eq. (18)] of $G_\lambda(W)$.[32] One has

$$J_{\lambda,1}^{\nu,\nu'}(W)=\frac{2^{\lambda+2}(2\lambda+1)!\,x^{2\lambda+1}}{[(x+\nu)\,(x+\nu')]^{2\lambda+2}}\left(\lambda+1-\frac{1}{x}\right)^{-1}$$
$$\times{}_2F_1(\lambda+1-1/x,2\lambda+2;\lambda+2-1/x;z)\,, \tag{54}$$

where $z=(\nu-x)\,(\nu'-x)/(\nu+x)\,(\nu'+x)$.

General integrals $J_{\lambda,p}^{\nu,\nu'}(W)$ are obtained from this result by noting that

$$J_{\lambda,p}^{\nu,\nu'}(W)=\left(\frac{\partial^{(p-1)}}{\partial\nu^{(p-1)}}\right)\left(\frac{\partial^{(p-1)}}{\partial\nu'^{(p-1)}}\right)J_{\lambda,1}^{\nu,\nu'}(W)\,, \tag{55}$$

which is symmetrical with respect to ν and ν' and by using repeatedly the derivation formula

$$\frac{\partial}{\partial\nu}J_{\lambda,1}^{\nu,\nu'}(W)=2(\nu^2-x^2)^{-1}$$
$$\times\left[\frac{(2\lambda+1)!}{(\nu+\nu')^{2\lambda+2}}\right.$$
$$\left.-\left(\nu(\lambda+1)-\frac{1}{x}\right)J_{\lambda,1}^{\nu,\nu'}(W)\right]. \tag{56}$$

Note that this last formula shows that the amplitude can be expressed in terms of the particular integral $J_{\lambda,1}^{\nu,\nu'}(W)$, i.e., of the corresponding Gauss hypergeometric function [Eq. (54)]. Expressions of the amplitudes $\langle nl\,|\,\vec{\mathrm{p}}\,G(E_n-\omega)\,\vec{\mathrm{p}}\,|\,nl\rangle$ for all the states $1S$ to $4P$ obtained in this way are listed in Table I. It should be noted that these expressions also may be used for the calculation of the amplitudes related to various other second-order processes such as, for instance, the elastic scattering of photons or the so-called "light shift" of the considered level of an atom irradiated by an intense laser light.

B. Calculation of the integrals $\Lambda_{n,l}(K)$

The second part of the calculation deals with the evaluation of the finite part of the integral $\Lambda_{n,l}(K)$ [Eq. (51)], which can be written, by introducing the integration variable $y=(nx)^{-1}=(1+n^2\omega)^{-1/2}$:

$$\Lambda_{n,l}(K)=\frac{2}{n^4}P\int_{(1+n^2K)^{-1/2}}^1 dy\,y^{-5}(1-y^2)$$
$$\times\langle nl\,|\,\vec{\mathrm{p}}\,G\vec{\mathrm{p}}\,|\,nl\rangle\,. \tag{57}$$

As a first step we have to extract from the integrand, the diverging terms corresponding to the factors $(-\frac{1}{4}nK+\delta_{l,0}\ln K)$ contained in Eq. (50) and which behave like y^{-3} or y^{-1} for $y\rightarrow 0$ (i.e., $K\rightarrow\infty$). It is thus necessary to study the corresponding asymptotic behavior of the Gauss hypergeometric functions involved in the expression of $\langle nl|\vec{\mathrm{p}}\,G(E_n-\omega)\vec{\mathrm{p}}\,|nl\rangle$. This may be conveniently achieved by using the known expansion of these functions[41]

TABLE I. Analytical expressions of the second-order amplitudes $\langle n,l\,|\,\vec{\mathrm{p}}\,G(E_n-\omega)\vec{\mathrm{p}}\,|\,n,l\rangle$. Notations: $y=(1+n^2\omega)^{-1/2}$; $0\le\omega\le K\to\infty$; $1\ge y\ge(1+n^2K)^{-1/2}\to 0$ for $K\to\infty$; $z=(1-y)^2(1+y)^{-2}$. The energies are given in Rydberg units.

State	$\langle n,l\,\vert\,\vec{\mathrm{p}}\,G(E_n-\omega)\vec{\mathrm{p}}\,\vert\,n,l\rangle$
1S	$2^7 3y^5(2-y)^{-1}(1+y)^{-8}\,{}_2F_1(4,2-y;3-y;z)$
2S	$2^8 3y^5(1-y)^{-1}(1+y)^{-8}\,{}_2F_1(4,2-2y;3-2y;z)\ +6y^2(y^2-1)^{-1}$
3S	$2^7 3y^5(2-3y)^{-1}(1+y)^{-8}(y^2-1)^{-1}(7y^2-3)^2\,{}_2F_1(4,2-3y;3-3y;z)+2^3y^2(y^2-1)^{-2}(5y^2-1)$
4S	$2^{10}3^{-1}y^5(1-2y)^{-1}(1+y)^{-8}(y^2-1)^{-4}(23y^4+18y^2+3)^2\,{}_2F_1(4,2-4y;3-4y;z)+2\ 3^{-1}y^2(y^2-1)^{-4}(733y^6-935y^4+375y^2-45)$
2P	$2^{12}3^{-1}5y^7(3-2y)^{-1}(1+y)^{-12}\,{}_2F_1(6,3-2y;4-2y;z)+2\ 3^{-2}y^2(y^2-1)^{-2}(5y^2+3)+2^8 3^{-2}y^7(1-2y)^{-1}(y^2-1)^{-2}(1+y)^{-4}\,{}_2F_1(2,1-2y;2-2y;z)$
3P	$2^9 3^{-1}y^7(1-3y)^{-1}(y-1)^{-4}(y+1)^{-8}(3y^2-1)\,{}_2F_1(6,3-2y;4-2y;z)+2\ 3^{-1}y^2(y^2-1)^{-4}(59y^6-29y^4+y^2+1)$
	$+2^{13}3^{-1}5y^7(1-y)^{-1}(1+y)^{-12}\,{}_2F_1(6,3-3y;4-3y;z)+2^2 3^{-1}5y^2(y^2-1)^{-1}$
4P	$2^2 3^{-1}y^2(y^2-1)^{-2}(35y^2-19)+2^{14}3^{-1}y^7(y^2-1)^{-2}(9y^2-5)^2(3-4y)^{-1}(1+y)^{-12}\,{}_2F_1(6,3-4y;4-4y;z)-2\ 3^{-1}y^2(y^2-1)^{-1}$
	$+2^{10}3^{-2}5^{-1}y^7(y^2-1)^{-6}(57y^4-38y^2+5)(1-4y)^{-1}(1+y)^{-4}\,{}_2F_1(2,1-4y;2-4y;z)+2^5 3^{-2}5^{-1}y^4(y^2-1)^{-6}(33y^4-18y^2+1)(57y^4-38y^2+5)$
3D	$2^{15}3\ 5^{-1}7y^9(4-3y)^{-1}(1+y)^{-16}\,{}_2F_1(8,4-3y;5-3y;z)+2^{11}3\ 5^{-1}y^9(2-3y)^{-1}(1+y)^{-10}(y-1)^{-2}\,{}_2F_1(4,2-3y;3-3y;z)+2^2 5^{-1}y^2(y^2-1)^{-2}(y^2+5)$

$$\begin{aligned} b^{-1}{}_2F_1(m,b;b+1;z) &= \frac{1}{m-1}(1-z)^{1-m}+\frac{(m-1-b)}{(m-1)(m-2)}(1-z)^{2-m}+\cdots \\ &\quad+\frac{(m-1-b)\cdots(2-b)}{(m-1)!}(1-z)^{-1} \\ &\quad+\frac{(m-1-b)\cdots(1-b)}{(m-1)!}\sum_{q=0}^{\infty}\frac{z^q}{q+b}\,. \end{aligned} \tag{58}$$

Since $(1-z)=4y(1+y)^{-2}$, it may be verified that we have separated in this way all the diverging terms containing negative powers of y. Then by replacing y by its value in terms of K at the lower limit of the integration range we have to verify that the factors $-\frac{1}{4}nK+\delta_{l,0}\ln K$, contained in the definition [Eq. (50)] of Bethe's logarithm, are exactly cancelled. Furthermore, it is interesting to note that this procedure provides a very good test *a posteriori* of the validity of the expressions of $\langle nl\,|\,\vec{\mathrm{p}}\,G(E_n-\omega)\vec{\mathrm{p}}\,|\,nl\rangle$ listed in Table I.

