Stimulated radiative corrections in hydrogen in the presence of a strong laser field

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(Received 6 May 1982)

The stimulated radiative corrections (or ac-Stark shift, or light shift . . .) of the energy levels of a hydrogenic atom irradiated by an intense, nonresonant, monomode laser field, are evaluated within the framework of the time-dependent perturbation theory, up to and including fourth order. The calculation is performed by using a Sturmian representation of the Coulomb Green's function. Whenever it is possible, comparison is made with other results, and several limiting cases are considered. If the laser frequency $\omega \to 0$, one recovers the results of the dc-Stark effect. If 2ω becomes larger than the ionization energy of the atomic state considered, the fourth-order level shift acquires an imaginary part which may be connected, via an extension of the optical theorem, to the two-photon ionization cross section. We point out also the difficulties encountered, when trying to get sensible estimates from partial summation of the infinite sums entering the perturbative expression of the fourth-order amplitudes. Finally, the order of magnitude of these various corrections is discussed.

I. INTRODUCTION

Following the early works on multiphoton processes it became apparent that the modifications of the atomic spectrum, in the presence of an intense laser field, play an essential role in explaining the observed behavior of multiphoton cross sections. As a consequence, in the later years, the theme "atoms in strong fields" has been extremely popular among atomic and laser physicists. In the presence of the strong external field the atomic levels are displaced, i.e., shifted and broadened, respectively.[1] The magnitude of these corrections can be obtained, at least in principle, from the location into the complex plane, of the poles of the resolvent $(z-H)^{-1}$ associated to the Hamiltonian H of the coupled system atom and field. As, in general, this task cannot be achieved exactly, one has to resort to approximation methods which may be, broadly speaking, classified into two categories according to whether they are perturbative or not. At very high intensity, i.e., when the laser intensity I becomes comparable to a characteristic atomic field strength intensity I_0, nonperturbative approaches become relevant. Let us recall that these methods either rely on unitary transformations,[2,3] or assume the atomic potential is a small perturbation with respect to the radiation field.[4,5] However, in addition to the fundamental difficulties raised by some of them,[6] the quantitative predictions of such theories have not been verified yet.

Another method, which has proven especially attractive for describing resonant or quasiresonant processes, i.e., when a few-level atomic model is well adapted, is the so-called "dressed-atom" approach which can be considered within either a fully quantum,[7] or semiclassical (Floquet[8]) framework. Note that this latter method has recently been successfully extended to more realistic atomic models, including in particular the continuous spectrum, and applied to the quantitative description of the ionization of atomic hydrogen in a very intense field.[9,10]

In nonresonant conditions, standard perturbative calculations provide nevertheless reliable estimates for interpreting most multiphoton experiments.[11] Moreover, it can be shown that the perturbative series itself may be formally resummed and reexpressed in closed form, in terms of operator-continued-fractions expansions, providing in some sense an analytic continuation of the series expansion beyond its domain of validity.[12–19] Although this had been recognized for a long time,[13] there has recently been a considerable body of work on the subject.[10,14–19] Note that it would be extremely interesting to compare the predictions of such "extended" perturbative theories with nonperturbative ones in the limit of very intense fields. Unfortunately, practical computations performed on realistic models meet with considerable difficulties, which has impeded making the connection between the two approaches.

Up to now, most perturbative calculations of these stimulated radiative corrections (or ac-Stark shifts, or light shifts, or dynamic polarizability,...)[1] were limited to the second order, i.e., the first nonvanishing term in the expansion.[20–24] Now, the advent of a variety of laser-aided high-resolution spectroscopies has permitted accurate measurements of such level displacements.[20,24] Recently, measurements have also been performed in atomic hydrogen, and the possibility of observing higher-order effects has arisen.[25] Consequently, it seemed of interest to carry out accurate higher-order perturbative calculations which could provide some reference marks for future discussions. More precisely, we present in this paper a perturbative calculation of the stimulated radiative corrections, including the fourth-order contributions, experienced by a nonrelativistic H atom irradiated by a nonresonant monomode laser. As by-products, we have gotten several interesting results related to the behavior of atoms in strong fields.

The organization of the paper is as follows. Section II is dedicated to the explicit derivation, within the framework of the time-dependent perturbation theory, of the various terms contributing, order by order, to the energy shifts. In Sec. III we present the computational method we have set up. By using the Sturmian representation of the Coulomb Green's function, we have been able to express each high-order perturbative amplitude as a series expansion in one (or several) variables. In order to improve the efficiency of the computation we have introduced Padé-related (ϵ-algorithm) methods for accelerating the convergence of the series. By the way, we have shown the usefulness of those methods when applied to the summation of multiple series.[26] Our numerical results are presented in Sec. IV, in which several limiting cases are also considered. In particular, when the laser frequency $\omega \rightarrow 0$ connection is made with the dc-Stark effect. On the contrary, as ω increases and two-photon ionization becomes possible one gets a generalization of the optical theorem, extended to multiphoton forward scattering. We show also that approximate computations, performed by retaining only a limited subset of discrete atomic states, i.e., on omitting the continuous spectrum contribution, can lead to misleading results in fourth-order calculations. This behavior is in sharp contrast with second-order calculations in which sensible estimates can be obtained even if truncated atomic basis sets are used.[27] Finally we conclude, in Sec. V, with a brief discussion of our results.

II. PERTURBATIVE EXPANSION AND FOURTH-ORDER LEVEL SHIFT

Most of the material of this section is directly inspired from the standard QED formalism as exposed, for instance, in Heitler's treatise.[28] A more specific account may be found in Ref. 29. One considers hereafter a nonrelativistic atom irradiated by an intense monomode electromagnetic field. The Hamiltonian H of the system is accordingly written

$$H = H_0 + V , \tag{1}$$

where $H_0 = H_{\text{at}} + H_{\text{rad}}$. Here H_{at} is the atomic Hamiltonian with stationary eigenstates $|n\rangle$ and eigenenergies E_n: $H_{\text{at}}|n\rangle = E_n|n\rangle$. H_{rad} is the Hamiltonian of the electromagnetic field. In the case of a monomode laser of frequency ω, one has

$$H_{\text{rad}}|N\rangle = (N + \tfrac{1}{2})\hbar\omega|N\rangle , \tag{2}$$

where N is the occupation number of the laser mode. As $N \gg 1$ one has

$$H_{\text{rad}}|N\rangle \simeq N\hbar\omega|N\rangle . \tag{3}$$

Within the dipole approximation, it is convenient to write the interaction Hamiltonian V in the electric dipole form

$$V = -e\vec{E}\cdot\vec{r} , \tag{4}$$

where the electric field operator $\vec{E}$ is, in Gaussian unrationalized units,

$$\vec{E} = i\left[\frac{2\pi\hbar\omega}{\mathcal{V}}\right]^{1/2}(a\vec{\epsilon} - a^{\dagger}\vec{\epsilon}^{*}) . \tag{5}$$

Here $\mathcal{V}$ is the normalization volume, a and $a^{\dagger}$ are the usual annihilation and creation operators, and $\vec{\epsilon}$ is a unit polarization vector. Note that, for computational purposes, we have preferred to use the $\vec{E}\cdot\vec{r}$ electric dipole form of the interaction Hamiltonian, instead of the $\vec{A}\cdot\vec{p} + \vec{A}^2$ Coulomb gauge representation. Although the equivalence between the two representations may be inferred from the general invariance properties of the S matrix under the corresponding gauge transformation,[30] several papers have recently been published on that matter.[31] This led us to verify that, in the problem considered here, strictly equivalent results are obtained by using either of the two forms.[32] Note also that, if one uses the $\vec{A}\cdot\vec{p} + \vec{A}^2$ form, the $\vec{A}^2$ term has to be consistently included at each step of the perturbation expansion and gives rise to nonvanishing contributions. Although this fact is well known in the case of two-photon elastic (Rayleigh) scattering, it does

not seem to have been recognized in the case of higher-order processes.[33]

The radiative shift of the atomic level $|n\rangle$, in the presence of the monomode laser field, may be obtained by expressing the probability amplitude $A_i(t)$, $t \geq 0$, of the state $|i\rangle = |n\rangle \otimes |N\rangle$, as a contour integral in the complex E plane:

$$A_i(t) = \lim_{\epsilon \to 0} \frac{1}{2\pi i} \oint dE\, e^{-iEt/\hbar} G_i(E - i\epsilon) , \quad (6)$$

where

$$G_i(z) = \langle i | (z - H_0 - V)^{-1} | i \rangle$$

is the diagonal matrix element of the resolvent $G(z)$ associated to the Hamiltonian $H = H_0 + V$. Then, it is convenient to rewrite

$$G_i(z) = [z - E_i - R_i(z)]^{-1} ,$$

where $R_i(z)$ is the diagonal element of the so-called level-shift operator $R(z)$, which, in turn, is the solution of the equation[34–36]

$$R(z)P_i = \left[V + V \frac{Q_i}{z - H_0} R(z) \right] P_i \quad (7)$$

with $P_i = |i\rangle\langle i|$ and $Q_i = 1 - P_i$. $R_i(z)$ is usually perturbatively expanded in powers of V, and one gets in our case

$$R_i(z) = P_i \left[V \frac{Q_i}{z - H_0} V + V \frac{Q_i}{z - H_0} V \frac{Q_i}{z - H_0} V \frac{Q_i}{z - H_0} V + \cdots \right] P_i \quad (8a)$$

$$= R_i^{(2)}(z) + R_i^{(4)}(z) + \cdots . \quad (8b)$$

At lower intensities and in nonresonant cases,[37] one can limit oneself to the second-order contribution, which, after replacing z by E_i, reduces to

$$\Delta E_i^{(2)} = R_i^{(2)}(E_i) = \mathsf{S}_i \frac{|V_{1i}|^2}{E_i - E_1} , \quad (9)$$

where $V_{ij} = \langle i | V | j \rangle$, $H_0 | j \rangle = E_j | j \rangle$, and the generalized sum S_j runs over the complete (discrete + continuous) spectrum of H_0, the state $|i\rangle$ being consistently excluded. Note that, after substituting $z = E_i$, the small imaginary quantity $i\epsilon$ has disappeared in the denominator.