The remaining integration over the interval $y\in\,]0,1]$ can be performed almost entirely analytically since the integrand reduces essentially to a linear combination of algebraic terms which are of the following general form:

$$\begin{aligned} J(p,q) &= \int_0^1 dy\, y^p(1+y)^{-q} \\ &= 2^{1-q}(p+1-q)^{-1}-p(p+1-q)^{-1}J(p-1,q)\,, \end{aligned} \tag{59}$$

with

$$J(p,p+1)=-2^{-p}p^{-1}+J(p-1,p)\,,$$

$$J(0,q)=(1-2^{1-q})(q-1)^{-1}\,,\quad J(0,1)=\ln 2\,.$$

The only term which requires a numerical computation comes from the infinite sum included in the expansion [Eq. (58)] of the hypergeometric function. This sum may be written as follows:

$$\sum_{q=0}^{\infty} z^q(q+b)^{-1}=b^{-1}+\sum_{q=1}^{\infty} z^q(q+b)^{-1}\,, \tag{60}$$

where $z=(1-y)^2(1+y)^{-2}$, $z\in[0,1[$, and $b=\lambda+1-ny$. The last sum clearly diverges at the limit $z\to 1$ (i.e., $y\to 0$). However, this difficulty is only apparent, since

$$\begin{aligned} \sum_{q=1}^{\infty}\frac{z^q}{q+b} &= \sum_{q=1}^{\infty}\frac{z^q}{q}-b\sum_{q=1}^{\infty}\frac{z^q}{q(q+1)} \\ &\quad-b(b-1)\sum_{q=1}^{\infty}\frac{z^q}{q(q+1)(q+b)} && (60') \\ &= -\ln(1-z)-b\left(1+\frac{1-z}{z}\ln(1-z)\right) \\ &\quad-b(b-1)\sum_{q=1}^{\infty}\frac{z^q}{q(q+1)(q+b)} && (60'') \end{aligned}$$

and that the integration reduces, in fact, to the calculation of elementary integrals of the following type:

$$\int_0^1 d\chi\, \chi^p \ln(1-\chi^2) = \frac{2}{p+1}[\psi(\tfrac{1}{2}(p+3)) - \psi(1)]\,, \quad p>-1\,, \tag{61}$$

where $\psi(z)$ is the logarithmic derivative of the gamma function, and to the numerical quadrature of an integral containing the remaining sum Eq. (60″), which converges at least as

$$\sum_{q=1}^{\infty} q^{-3} = \zeta(3) = 1.202\,056\,903\,1\,.$$

The routine used was a double-precision-arithmetic 24-point Gaussian quadrature which ensured a very good precision.

The numerical results obtained by this method are listed in Table II. As an independent check of our analytical calculations we also performed an entirely numerical integration of the finite part of the integral $\Lambda_{n,l}(K)$. The results obtained by these two methods coincide up to 12 significant digits.

The comparison of our results with those previously obtained by the above-mentioned authors shows an excellent agreement with the most precise results of Huff[39] for the lowest-lying states. Note that the very recent calculation by Shimamura[42] agrees equally well with our results. On the other hand one observes some discrepancies with Harriman's results,[38] the most important one being for the $3p$ state. However, even such differences remain hardly significant from the experimental point of view.

TABLE II. Numerical values of the Bethe logarithm for states 1S to 4P. The numbers in parentheses represent the estimated error for the previous results. The uncertainty in our values should be less than three units in the last decimal place. Note that Shimamura's results (Ref. 42) were published after this calculation was completed.

State	Previous results	Ref.	This work (Ref. 33)
1S	2.984 149(3)	38	2.984 128 555 8
	2.984 128 5(3)	15	
	2.984 128 555 9(3)	39	
	2.984 128 555 765 50	42	
2S	2.811 798(9)	38	2.811 769 893 1
	2.811 769 883(28)	2	
	2.811 769 8(3)	15	
	2.811 769 893 2(5)	39	
	2.811 769 893 131	42	
3S	2.767 699(8)	38	2.767 663 612 5
4S	2.749 859(9)	38	2.749 811 840 5
2P	−0.030 016 37(1)	38	−0.030 016 708 6
	−0.030 016 697(12)	2	
	−0.030 016 75(6)	15	
	−0.030 016 708 9(3)	39	
3P	−0.038 188 52(1)	38	−0.038 190 229 4
4P	−0.041 954 0(3)	38	−0.041 954 894 6
3D	−0.005 232 1(2)	38	−0.005 232 148 1

As a comment on this part of our work, we note that the use of Hostler's integral representation [Eq. (18)] of the CGF has proven to be very useful in the analytical calculation of the second-order amplitudes considered here. The method is quite general and can be applied successfully to the study of any second-order radiative process within the limits of validity of the dipole approximation. However, we encounter some limitations in its application when highly excited atomic states are involved. As a matter of fact, the analytic derivation of the expressions of the transition amplitudes becomes very intricate since these expressions are more and more complicated as may be seen in Table I. This circumstance has induced us to use a somewhat different approach in order to compute the Bethe logarithm in higher excited states $4<n\leqslant 8$. The entirely numerical technique used is analogous to Harriman's, and the results we obtained in this way are given in Ref. 33.

On the contrary, the method we used in studying higher-order multiphoton processes does not suffer from these limitations. We present it in Sec. V.

V. MULTIPHOTON IONIZATION OF ATOMIC HYDROGEN

Since the advent of powerful laser sources, the study of multiphoton processes has become an important subject of experimental and theoretical investigations.[43] Most of the theoretical work was done within the framework of the perturbation theory and thus required the evaluation of higher-order ($N\geqslant 2$) amplitudes. As is well known, "exact" calculations of this type can be achieved for hydrogenic systems by using either Dalgarno's method[1-5] or the CGF method.

As previously mentioned, the calculation of second-order amplitudes presents no major difficulty within the framework of both methods, but the generalization to higher orders is not straightforward. Gontier and Trahin[5] have succeeded in generalizing Dalgarno's method to the calculation of higher-order amplitudes. On the contrary, application of the CGF representations previously used in this paper to such calculations proved rather deceptive owing to the increasing complexity of the formalism. However, as we will show, such difficulties may be overcome by using the Sturmian representation of the CGF defined in Sec. II D. The first attempts in this direction were done by Karule[24] and Mizuno.[25] We present here our own computations of cross sections for multiphoton ionization in hydrogen.

A. Three-photon ionization

For the sake of clarity we shall first outline in detail the calculation for the three-photon process. As a matter of fact it represents an immediate generalization of the two-photon case, and a further extension to higher-order calculations can be carried out without additional difficulty, as we will show further on.