If one wishes to obtain the fourth-order contribution it is not enough to make the replacement $z = E_i$ into $R_i^{(4)}(z)$: One has, in addition, to take into account the fact that $R_i^{(2)}(z)$ is z dependent. This can be done in several ways. For instance, one can use a Taylor expansion of $R_i^{(2)}(z)$ for $z \simeq E_i$.

$$R_i^{(2)}(z) = R_i^{(2)}(E_i) + (z - E_i) \frac{dR_i^{(2)}(E_i)}{dE_i} + \cdots .$$

This leads to the following expansion for G_i, correct up to fourth order:

$$G_i(z) \simeq \left\langle i \left| \left[1 - \frac{dR_i^{(2)}}{dE_i} \right]^{-1} \left[z - E_i - \frac{R_i^{(2)}(E_i) + R_i^{(4)}(E_i) + \cdots}{1 - dR_i^{(2)}/dE_i} \right]^{-1} \right| i \right\rangle . \quad (10)$$

Since

$$(1 - dR_i^{(2)}/dE_i)^{-1} \simeq 1 + dR_i^{(2)}/dE_i + \cdots ,$$

one easily verifies that the total fourth-order contribution to the level shift becomes

$$\Delta E_i^{(4)} = \mathsf{S}_{1,2,3} \frac{V_{i3} V_{32} V_{21} V_{1i}}{(E_i - E_3)(E_i - E_2)(E_i - E_1)} + \Delta E_i^{(2)} \frac{d\Delta E_i^{(2)}}{dE_i} . \quad (11)$$

This result could have been obtained more directly, without replacing in $G_i(z)$, by using a Lagrange expansion of $R_i(z)$ in the neighborhood of $z = E_i$ (Ref. 38):

$$R_i(z)=R_i(E_i)+\sum_{n=1}^{\infty}\frac{1}{n!}\frac{d^{n-1}}{dE_i^{n-1}}[R_i'(E_i)R_i^n(E_i)]\ , \tag{12}$$

which leads in a straightforward fashion to (9) and (11). It is interesting to note the formal equivalence of these results with the usual Rayleigh-Schrödinger expressions of the second- and fourth-order energy corrections in stationary perturbation theory.[39]

In the foregoing we have used expansions (Taylor or Lagrange) valid in the neighborhood of $z=E_i$. One can show that, if one considers the expansion of G_i in terms of R_i in the integral (6),

$$A_i(t)=\lim_{\epsilon\to 0}\frac{1}{2\pi i}\oint dE\, e^{-iEt/\hbar}\left[\frac{1}{E-E_i-i\epsilon}+\frac{1}{E-E_i-i\epsilon}R_i(E-i\epsilon)\frac{1}{E-E_i-i\epsilon}+\cdots\right], \tag{13}$$

this is equivalent to neglecting the contributions of the poles $z\neq E_i$ contained in $R_i(z)$. This is certainly not correct in a time-dependent approach, since every pole does contribute to the probability amplitude $A_i(t)$. In fact, if those contributions are properly taken into account, one gets[40]

$$\Delta E_i^{(2)}(t)=\mathop{S}_{1} V_{i1}\frac{1-e^{i(E_i-E_1)t/\hbar}}{E_i-E_1}V_{1i}\ , \tag{14}$$

and a similar expression for $\Delta E_i^{(4)}$. Now, for large values of t, one has[28]

$$\lim_{t\to\infty}\frac{1-e^{izt}}{z}=\frac{P}{z}-i\pi\delta(z)=\lim_{\eta\to 0}\frac{1}{z+i\eta}\ . \tag{15}$$

Thus, by taking into account the poles of $G_i(z)$ one obtains, *in the limit of large times t,*

$$\Delta E_i^{(2)}=\lim_{\eta\to 0}\mathop{S}_{1}\frac{|V_{i1}|^2}{E_i-E_1+i\eta}\ , \tag{16a}$$

$$\Delta E_i^{(4)}=\lim_{\eta\to 0}\mathop{S}_{1,2,3}\frac{V_{i3}V_{32}V_{21}V_{1i}}{(E_i-E_3+i\eta)(E_i-E_2+i\eta)(E_i-E_1+i\eta)}+\Delta E_i^{(2)}\frac{d\Delta E_i^{(2)}}{dE_i}\ . \tag{16b}$$

Note the presence of the small imaginary part $i\eta$ in the denominators which permits us to treat the case of transitions involving the continuous spectrum.

Since the state of the field can be considered as unaffected by the presence of the small atomic system, it appears that these corrections are, in fact, the radiative shifts of the atomic state $|n\rangle$: $\Delta E_i=\Delta E_n$. This assumption is consistent with the condition $N\gg 1$.

Then, by factorizing out the field-dependent quantities, one easily gets the intensity dependence of the second-order atomic level shift $\Delta E_n^{(2)}$. For instance, by introducing the laser intensity I and the characteristic atomic field strength intensity

$$I_0=\frac{c}{4\pi}|E_0|^2=7.016\times 10^{16}\ \mathrm{W/cm^2}\ ,$$

where $E_0=e/a_0^2$ is the atomic unit of electric field strength intensity, one has

$$\Delta E_n^{(2)}=\frac{I}{2I_0}\tau_n^{(2)}(\omega)\ . \tag{17}$$

Here $\tau_n^{(2)}(\omega)$ is an atomic matrix element which, in the case of a laser beam linearly polarized along the Oz axis, reads

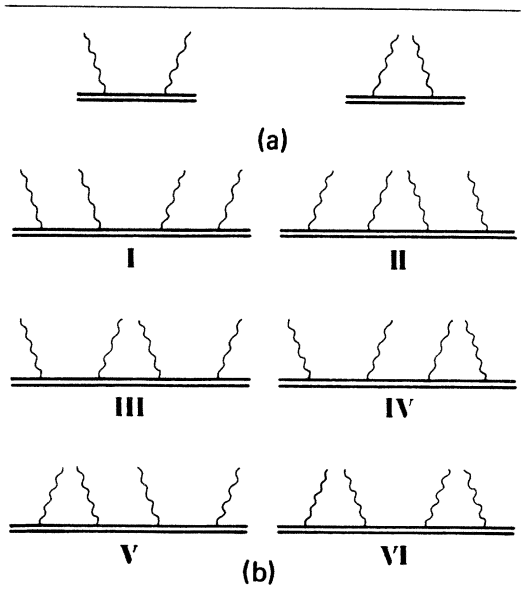

FIG. 1. Diagrams contributing to the second-order (a) and fourth-order (b) stimulated radiative corrections in the electric dipole gauge. The diagrams denoted I and II (b) are referred to as proper diagrams in the text. Those numbered III to VI, in which the system can return in the initial state after two interactions, are referred to as improper diagrams.

$$\tau_n^{(2)}(\omega)=\sum_{n_1}|(z)_{n,n_1}|^2\left[\frac{1}{E_n-E_{n_1}+\hbar\omega}+\frac{1}{E_n-E_{n_1}-\hbar\omega}\right], \tag{18}$$

where $(z)_{n,n_1}=\langle n\,|\,z\,|\,n_1\rangle$ and $|\,n_1\rangle=|\,nlm\,\rangle$ and $|\,n_1\rangle=|\,n_1l_1m_1\,\rangle$ are the atomic kets in the initial and intermediate virtual states, respectively. The infinite sum runs over the complete (discrete + continuous) atomic spectrum and we have dropped, for notational convenience, the small imaginary part $i\eta$ in the denominators. Note the two terms inside the bracket may be associated, respectively, to the two diagrams displayed in Fig. 1(a).

As this point will be considered later, it is interesting to express $\Delta E_n^{(2)}$ in terms of the average of the square of the electric field operator, Eq. (4). One has[41]

$$\overline{E^2}=\langle N\,|\,E^2\,|\,N\rangle=\frac{4\pi N\hbar\omega}{\mathcal{V}}=\frac{4\pi}{c}I \tag{19}$$

and

$$\Delta E_n^{(2)}=\frac{\overline{E^2}}{2E_0^2}\tau_n^{(2)}(\omega)\,. \tag{20}$$

The field dependence of $\Delta E_n^{(4)}$ may be similarly expressed in terms either of the laser intensity or of the average value of the fourth power of the electric field operator, and one has

$$\Delta E_n^{(4)}=\left[\frac{I}{2I_0}\right]^2\tau_n^{(4)}(\omega)=\frac{1}{6}\frac{\overline{E^4}}{E_0^4}\tau_n^{(4)}(\omega)\,. \tag{21}$$

Here $\tau_n^{(4)}(\omega)$ is an atomic fourth-order matrix element, which, in the case of a laser beam linearly polarized along the Oz axis, reads explicitly

$$\begin{aligned}\tau_n^{(4)}(\omega)=&\sum_{n_3}\sum_{n_2}\sum_{n_1}(z)_{nn_3}(z)_{n_3n_2}(z)_{n_2n_1}(z)_{n_1n}\left[\frac{1}{(E_n-E_{n_3}+\hbar\omega)(E_n-E_{n_2}+2\hbar\omega)(E_n-E_{n_1}+\hbar\omega)}\right.\\&\left.+\frac{1}{(E_n-E_{n_3}-\hbar\omega)(E_n-E_{n_2}-2\hbar\omega)(E_n-E_{n_1}-\hbar\omega)}\right]\\&+\sum_{n_3}\sum_{n_2\neq n}\sum_{n_1}\frac{(z)_{nn_3}(z)_{n_3n_2}(z)_{n_2n_1}(z)_{n_1n}}{E_n-E_{n_2}}\left[\frac{1}{(E_n-E_{n_3}+\hbar\omega)(E_n-E_{n_1}+\hbar\omega)}+\frac{2}{(E_n-E_{n_3}-\hbar\omega)(E_n-E_{n_1}+\hbar\omega)}\right.\\&\left.+\frac{1}{(E_n-E_{n_3}-\hbar\omega)(E_n-E_{n_1}-\hbar\omega)}\right]\\&-\tau_n^{(2)}(\omega)\sum_{n_1}|(z)_{nn_1}|^2\left[\frac{1}{(E_n-E_{n_1}+\hbar\omega)^2}+\frac{1}{(E_n-E_{n_1}-\hbar\omega)^2}\right].\end{aligned} \tag{22}$$

The first term corresponds to the contribution of the so-called proper diagrams,[14,36] numbered I and II in Fig. 1(b). Other terms may be associated to the four improper diagrams,[14,36] corresponding to processes in which the system can return in the initial state after two interactions with the field. These diagrams are numbered III to VI in Fig. 1(b). The evaluation of the infinite sums which appear in those expressions will be presented in the next section.