The differential cross section for three-photon ionization reads, in atomic units,

$$\frac{d\sigma}{d\Omega(\hat{k})}=\frac{\alpha}{2\pi}a_0^2\left(\frac{I}{2I_0}\right)^2\omega k\,|\mathfrak{M}^{(3)}_{f/i}|^2\,, \tag{62}$$

where α is the fine-structure constant, $a_0=5.2917\times10^{-9}$ cm is the radius of the first Bohr orbit, I is the field strength intensity of the radiation field, $I_0=7.019\times10^{16}$ W/cm^2 is the atomic unit of field-strength intensity, k is the momentum of the photoelectron ejected in direction of the unit vector $\hat{k}$. The magnitude of k is given by conservation of energy:

$$\tfrac{1}{2}k^2=E_i+3\omega\,,$$

where E_i is the energy of the initial atomic bound state and ω is the energy of the incoming photons.

The third-order amplitude $\mathfrak{M}^{(3)}_{f/i}$ corresponding to the transition of the atomic system from the initial bound state $|i\rangle$ to the final one $|f\rangle$ belonging to the continuum, reads

$$\mathfrak{M}^{(3)}_{f/i}=\sum_{s_1,s_2}\frac{\langle f|\vec{\epsilon}\cdot\vec{r}\,|s_2\rangle\langle s_2|\vec{\epsilon}\cdot\vec{r}\,|s_1\rangle\langle s_1|\vec{\epsilon}\cdot\vec{r}\,|i\rangle}{(E_i+2\omega-E_{s_2})(E_i+\omega-E_{s_1})}\,, \tag{63}$$

where $\vec{\epsilon}$ is the unit polarization vector of the incident radiation field.

The atomic system is presumed to be initially in an arbitrary bound state

$$\langle\vec{r}\,|i\rangle=\langle\vec{r}\,|nlm\rangle=R_{n,l}(r)\,Y_{l,m}(\hat{r})\,.$$

The intermediate (virtual) states $|s_i\rangle$ belong to the complete set of eigenstates of the atomic Hamiltonian

$$\langle\vec{r}\,|s_i\rangle=\langle\vec{r}\,|\nu_i\lambda_i\mu_i\rangle=R_{\nu_i\,\lambda_i}(r)\,Y_{\lambda_i\,\mu_i}(\hat{r})$$

and the wave function of the photoelectron $\langle\vec{r}\,|f\rangle=\langle\vec{r}\,|\vec{k}\rangle$ is given as the partial-wave expansion

$$\langle\vec{r}\,|\vec{k}\rangle=\left(\frac{8\pi^3}{k}\right)^{1/2}\sum_{L,M}i^L e^{-i\eta_L}R_{k,L}(r)\times Y_{L,M}(\hat{r})\,Y^*_{L,M}(\hat{k})\,, \tag{64}$$

where $\eta_L=\arg\Gamma(L+1-i/k)$ is the Coulomb phase shift and $R_{k,l}(r)$ is the radial wave function.

The calculation is comprised of two steps which correspond to the evaluation of, respectively, the angular part and of the radial part, the latter representing the main difficulty. It should be noted that the angular part depends explicitly on the polarization state of the incident light which induces a "polarization effect" characteristic for multiphoton processes.[44,45] In consequence, it seemed of interest to display the result of the calculation for both linear and circular polarization.

1. Angular calculation

With a convenient choice of the coordinate-system orientation one shows that the dipole interaction operator assumes the forms[46]:

$$\vec{\epsilon}\cdot\vec{r}=(\tfrac{4}{3}\pi)^{1/2}r\,Y_{1,0}(\hat{r}) \tag{65}$$

for linear polarization, and

$$\vec{\epsilon}\cdot\vec{r}=-(\tfrac{4}{3}\pi)^{1/2}r\,Y_{1,1}(\hat{r}) \tag{65$'$}$$

for right circular polarization.

The angular calculation is thus reduced to the evaluation of known integrals over a product of three spherical harmonics. Such integrals may be expressed in terms of Wigner 3-j symbols,[47] and one easily gets

$$\left(\frac{4\pi}{3}\right)^{1/2}\langle Y_{\lambda,m}|\,Y_{1,0}|\,Y_{l,m}\rangle=\left(\frac{l_>^2-m^2}{(2l+1)(2\lambda+1)}\right)^{1/2}, \qquad l_>=\sup(l,\lambda)\,, \tag{66}$$

$$\left(\frac{4\pi}{3}\right)^{1/2}\langle Y_{\lambda,m+1}|\,Y_{1,1}|\,Y_{l,m}\rangle=\left(\frac{(l+1+m)(\lambda+1+m)}{2(2l+1)(2\lambda+1)}\right)^{1/2}\delta_{\lambda,l+1}-\left(\frac{(l+1-m)(\lambda+1-m)}{2(2l+1)(2\lambda+1)}\right)\delta_{\lambda,l-1}\,. \tag{66$'$}$$

As a consequence of the selection rule, $\lambda=l\pm1$, the amplitude $\mathfrak{M}^{(3)}_{f/i}$ can be split into four components, each of which corresponds to one of the accessible values for the final angular momentum L:

$$\mathfrak{M}^{(3)}_{f/i}=(8\pi^3/k)^{1/2}(M_{l+3}+M_{l+1}+M_{l-1}+M_{l-3})\,. \tag{67}$$

Clearly the expressions of the partial amplitudes M_L differ according to the polarization state of the light. They are displayed in Table III.

After inserting $\mathfrak{M}^{(3)}_{f/i}$ [Eq. (67)] in Eq. (62) and summing over the magnetic quantum number in the final state, one gets the total cross section by integrating over the propagation direction of the photoelectron. Eventually, by averaging over the magnetic quantum number m in the initial state:

$$\sigma_{n,l}=\frac{1}{2l+1}\sum_{m=-l}^{+l}\sigma_{nlm}\,,$$

one obtains the expressions given in Table IV for three-photon ionization cross section either for linear (σ) or circular (σ') polarization of the incident light.

Incidentally, from these results one can check that the maximum allowed value of the ratio σ'/σ is equal to $\frac{5}{2}$ as previously derived for s states by Lambropoulos[44] and independently by Klarsfeld and Maquet.[45] Moreover, one can demonstrate that this maximum value is independent of the initial state $|n,l\rangle$ considered by verifying that the difference $\frac{5}{2}\sigma_{n,l}-\sigma'_{n,l}$ remains positive. The result reads

$$\tfrac{5}{2}\sigma_{n,l}-\sigma'_{n,l}=C\,\frac{1}{30(2l+1)}\left[(l+1)\,|Q_{l+1}|^2+l\,|Q_{l-1}|^2\right],\tag{68}$$

where

$$Q_{l+1}=\frac{2(l+2)}{2l+3}T_{l+1,l+2,l+1}+\frac{4l^2+8l+5}{(2l+1)(2l+3)}T_{l+1,l,l+1}+\frac{2l}{2l+1}T_{l-1,l,l+1}\,,$$

$$Q_{l-1}=\frac{2(l+1)}{2l+3}T_{l+1,l,l-1}+\frac{4l^2+1}{(2l-1)(2l+1)}T_{l-1,l,l-1}+\frac{2(l-1)}{2l-1}T_{l-1,l-2,l-1}\,.$$

Before concluding the angular part of the calculation related to multiphoton ionization, it is worth noting that we get some interesting sum

TABLE III. Angular dependence of the partial-wave components of the three-photon ionization transition amplitudes. For each polarization state of the incident light there are four components corresponding, respectively, to the accessible values of the photoelectron angular momentum $L=l\pm 1,\ l\pm 3$. The quantities denoted $T_{\lambda_1,\lambda_2,L}$ represent the radial part of the amplitude: $T_{\lambda_1,\lambda_2,L}=\langle k,L\,|\,rG_{\lambda_2}(E_n+2\omega)rG_{\lambda_1}(E_n+\omega)r\,|\,n,l\rangle$.