III. COMPUTATIONAL PROCEDURE

In the case of a nonrelativistic hydrogenic atom, the infinite sums, entering Eqs. (18) and (22), may be evaluated exactly, either by solving a system of inhomogeneous differential equations,[42] or by using compact representations of the Coulomb Green's function.[43–45] Here we have used the latter technique and more precisely the so-called Sturmian representation,[45] which has proven to be extremely useful in multiphoton calculations.[26,46,47] The main advantage encountered in using this representation lies in the fact that the infinite sums running over the physical (discrete + continuous) atomic spectrum are replaced by sums over the (discrete) Sturmian spectrum.[47,48] One is thus faced with the standard problem of the numerical computation of infinite (possibly multiple) series. These series are usually convergent, but, as we shall show, in some cases of physical interest, they may diverge. Thus

we have found it useful to accelerate their convergence with the help of the so-called ϵ-algorithm,[49,50] which is closely connected to the Padé approximants.[51]

Before going further and presenting the computational procedure we have used, we should emphasize that we have not introduced the concept of quasienergy spectrum of the atom in the presence of the strong electromagnetic field. As a matter of fact, it has been pointed out by several authors that the external field mixes atomic states with the same principal quantum number and the same parity.[22,52] As a consequence, the atomic wave functions should be expressed as linear combinations of unperturbed wave functions, which obviously renders the computation much more intricate. However, by comparing with our own results, we have verified that the magnitude of the corrections so introduced remains relatively small (see below). Moreover it may be demonstrated that the shift of the barycenter of a given level is unaffected by such a transformation,[22] and, as the level shifts themselves are small corrections, we are confident that our results are very good approximations. This is obviously the case in fourth-order calculations, in which the quasienergy formalism would lead to unnecessary complications.

A. Second-order amplitude

The reduced atomic second-order amplitude, Eq. (18), is conveniently rewritten in terms of the Coulomb Green's function

$$G(E)=\mathsf{S}_{n_1}\frac{|n_1\rangle\langle n_1|}{E-E_{n_1}} \tag{23}$$

and becomes

$$\tau^{(2)}_{nlm}(\omega)=\langle nlm\,|\,z[G(E_n+\omega)+G(E_n-\omega)]z\,|\,nlm\rangle\ . \tag{24}$$

Note that hereafter atomic units will be used.

After factorizing the angular part one has

$$\tau^{(2)}_{nlm}(\omega)=\sum_{\lambda}\frac{l_>^2-m^2}{(2l+1)(2l_>+1)}\times[T^{(2)}_{\lambda}(\omega)+T^{(2)}_{\lambda}(-\omega)]\ , \tag{25}$$

where $\lambda=l-1,l+1$; $l_>=\sup(l,\lambda)$ and the second-order radial amplitudes $T^{(2)}_{\lambda}(\pm\omega)$ are

$$T^{(2)}_{\lambda}(\pm\omega)=\langle nl\,|\,rG_{\lambda}(E_n\pm\omega)r\,|\,nl\rangle\ . \tag{26}$$

Here

$$\langle r\,|\,nl\rangle=R_{nl}(r)=C_{n,l}e^{-r/n}r^{l}\,{}_1F_1(-n+l+1;2l+2;2r/n) \tag{27a}$$

with

$$C_{n,l}=2^{l+1}n^{-l-2}\frac{1}{(2l+1)!}\left[\frac{(n+l)!}{(n-l-1)!}\right]^{1/2} \tag{27b}$$

being radial hydrogenic wave functions.[53]

$G_{\lambda}(E)$ is the radial component of the Coulomb Green's function for angular momentum λ, whose expansion over the Coulomb Sturmian basis reads[47]

$$\langle r'\,|\,G_{\lambda}(E)\,|\,r\rangle=\sum_{\nu=\lambda+1}^{\infty}\frac{S_{\nu\lambda}(p_0r')S_{\nu\lambda}(p_0r)}{1-\nu p_0},\quad p_0=\sqrt{-2E}\ , \tag{28}$$

where

$$S_{\nu\lambda}(p_0r)=N_{\nu,\lambda}(p_0)e^{-p_0r}r^{\lambda}\,{}_1F_1(-\nu+\lambda+1;2\lambda+2;2p_0r)\ , \tag{29a}$$

with

$$N_{\nu,\lambda}(p_0)=(2p_0)^{\lambda+1}\frac{1}{(2\lambda+1)!}\left[\frac{(\nu+\lambda)!}{(\nu-\lambda-1)!}\right]^{1/2} \tag{29b}$$

being the radial Coulomb Sturmian functions for angular momentum λ and energy $E=-p_0^2/2<0$.