Linear polarization

$$M_{l+3}=(-i)^{l+3}e^{i\eta_{l+3}}Y_{l+3,m}(\hat{k})\left(\frac{[(l+1)^2-m^2][(l+2)^2-m^2][(l+3)^2-m^2]}{(2l+1)(2l+3)^2(2l+5)^2(2l+7)}\right)^{1/2}T_{l+1,l+2,l+3}$$

$$\begin{aligned}M_{l+1}=(-i)^{l+1}e^{i\eta_{l+1}}Y_{l+1,m}(\hat{k})\Bigg[&\left(\frac{(l+1)^2-m^2}{(2l+1)(2l+3)}\right)^{3/2}T_{l+1,l,l+1}\\&+\left(\frac{[(l+1)^2-m^2][(l+2)^2-m^2]^2}{(2l+1)(2l+3)^3(2l+5)^2}\right)^{1/2}T_{l+1,l+2,l+1}+\left(\frac{(l^2-m^2)^2[(l+1)^2-m^2]}{(2l-1)^2(2l+1)^3(2l+3)}\right)^{1/2}T_{l-1,l,l+1}\Bigg]\end{aligned}$$

$$\begin{aligned}M_{l-1}=(-i)^{l-1}e^{i\eta_{l-1}}Y_{l-1,m}(\hat{k})\Bigg[&\left(\frac{(l^2-m^2)[(l+1)^2-m^2]^2}{(2l-1)(2l+1)^3(2l+3)^2}\right)^{1/2}T_{l+1,l,l-1}\\&+\left(\frac{l^2-m^2}{(2l-1)(2l+1)}\right)^{3/2}T_{l-1,l,l-1}+\left(\frac{(l^2-m^2)[(l-1)^2-m^2]^2}{(2l-3)^2(2l-1)^3(2l+1)}\right)^{1/2}T_{l-1,l-2,l-1}\Bigg]\end{aligned}$$

$$M_{l-3}=(-i)^{l-3}e^{i\eta_{l-3}}Y_{l-3,m}(\hat{k})\left(\frac{(l^2-m^2)[(l-1)^2-m^2][(l-2)^2-m^2]}{(2l-5)(2l-3)^2(2l-1)^2(2l+1)}\right)^{1/2}T_{l-1,l-2,l-3}$$

Circular polarization

$$M'_{l+3}=(-i)^{l+3}e^{i\eta_{l+3}}Y_{l+3,m+3}(\hat{k})\left(\frac{(l+1+m)(l+2+m)(l+3+m)(l+4+m)(l+5+m)(l+6+m)}{8(2l+1)(2l+3)^2(2l+5)^2(2l+7)}\right)^{1/2}T_{l+1,l+2,l+3}$$

$$\begin{aligned}M'_{l+1}=(-i)^{l+1}e^{i\eta_{l+1}}Y_{l+1,m+3}(\hat{k})&\left(\frac{(l-1-m)(l-m)(l+1+m)(l+2+m)(l+3+m)(l+4+m)}{8(2l+1)(2l+3)}\right)^{1/2}\\\times&\left(\frac{T_{l-1,l,l+1}}{(2l-1)(2l+1)}+\frac{T_{l+1,l+2,l+1}}{(2l+3)(2l+5)}+\frac{T_{l+1,l,l+1}}{(2l+1)(2l+3)}\right)\end{aligned}$$

$$\begin{aligned}M'_{l-1}=(-i)^{l-1}e^{i\eta_{l-1}}Y_{l-1,m+3}(\hat{k})&\left(\frac{(l-3-m)(l-2-m)(l-1-m)(l-m)(l+1+m)(l+2+m)}{8(2l-1)(2l+1)}\right)^{1/2}\\\times&\left(\frac{T_{l-1,l,l-1}}{(2l-1)(2l+1)}+\frac{T_{l-1,l-2,l-1}}{(2l-3)(2l-1)}+\frac{T_{l+1,l,l-1}}{(2l+1)(2l+3)}\right)\end{aligned}$$

$$M'_{l-3}=(-i)^{l-3}e^{i\eta_{l-3}}Y_{l-3,m+3}(\hat{k})\left(\frac{(l-5-m)(l-4-m)(l-3-m)(l-2-m)(l-1-m)(l-m)}{8(2l-5)(2l-3)^2(2l-1)^2(2l+1)}\right)^{1/2}T_{l-1,l-2,l-3}$$

TABLE IV. Explicit expressions of the total three-photon ionization cross section for an atomic $|n,l\rangle$ state according to the polarization state of the incident light.

Linear polarization

$$\begin{aligned}\sigma_{n,l}=4\pi^2\alpha a_0^2\omega\left(\frac{I}{2I_0}\right)^2\frac{1}{105}\Bigg(&\frac{6(l+1)(l+2)(l+3)}{(2l+1)(2l+3)(2l+5)}|T_{l+1,l+2,l+3}|^2+\frac{(l+1)(24l^4+96l^3+150l^2+108l+35)}{(2l+1)^3(2l+3)^2}|T_{l+1,l,l+1}|^2\\&+\frac{2(l+1)(l+2)(6l^2+25l+28)}{(2l+1)(2l+3)^2(2l+5)}|T_{l+1,l+2,l+1}|^2+\frac{2l(l+1)(6l^2-l+2)}{(2l-1)(2l+1)^3}|T_{l-1,l,l+1}|^2\\&+\frac{4l(l+1)(6l^2+13l+9)}{(2l+1)^3(2l+3)}\operatorname{Re}(T_{l+1,l,l+1}T^*_{l-1,l,l+1})+\frac{4(l+1)(l+2)(6l^2+11l+7)}{(2l+1)^2(2l+3)^2}\operatorname{Re}(T_{l+1,l,l+1}T^*_{l+1,l+2,l+1})\\&+\frac{l(24l^4+6l^2+5)}{(2l-1)^2(2l+1)^3}|T_{l-1,l,l-1}|^2+\frac{12l(l+1)(l+2)}{(2l+1)^2(2l+3)}\operatorname{Re}(T_{l-1,l,l+1}T^*_{l+1,l+2,l+1})\\&+\frac{2l(l-1)(6l^3-13l+9)}{(2l-3)(2l-1)^2(2l+1)}|T_{l-1,l-2,l-1}|^2+\frac{2l(l+1)(6l^2+13l+9)}{(2l+1)^3(2l+3)}|T_{l+1,l,l-1}|^2\\&+\frac{4l(l-1)(6l^2+l+2)}{(2l-1)^2(2l+1)^2}\operatorname{Re}(T_{l-1,l,l-1}T^*_{l-1,l-2,l-1})+\frac{4l(l+1)(6l^2-l+2)}{(2l+1)^3(2l-1)}\operatorname{Re}(T_{l-1,l,l-1}T^*_{l+1,l,l-1})\\&+\frac{12l(l-1)(l+1)}{(2l-1)(2l+1)^2}\operatorname{Re}(T_{l-1,l-2,l-1}T^*_{l+1,l,l-1})+\frac{6l(l-1)(l-2)}{(2l-3)(2l-1)(2l+1)}|T_{l-1,l-2,l-3}|^2\Bigg)\end{aligned}$$

Circular polarization

$$\begin{aligned}\sigma'_{n,l}=4\pi^2a_0^2\alpha\omega\left(\frac{I}{2I_0}\right)^2\frac{1}{7}\Bigg(&\frac{(l+1)(l+2)(l+3)}{(2l+1)(2l+3)(2l+5)}|T_{l+1,l+2,l+3}|^2\\&+\frac{l(l+1)(l+2)(2l-1)(2l+5)}{15(2l+1)}\left|\frac{T_{l+1,l+2,l+1}}{(2l+3)(2l+5)}+\frac{T_{l+1,l,l+1}}{(2l+1)(2l+3)}+\frac{T_{l-1,l,l+1}}{(2l-1)(2l+1)}\right|^2\\&+\frac{l(l-1)(l+1)(2l-3)(2l+3)}{15(2l+1)}\left|\frac{T_{l-1,l,l-1}}{(2l-1)(2l+1)}+\frac{T_{l-1,l-2,l-1}}{(2l-3)(2l-1)}+\frac{T_{l+1,l,l-1}}{(2l+1)(2l+3)}\right|^2\\&+\frac{l(l-1)(l-2)}{(2l-3)(2l-1)(2l+1)}|T_{l-1,l-2,l-3}|^2\Bigg)\end{aligned}$$

rules verified by particular products of Wigner 3-j coefficients.[48]

2. *Radial part calculation*

The second part of the calculation deals with the evaluation of the radial part of the amplitude:

$$T_{\lambda_1\lambda_2L}=\langle R_{k,L}|rG_{\lambda_2}(E_n+2\omega)rG_{\lambda_1}(E_n+\omega)r|R_{n,l}\rangle \tag{69}$$

where $R_{n,l}(r)$ is the hydrogenic radial function for the initial bound state $|n,l\rangle$ [Eq. (52″)]; $R_{k,L}$ corresponds to the final continuum state for angular momentum L and is given by

$$R_{k,L}=C_{k,L}e^{-ikr}r^L\,{}_1F_1(ik^{-1}+L+1;2L+2;2ikr)\,, \tag{70}$$

with

$$C_{k,L}=\frac{2^{L+1}}{(2L+1)!}k^L\left(\frac{\prod_{S=1}^{L}(s^2+k^{-2})}{1+\exp(-2\pi/k)}\right)^{1/2},$$

$$\tfrac{1}{2}k^2=3\omega-1/2n^2\,.$$

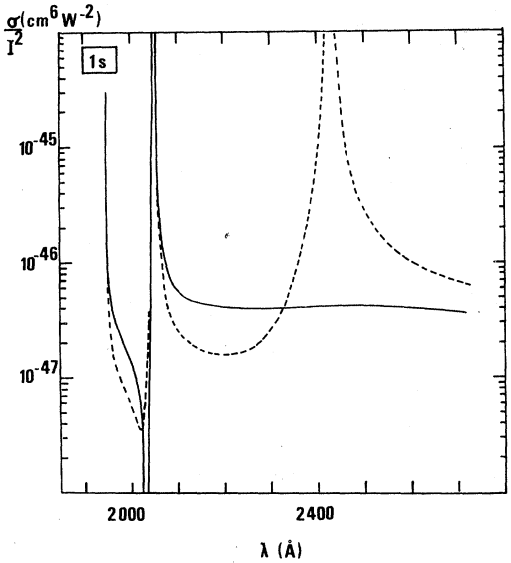

FIG. 3. Three-photon ionization rates from the hydrogenic state 1s. Solid line: circularly polarized light; dashed line: linearly polarized light.

By replacing in Eq. (69) $G_{\lambda_i}(\omega)$ by its Sturmian representation (32), one gets

$$T_{\lambda_1,\lambda_2,L} = C_{n,l}C_{k,L}2^2\left(\prod_{q=1}^{2}\frac{(2x_q)^{2\lambda_q+1}}{[(2\lambda_q+1)!]^2}\right)\sum_{m_1=\lambda_1+1}^{\infty}K_1J(n,l,(1/n)\,|\,m_1,\lambda_1,x_1)$$
$$\times\sum_{m_2=\lambda_2+1}^{\infty}K_2J(m_1,\lambda_1,x_1\,|\,m_2,\lambda_2,x_2)J(m_2,\lambda_2,x_2\,|\,-i/k,L,ik)\,, \tag{71}$$

where

$$x_j=(n^{-2}-2j\omega)^{1/2},\quad K_j=\left(m_j-\frac{1}{x_j}\right)^{-1}\frac{(m_j+\lambda_j)!}{(m_j-\lambda_j-1)!}\,,$$

and

$$J(m,\lambda,x\,|\,m',\lambda',x')=\int_0^\infty dr\,r^{3+\lambda+\lambda'}e^{-r(x+x')}\,{}_1F_1(\lambda+1-m,2\lambda+2;2xr)\,{}_1F_1(\lambda'+1-m',2\lambda'+2;2x'r)\,. \tag{72}$$

Since the coupling of angular momenta imposes the selection rule $\lambda'=\lambda\pm1$, these integrals turn out to be of a well-known form, studied by Gordon.[49] One finally gets

$$J(m,\lambda,x\,|\,m',\lambda',x')=(-1)^{\lambda_<}\frac{(2\lambda_>+1)!}{2x_>^2(x_<-x_>)^{2\lambda_>+2}}\left(\frac{x_>-x_<}{x_>+x_<}\right)^{m_<}\left(\frac{x_<-x_>}{x_>+x_<}\right)^{m_>}$$
$$\times\sum_{q=0}^{2}\binom{2}{q}[x_>(m_>-1+q)-x_<m_<]\left(\frac{x_>-x_<}{x_>+x_<}\right)^q$$
$$\times{}_2F_1(\lambda_>+1-m_>-q,\lambda_<+1-m_<;2\lambda_>;-4x_>x_</(x_>-x_<)^2)\,, \tag{73}$$

where $\lambda_>=\sup(\lambda,\lambda')$, $\lambda_<=\inf(\lambda,\lambda')$, and $m_>,x_>$ $(m_<,x_<)$ are the values of the parameters m,x corresponding to $\lambda_>$ $(\lambda_<)$.

This is nothing but a generalization of Gordon's formula to the case of a transition between two "Sturmian" states. One can verify that by substituting $x\to1/n$, $x'\to1/n'$ and $m\to n$, $m'\to n'$ one recovers Gordon's formula for the radial transition matrix element between two hydrogenic bound states.

It should be noted that the expression of $J(m,\lambda,x\,|\,m',\lambda',x')$ [Eq. (73)] is valid also for complex values of the parameters m (m') or x (x'). However, in this case it is more convenient, for computational purposes, to use equivalent forms, given in terms of real variables, for these expressions. With this aim we have adapted to our problem a method proposed by Karzas and Latter.[50] We obtained in this way the following results:

$$J(m_2,\lambda_2,x_2\,|\,-i/k,\lambda_2-1,ik)=\frac{(2\lambda_2+1)!\exp[k^{-1}(2\tan^{-1}\rho-\pi)]}{2x_2^2k^{2\lambda_2+2}(1+\rho^2)^{m_2+2}}$$
$$\times\sum_{q=0}^{2}(-1)^q\binom{2}{q}(1+\rho^2)^q\,[x_2(m_2+1-q)-1]\,G_{\lambda_2}(\lambda_2+q-1-m_2;1/k;\rho)\,, \tag{74}$$