Thus, $T^{(2)}_{\lambda}(\pm\omega)$ may be rewritten as

$$T^{(2)}_{\lambda}(\pm\omega)=C^2_{n,l}\sum_{\nu=\lambda+1}^{\infty}[N_{\nu,\lambda}(p_0^{\pm})]^2(1-\nu p_0^{\pm})^{-1}[J(n,l,1/n\,|\,\nu,\lambda,p_0^{\pm})]^2\ , \tag{30}$$

where $p_0^{\pm}=[-2(E_n\pm\omega)]^{1/2}$ and the quantity denoted J is an integral of the following general form:

$$J(\nu,\lambda,p_0\,|\,\nu',\lambda',p_0')=\int_0^\infty dr\, r^{3+\lambda+\lambda'}e^{-r(p_0+p_0')}\,{}_1F_1(\lambda+1-\nu;2\lambda+2;2p_0r)\,{}_1F_1(\lambda'+1-\nu';2\lambda'+2;2p_0'r)\ . \tag{31}$$

In the particular case $\lambda'=\lambda\pm1$ we are concerned with, those integrals can be evaluated in closed form via a generalization of Gordon's formula,[47,53] and may be conveniently factorized as follows[54]:

$$J(\nu,\lambda,p_0\,|\,\nu',\lambda',p_0')=\left[\frac{p_0'-p_0}{p_0'+p_0}\right]^{\nu+\nu'}\tilde{J}(\nu,\lambda,p_0\,|\,\nu',\lambda',p_0')\ , \tag{32}$$

where $\tilde{J}$ is now a combination of hypergeometric polynomials times algebraic factors, and

$$\begin{aligned}\tilde{J}(\nu,\lambda,p_0\,|\,\nu',\lambda',p_0')=(-1)^{\nu+\lambda_<}&\frac{(2\lambda_>+1)!}{2p_{0>}^2(p_0-p_0')^{2\lambda_>+2}}\\ &\times\sum_{q=0}^{2}\begin{bmatrix}2\\q\end{bmatrix}[p_{0>}(\nu_>-1+q)-p_{0<}\nu_<]\\ &\times\left[\frac{p_{0>}-p_{0<}}{p_{0>}+p_{0<}}\right]^q{}_2F_1\left[\lambda_>+1-\nu_>-q,\lambda_<+1-\nu_<;2\lambda_>;\frac{-4p_0p_0'}{(p_0-p_0')^2}\right]\ .\end{aligned} \tag{33}$$

Here $\lambda_>=\sup(\lambda,\lambda')$, $\lambda_<=\inf(\lambda,\lambda')$, and $p_{0>},\nu_>$ ($p_{0<},\nu_<$) are the values of the parameters p_0,ν corresponding to $\lambda_>$ ($\lambda_<$).

From those results one can easily reexpress the amplitude $T_\lambda^{(2)}(\pm\omega)$ as an infinite series in the variable

$$\zeta_\pm=\left[\frac{p_0^\pm-1/n}{p_0^\pm+1/n}\right]^2\ .$$

We have

$$T_\lambda^{(2)}(\pm\omega)=\sum_{r=0}^{\infty}a_r(\pm\omega)\zeta_\pm^r\ , \tag{34}$$

where the coefficients $a_r(\pm\omega)$, which exhibit a somewhat complicated structure, can be deduced from the formulas (30)–(33). Such an expression for the radial amplitude is general, whatever hydrogenic state $|n,l\rangle$ is considered, and represents, in some sense, an extension of the analytical formulas previously obtained for the low-lying states by using integral representations of the Coulomb Green's function.[47,55–57] As a matter of fact, one can show (see Appendix A) that the series Eq. (34) may always be expressed as a sum of algebraic factors, plus one term proportional to a Gauss hypergeometric function of the general form ${}_2F_1(1,b;b+p;\zeta)$, where $p>1$ is an integer.[26] This result enables us to make the connection with the above-mentioned compact formulas and to gain some insight into the analytical properties of the series Eq. (34). On the other hand, the hypergeometric functions ${}_2F_1(1,b;c;z)$ have a continued-fraction expansion converging in the whole complex plane, except on the cut $(+1,+\infty)$.[58] This theorem permits us to assess the validity of the ϵ-algorithm technique we have used for accelerating the convergence of the series (34) (see below).

B. Fourth-order amplitude

Again the reduced fourth-order hydrogenic amplitudes entering expression (22) may be reexpressed in terms of Coulomb Green's functions. Note, however, that two cases should be distinguished according to whether the initial state $|nl\rangle$ is excluded or not from the generalized sum over the states $|n_2l_2\rangle$, i.e., if the corresponding diagram Fig. 1(b) is improper or not. As a matter of fact, if the state $|nl\rangle$ is excluded from the infinite summation, one has to introduce the so-called reduced Coulomb Green's function $G^{(n)}(E_n)$ whose expression will be given below.

1. Contribution of the proper diagrams

In the following, for the sake of simplicity, we shall consider the case of a typical amplitude corresponding, for instance, to the diagram I, in Fig. 1(b). After the angular part is factorized out, any fourth-order amplitude of the general form

$$\langle nlm \mid zG(E_n+\omega)zG(E_n+2\omega) \times zG(E_n+\omega)z \mid nlm \rangle$$

may be written in terms of the following reduced radial amplitudes:

$$T^{(4)}_{\lambda_1\lambda_2\lambda_3} = \langle nl \mid rG_{\lambda_1}(E_n+\omega)rG_{\lambda_2}(E_n+2\omega)rG_{\lambda_3}(E_n+\omega)r \mid nl \rangle \tag{35}$$