$$J(m_2,\lambda_2,x_2\,|\,-i/k,\lambda_2+1,ik)=-2\,\frac{(2\lambda_2+1)!\exp[k^{-1}(2\tan^{-1}\rho-\pi)]}{k^{2\lambda_2+6}(1+\rho^2)^{m_2+2}}$$
$$\times\sum_{q=0}^{2}\binom{2}{q}\alpha\beta(1+\rho^2)^q[1-x_2(m_2+1-q)]\,G_{\lambda_2+2}(\lambda_2+q+1-m_2;1/k;\rho)\,, \tag{74'}$$

where

$$\rho=x_2k^{-1},\quad \alpha=\lambda_2+1-m_2(q^2-q-1),$$

$$\beta=\lambda_2+2+(m_2+1)\,q^2-(3m_2+2)\,q+m_2\,,$$

and the real polynomials $G_l(-m;\eta;\rho)$ are defined as follows:

$$G_l(-m;\eta;\rho)=(1-i\rho)^{2m}$$
$$\times{}_2F_1(-m,l-i\eta;2l;-4i\rho(1-i\rho)^{-2})$$
$$=\sum_{S=0}^{2m}b_S\rho^S\,, \tag{75}$$

with

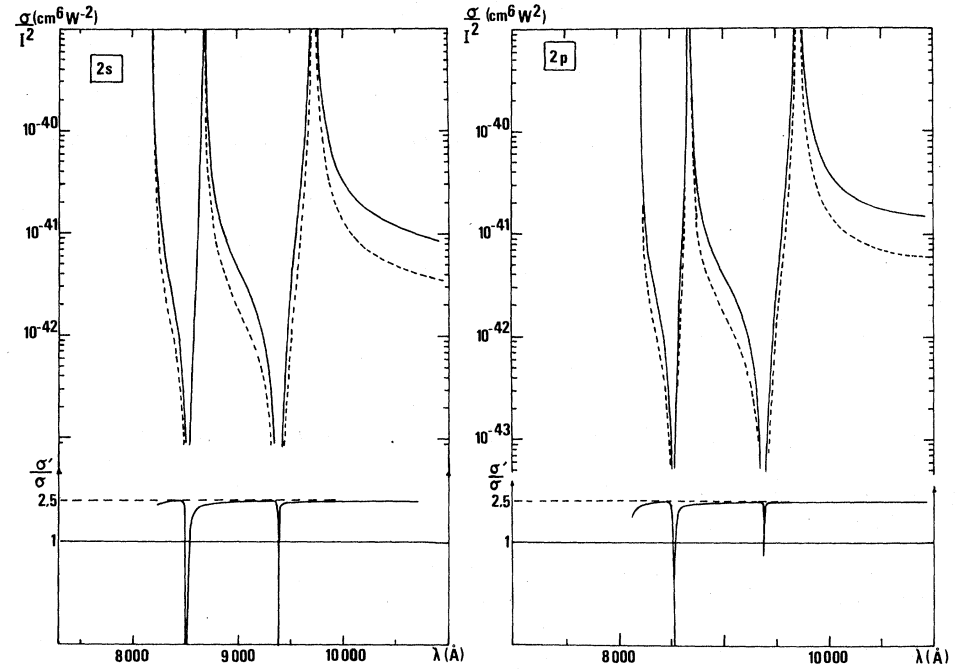

FIG. 4. Polarization effect in three-photon ionization from the hydrogenic state $n=2$. (a) 2s state; (b) 2p state. Solid line: circular polarization; dashed line: linear polarization. In addition we display the variation of the ratio σ'/σ vs wavelength. It appears that this ratio remains often near the theoretically maximum allowed value of $\frac{5}{2}$.

$$b_0=1,\quad b_1=2m\eta/l,$$
$$b_S=-s^{-1}(2l+s+1)^{-1}\times[4\eta(s-1-m)b_{S-1}+(2m+2-s)(2l+2m+1-s)b_{S-2}].$$

As a consequence of these results, one sees that irrespective of whether the functions $J(m,\lambda,x\,|\,m',\lambda',x')$ have imaginary or real arguments x (x') or m (m') they can be evaluated to any desired accuracy owing to their polynomial character. The reduced amplitude $T_{\lambda_1\lambda_2L}$ [Eq. (71)] is thus given as an infinite double series each term of which is expressed in terms of the polynomials $J(m,\lambda,x\,|\,m',\lambda',x')$. We have verified that such series converge quite rapidly. Thus one may choose to cutoff the sum when the ratio of the nth term u_n to the partial sum S_n is lower than an arbitrarily small parameter ϵ: $|u_n/S_n|<\epsilon$.

Our calculations have been performed with $\epsilon=10^{-4}$, but we have checked that the total value of the sums were not affected, within an accuracy of one part in one thousand, if one chooses $\epsilon=10^{-6}$ or $\epsilon=10^{-8}$.

In this way we easily get the transition amplitude and then the three-photon ionization cross section from any given bound state. These results have allowed us to study in greater detail the polarization effect for states 1s through 3d. Our results are summarized by the curves displayed in Figs. 3–5.

For the lowest-lying states 1s and 2s our results coincide with those of Gontier and Trahin.[51] For more excited states our results confirm the overall advantage of circular polarization, since the ratio σ'/σ remains greater than unity for large domains of frequency of the incident light.

We can mention also that values of the cross sections at the three-photon ionization threshold may be obtained easily from the above-given expressions. As a matter of fact, this task may be achieved by taking the limiting value of the function $J(m_2,\lambda_2,x_2\,|-i/k,L,ik)$ for $k\to 0$ (corresponding to zero kinetic energy of the photoelectron). This procedure corresponds to a confluence transformation of the Gauss hypergeometric functions ${}_2F_1$ involved in the expression of $J(m_2,\lambda_2,x_2\,|-i/k,L,ik)$ [Eq. (73)]. One gets eventually

$$\lim_{k\to 0}[J(m_2,\lambda_2,x_2\,|-i/k,\lambda_2-1,ik)]$$
$$=(-1)^{m_2-\lambda_2-1}\frac{e^{-2/x_2}(2\lambda_2+1)!}{2x_2^{2\lambda_2+4}}\sum_{q=0}^{2}\binom{2}{q}[x_2(m_2+1-q)-1]\,{}_1F_1(\lambda_2+q-m_2-1;2\lambda_2;4x_2^{-1}),\qquad(76)$$

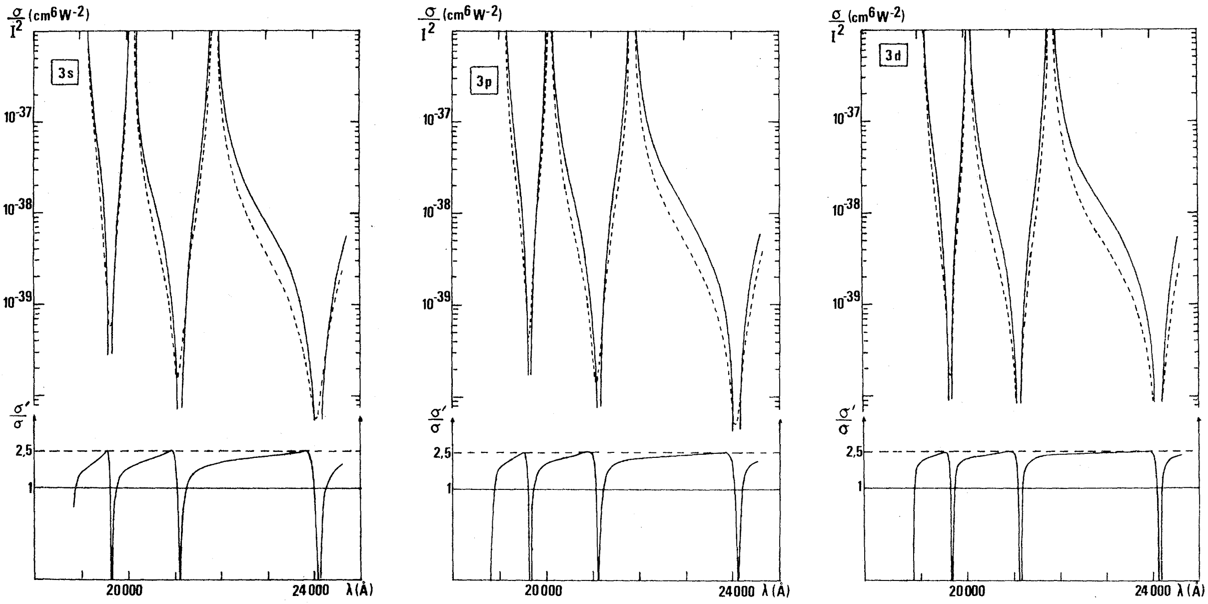

FIG. 5. Polarization effect in three-photon ionization rates from the hydrogenic state $n=3$. (a) $3s$ state; (b) $3p$ state; (c) $3d$ state. Solid line: circular polarization; dashed line: linear polarization. From the variation of the ratio σ'/σ vs wavelength it may be verified that for a given value of the wavelength the magnitude of this ratio increases with the angular momentum of the state considered.

$$\lim_{k\to 0}\left[J(m_2,\lambda_2,x_2\,|\,-i/k,\lambda_2+1,ik)\right]$$

$$=(-1)^{m_2+\lambda_2}\frac{2e^{-2/x_2}(2\lambda_2+1)!}{x_2^{2\lambda_2+6}}\sum_{q=0}^{2}\binom{2}{q}\alpha\beta\left[1-x_2(m_2+1-q)\right]\,{}_1F_1(\lambda_2+q+1-m_2;2\lambda_2;4x_2^{-1})\,, \qquad (76')$$

where ${}_1F_1(a;c;z)$ denotes the confluent hypergeometric function.

Substitution into Eq. (71) then yields the desired result.[52] We list in Table V our values for states $1s$ through $6d$.

B. Higher-order calculations

The generalization of the above-described formalism to higher-order calculations is quite straightforward. However, we shall not present expressions of the total cross sections, as in Tables III and IV, since they are too cumbersome. Instead the angular part of the amplitude was computed entirely numerically. On the other hand, the radial part (corresponding to the reduced amplitudes T) was evaluated in a way exactly similar to that we described in Sec. V A. The increase in the number of involved photons corresponds to the insertion in the expression of the reduced amplitude T of new infinite sums extended to the whole "Sturmian" spectrum of the H atom. As a rule, an Nth-order reduced amplitude corresponding to the ionization by N photons of energy ω contains $N-1$ such sums:

$$T_{\lambda_1\lambda_2\cdots\lambda_{N-1}L}=C_{n,l}C_{k,L}2^{N-1}\left(\prod_{q=1}^{N-1}\frac{(2x_q)^{2\lambda_q+1}}{[(2\lambda_q+1)!]^2}\right)$$

$$\times\sum_{m_1=\lambda_1+1}^{\infty}K_1J(n,l,(1/n)\,|\,m_1,\lambda_1,x_1)\sum_{m_2=\lambda_2+1}^{\infty}K_2J(m_1,\lambda_1,x_1\,|\,m_2,\lambda_2,x_2)\cdots$$

$$\times\sum_{m_{N-1}=\lambda_{N-1}+1}^{\infty}K_{N-1}J(m_{N-2},\lambda_{N-2},x_{N-2}\,|\,m_{N-1},\lambda_{N-1},x_{N-1})J(m_{N-1},\lambda_{N-1},x_{N-1}\,|\,-i/k,L,ik)\,, \qquad (77)$$

where the notations are identical to those of Sec. VA, Eq. (71).

The numerical computation of such multiple sums does not present major difficulties since, as we have shown, each term of the series may be evaluated to any desired accuracy.

We have tested our results by comparing them with those previously obtained by Gontier and Trahin[51] for 1*s* and 2*s* states, and for processes involving up to $N=8$ photons. This verification provides an independent check of the numerical results obtained within the framework of the perturbation theory.

On the other hand, we have used this formalism in order to study the role of the initial excited state on the polarization effect. We present some of our results for the six-photon ionization of the states 3*s* through 3*d* (Fig. 6). The results confirm the generally accepted analysis according to which the linear polarization becomes more and more advantageous as the number of photons involved increases.[51,52] Results at the threshold may be obtained in the same way by generalizing the formulas (76) (see Table V).

Moreover, it appears that this method may be used also for even higher-order calculations. So we have evaluated the 12-photon ionization cross section from the 1*s* state by circularly polarized neodymium laser light ($\lambda = 10\,600$ Å):

$$\sigma_{(N=12)} = 1.4\times10^{-174}\,\text{cm}^{24}\,\text{W}^{-11}\,.$$

This result has been obtained independently by

TABLE V. Polarization effect in N-photon ionization transition rates at threshold, i.e., for a zero kinetic energy of the photoelectron. We give data for the atomic states 1*S* to 6*D* included and transitions involving $N=3,5,6,7$ photons. For fourth- and eighth-order transitions the cross sections do not remain finite since the process becomes resonant (at least with linearly polarized light). The format $A(n)$ means $A\times10^n$.