with $\lambda_1 = l\pm 1$, $\lambda_2 = l, l\pm 2$, $\lambda_3 = l\pm 1$. By using the Sturmian representation of the Coulomb Green's function, amplitudes of this kind may be rewritten as a triple sum, generalizing in a straightforward way the second order. With similar notations one has

$$\begin{aligned} T^{(4)}_{\lambda_1\lambda_2\lambda_3} = (C_{nl})^2 & \sum_{\nu_3=\lambda_3+1}^{\infty} \frac{N^2_{\nu_3\lambda_3}(p_{03})}{1-\nu_3 p_{03}} J(n,l,1/n \mid \nu_3,\lambda_3,p_{03}) \\ & \times \sum_{\nu_2=\lambda_2+1}^{\infty} \frac{N^2_{\nu_2\lambda_2}(p_{02})}{1-\nu_2 p_{02}} J(\nu_3,\lambda_3,p_{03} \mid \nu_2,\lambda_2,p_{02}) \\ & \times \sum_{\nu_1=\lambda_1+1}^{\infty} \frac{N^2_{\nu_1\lambda_1}(p_{01})}{1-\nu_1 p_{01}} J(\nu_2,\lambda_2,p_{02} \mid \nu_1,\lambda_1,p_{01}) J(\nu_1,\lambda_1,p_{01} \mid n,l,1/n) , \end{aligned} \tag{36}$$

where $p_{01}=p_{03}=[-2(E_n+\omega)]^{1/2}$ and $p_{02}=[-2(E_n+2\omega)]^{1/2}$. The same analysis as in the second-order case leads us to reexpress this amplitude in terms of a triple series, in three variables[59]:

$$T^{(4)}_{\lambda_1\lambda_2\lambda_3} = \sum_{r_1 r_2 r_3} a_{r_1 r_2 r_3} \zeta_1^{r_1} \zeta_2^{r_2} \zeta_3^{r_3} , \tag{37}$$

where

$$\zeta_1 = \frac{(1/n-p_{01})(p_{02}-p_{01})}{(1/n+p_{01})(p_{02}+p_{01})} ,$$

$$\zeta_2 = \frac{(p_{01}-p_{02})(p_{03}-p_{02})}{(p_{01}+p_{02})(p_{03}+p_{02})} ,$$

$$\zeta_3 = \frac{(p_{02}-p_{03})(1/n-p_{03})}{(p_{02}+p_{03})(1/n+p_{03})} ,$$

and the explicit expression of $a_{r_1 r_2 r_3}$, which is too long to be reproduced here, may be found by inspection with the help of formulas (27b), (29b), and (33).

It should be stressed that such a Sturmian series expansion is valid, whatever initial hydrogenic bound state is considered. Although it is likely that these series have a structure similar to those of Lauricella's hypergeometric functions of several variables,[60] one cannot derive their analytic properties so easily as in the second-order case. However, as we shall show in the following section, the numerical convergence of the series (37) is usually very good. Note also that if one had used integral representations of the Coulomb Green's function, the amplitude $T^{(4)}_{\lambda_1\lambda_2\lambda_3}$ would have been expressed in terms of multiple integrals. For a general derivation of such a result, based on a group-theoretical approach, see Ref. 61.

2. *Contributions of the improper diagrams*

The general expression of these contributions, which has been given in Eq. (22), can be split into a fourth-order term and a product of two second-order amplitudes, respectively. Again, for the sake of simplicity, we shall consider only a typical case exemplified by the diagram III, in Fig. 1(b). In the fourth-order term, the initial state $|nl\rangle$ has to be consistently excluded from the infinite sum over the states $|n_2\lambda_2\rangle$, which leads us to consider the following reduced radial amplitudes:

$$T'^{(4)}_{\lambda_1\lambda_2\lambda_3} = \langle nl \mid rG_{\lambda_3}(E_n+\omega)rG^{(n)}_{\lambda_2}(E_n)rG_{\lambda_1}(E_n+\omega)r \mid nl \rangle , \tag{38}$$

where $G^{(n)}_{\lambda}(E_n)$ is the radial component, for angular momentum λ, of the reduced Coulomb Green's function[62,63], and

$$G^{(n)}_{\lambda}(E_n) = \sum_{n' \neq n} \frac{|n'\lambda\rangle\langle n'\lambda|}{E_n - E_{n'}} = \left[G_{\lambda}(E) - \frac{|n\lambda\rangle\langle n\lambda|}{E-E_n} \right]_{E=E_n} = \frac{\partial}{\partial E}(E-E_n)G_{\lambda}(E)\Big|_{E=E_n} . \tag{39}$$

Several representations of $G_\lambda^{(n)}(E_n)$ in terms of transcendental functions have been proposed so far,[62,63] but unfortunately they share a common lack of symmetry in the variables which hinder us from obtaining closed expressions of the amplitude. Instead we have found it useful to rely upon a Sturmian expansion for $G_\lambda^{(n)}(E_n)$ (see Appendix B and Ref. 64). One gets the following general expression:

$$\langle r \mid G_\lambda^{(n)}(E_n) \mid r' \rangle = \sum_{\nu \neq n} \frac{S_{\nu,\lambda}(r/n) S_{\nu,\lambda}(r'/n)}{1-\nu/n} - \tfrac{1}{2} S_{n,\lambda}(r/n) S_{n,\lambda}(r'/n)$$
$$- \tfrac{1}{2}[(n+\lambda+1)(n-\lambda)]^{1/2}[S_{n,\lambda}(r/n) S_{n+1,\lambda}(r/n) + S_{n+1,\lambda}(r/n) S_{n,\lambda}(r'/n)]$$
$$+ \tfrac{1}{2}[(n+\lambda)(n-\lambda-1)]^{1/2}[S_{n,\lambda}(r/n) S_{n-1,\lambda}(r'/n) + S_{n-1,\lambda}(r/n) S_{n,\lambda}(r'/n)] \; . \tag{40}$$