State $\backslash N$	3	5	6	7
1*S*	$\frac{3.50(-47)}{6.10(-47)}=5.73(-1)$	$\frac{1.83(-76)}{6.93(-75)}=2.64(-2)$	$\frac{4.49(-91)}{2.54(-88)}=1.77(-3)$	$\frac{1.14(-105)}{1.33(-101)}=8.53(-5)$
2*S*	$\frac{8.60(-42)}{3.60(-42)}=2.39$	$\frac{4.45(-67)}{6.63(-66)}=6.71(-2)$	$\frac{5.45(-79)}{1.16(-75)}=4.70(-4)$	$\frac{7.74(-91)}{2.04(-89)}=3.80(-2)$
2*P*	$\frac{1.48(-41)}{6.01(-42)}=2.46$	$\frac{3.86(-66)}{9.20(-66)}=4.19(-1)$	$\frac{2.60(-78)}{9.88(-76)}=2.63(-3)$	$\frac{1.40(-90)}{3.49(-89)}=4.02(-2)$
3*S*	$\frac{4.17(-39)}{3.32(-39)}=1.79$	$\frac{8.65(-60)}{6.80(-59)}=1.27(-1)$	$\frac{1.27(-72)}{5.55(-71)}=2.30(-2)$	$\frac{2.80(-79)}{4.84(-77)}=5.78(-3)$
3*P*	$\frac{6.22(-39)}{3.00(-39)}=2.07$	$\frac{1.41(-60)}{7.69(-59)}=1.83(-2)$	$\frac{9.87(-72)}{7.97(-71)}=1.24(-1)$	$\frac{7.93(-80)}{5.76(-77)}=1.38(-3)$
3*D*	$\frac{6.45(-39)}{2.82(-39)}=2.28$	$\frac{1.58(-59)}{6.18(-59)}=2.55(-1)$	$\frac{1.34(-70)}{8.75(-71)}=1.53$	$\frac{1.51(-80)}{4.34(-77)}=3.49(-4)$
4*S*	$\frac{1.63(-34)}{8.10(-35)}=1.68$	$\frac{1.23(-54)}{2.99(-55)}=4.12$	$\frac{7.53(-63)}{1.86(-62)}=4.05(-1)$	$\frac{5.38(-75)}{5.94(-75)}=9.07(-1)$
4*P*	$\frac{2.00(-34)}{1.03(-34)}=1.94$	$\frac{2.93(-54)}{4.87(-55)}=6.01$	$\frac{7.95(-63)}{2.23(-62)}=3.57(-1)$	$\frac{4.68(-75)}{8.24(-75)}=5.67(-1)$
4*D*	$\frac{2.69(-34)}{1.23(-34)}=2.18$	$\frac{6.43(-54)}{8.84(-55)}=7.28$	$\frac{3.80(-63)}{2.39(-62)}=1.59(-1)$	$\frac{4.32(-76)}{1.09(-74)}=3.98(-2)$
5*S*	$\frac{2.40(-35)}{1.42(-35)}=1.70$	$\frac{4.38(-51)}{1.37(-52)}=3.20$	$\frac{5.75(-63)}{2.41(-61)}=2.38(-2)$	$\frac{1.70(-69)}{4.40(-69)}=3.26(-1)$
5*P*	$\frac{3.40(-35)}{1.78(-35)}=1.90$	$\frac{7.89(-51)}{1.87(-51)}=4.21$	$\frac{1.42(-63)}{2.57(-61)}=5.53(-3)$	$\frac{2.31(-69)}{4.94(-69)}=4.67(-1)$
5*D*	$\frac{4.86(-35)}{2.29(-35)}=2.12$	$\frac{1.48(-50)}{2.76(-51)}=5.34$	$\frac{1.53(-62)}{2.59(-61)}=5.90(-2)$	$\frac{2.50(-69)}{5.40(-69)}=4.64(-1)$
6*S*	$\frac{2.98(-35)}{2.99(-35)}=9.95(-1)$	$\frac{4.36(-50)}{1.31(-50)}=3.32$	$\frac{6.33(-58)}{9.57(-59)}=6.61$	$\frac{5.16(-63)}{2.19(-62)}=2.36(-1)$
6*P*	$\frac{3.89(-35)}{3.20(-35)}=1.21$	$\frac{7.52(-50)}{1.78(-50)}=4.23$	$\frac{1.15(-57)}{1.38(-58)}=8.37$	$\frac{7.13(-63)}{2.35(-62)}=3.03(-1)$
6*D*	$\frac{5.40(-35)}{3.47(-35)}=1.55$	$\frac{1.42(-49)}{2.68(-50)}=5.29$	$\frac{2.51(-57)}{2.50(-58)}=10.1$	$\frac{9.21(-63)}{2.51(-62)}=3.66(-1)$

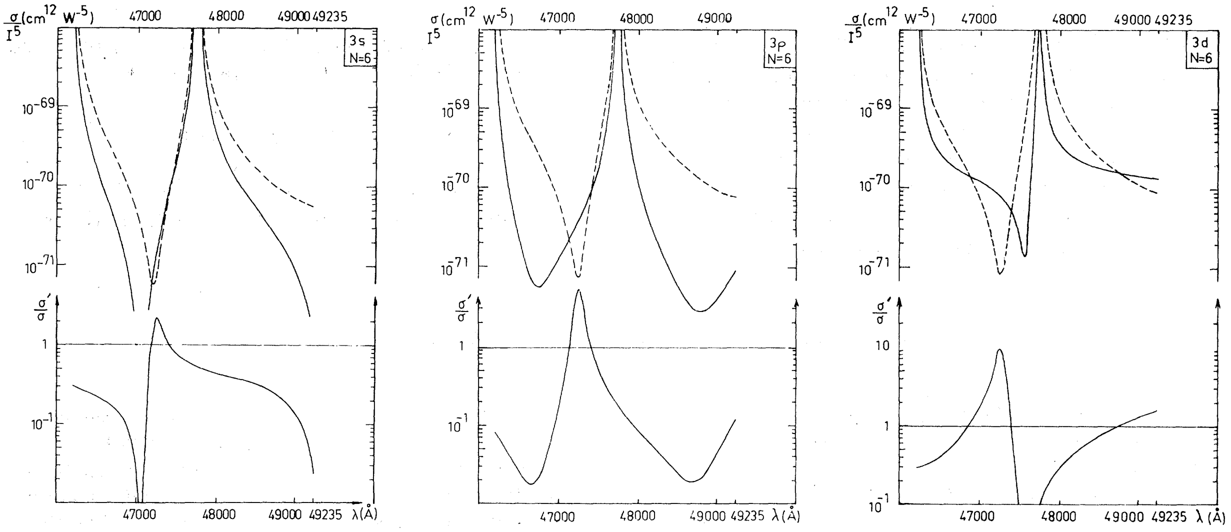

FIG. 6. Influence of the initial atomic sublevel on the polarization effect for six-photon ionization rates from the hydrogenic state $n=3$. (a) 3*s* state; (b) 3*p* state; (c) 3*d* state. Solid line: circular polarization; dashed line: linear polarization. The predominance of the linearly polarized light is reduced as the angular momentum increases.

Karule[24] who used a similar technique.

Before summarizing our results in Sec. VI we wish to emphasize that the Sturmian representation of the CGF seems to us the best adapted for this type of calculation. As a matter of fact, any amplitude corresponding to a higher-order transition may be expressed in a form similar to that given in Eq. (77), whatever the considered initial and final hydrogenic states. Moreover, as we previously pointed out, the numerical calculation consists of summing up infinite (convergent!) series of polynomials, which consequently can be computed to any desired accuracy. These features are characteristic of the Sturmian analysis and strongly facilitate the programming work, since it does not require sophisticated numerical methods. For instance, we think that the "Sturmian" technique compares very favorably with Dalgarno's method. As a last comment, we strongly recommend the use of the Sturmian representation to future CGF users!

VI. SUMMARY

In this paper we presented some analytical and numerical results concerning various higher-order processes occurring when a nonrelativistic hydrogenic system interacts with an electromagnetic radiation field. It should be noted, more precisely, that we described these processes within the common framework of the time-dependent perturbation theory. Our contribution may be considered as twofold.

(i) We derive analytical expressions of some second-order amplitudes. So we studied the retardation effect in two-photon bound-bound transitions, and we evaluated the nonrelativistic part of the self-energy contribution (Bethe logarithm) to the overall Lamb shift for the levels 1*S*–4*P*. The analytical expressions obtained allowed us to get very accurate numerical values of the Bethe logarithm.

We also studied higher-order processes and, in particular, we have been able to compute transition rates for multiphoton ionization from any given hydrogenic state. Our results provide an independent check of those obtained by Gontier and Trahin,[51] who used a different method.

(ii) From a different point of view our work may be considered as an investigation of the respective advantages offered by the various CGF representations. For instance, if we restrict ourselves to second-order amplitudes and if one needs analytical expressions which easily lend themselves to formal manipulations such as analytic continuation, etc., it seems preferable to use integral representations of the CGF. Thus for calculations performed within the dipole approximation Hostler's representation [Eq. (18)] is convenient; see Sec. IV. On the contrary, if one has to include retardation effects, one may use either Schwinger's four-dimensional form [Eq. (13), see Sec. III], or Hostler's expression, see Ref. 20.

On the other hand, it should be noted that without the above-mentioned conditions the Sturmian representation [Eq. (32)] seems strongly recommendable in view of its ease of use particularly for higher-order calculations. As a matter of fact, higher-order amplitudes are then given in terms of multiples series, the evaluation of which does not require sophisticated numerical methods.

Note added in proof. Very recently Khristenko and Vetchinkin[55] used also the Sturmian representation of the CGF in two- and three-photon ionization calculations. The results they obtained are in good agreement with our own data.

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