After being replaced in Eq. (38), $T'^{(4)}_{\lambda_1\lambda_2\lambda_3}$ can be reexpressed as a triple series, similar to that of Eq. (37), plus simpler terms reducing to a product of two second-order amplitudes.

All that remains is the contribution of the product $\Delta E_n^{(2)}(\partial/\partial E)\Delta E_n^{(2)}$, whose radial part, in the case of the graph III considered here, reduces to

$$-T_{l\pm1}^{(2)}(+\omega) \left[\frac{\partial}{\partial E} \sum_{n_1} \frac{|\langle nl \mid r \mid n_1\lambda_1 \rangle|^2}{E+\omega-E_{n_1}} \right]_{E=E_n} = T_{l\pm1}^{(2)}(+\omega) \langle nl \mid r[G_{\lambda_1}(E_n+\omega)]^2 r \mid nl \rangle \; , \tag{41}$$

where we have dropped the small imaginary part $i\eta$ in the denominator. Again we have used a Sturmian representation of the derivative of the Coulomb Green's function:

$$-\frac{\partial}{\partial E} G(E) = (E - H_{at})^{-2} \; . \tag{42}$$

The expansion, which has been obtained as a by-product when establishing the one for $G_\lambda^{(n)}(E_n)$, is given in Appendix B.

IV. NUMERICAL RESULTS

The numerical results, which we shall present and discuss now, were obtained by summing the series given in Eq. (34) for second-order amplitudes and in Eq. (37) for the fourth-order ones. In order to improve the efficiency of the computation we have used routinely the ϵ algorithm which has provided reliable results in case of slow convergence (and even in case of divergence) of the series. In subsection A we present our results for the second-order contribution to the level shift and compare them with previous calculations. Fourth-order contributions are given in subsection B. In order to get an independent check of our results, we have performed also a crude evaluation of the fourth-order amplitudes by retaining only a limited set of bound states in the infinite summations $\sum_{n_1}$. We have found that, in contradistinction with the second-order case, such an approximation leads to very poor results and we analyze the reasons of this rather deceptive behavior. In subsection C, we consider the limiting case $\omega \to 0$ and make the connection with the dc-Stark effect. Finally, the case $2\omega > |E_n|$, i.e., when two-photon ionization is possible, is investigated in subsection D, leading to an extension of the optical theorem to the case of multiphoton forward-scattering processes.

A. Second-order level shift

The computation merely consists of summing the series Eq. (34). Two cases should be distinguished according to whether the laser frequency ω is smaller or greater than the ionization energy of the atomic state $|E_n| = 1/2n^2$. In the first case the parameters $p_0^{\pm} = (n^{-2} \mp 2\omega)^{1/2}$ are real positive and $|\zeta_1| < 1$. Then the series (34) converges very well and the computation of approximate values of the sum does not give rise to any particular trouble. Our results are in excellent agreement with the most accurate values given by Gavrila[55] for the two-photon amplitude $\tau_{1s}^{(2)}(\omega)$, Eq. (24) (see Table I). One observes also a good coincidence between our results and those obtained by Dubreuil[25] for the second-order contribution to the light shift of excited states ($n=2,3$) irradiated by a neodymium laser ($\omega = 9440\ \text{cm}^{-1} = 4.3 \times 10^{-2}$ a.u.) (see Table II). In this table we present also the data obtained by Zon *et al.*[22] who used the quasienergy formalism: The

TABLE I. Comparison of our results with those of Gavrila (Ref. 55) for the second-order transition amplitude $\tau_{1S}^{(2)}(\omega)$, in the $1S$ state of hydrogen. An excellent agreement is found between these results obtained by using two different representations of the Coulomb Green's function.

ω	This work		Gavrila	
a.u.	$\mathrm{Re}(\tau_{1S}^{(2)})$	$\mathrm{Im}(\tau_{1S}^{(2)})$	$\mathrm{Re}(\tau_{1S}^{(2)})$	$\mathrm{Im}(\tau_{1S}^{(2)})$
0.02	−0.001 804		−0.001 804	
0.04	−0.007 269		−0.007 269	
0.08	−0.029 937		−0.029 936	
0.1	−0.047 843		−0.047 843	
0.2	−0.237 667		−0.237 667	
0.4	2.691 623		2.691 623	
0.8	1.226 119	−0.544 400	1.226 119	−0.544 400
1.0	1.205 980	−0.362 705	1.205 980	−0.362 705
2.0	1.100 074	−0.095 814	1.100 074	−0.095 814
5.0	1.025 888	−0.014 455	1.025 888	−0.014 455
10	1.008 098	−0.003 193	1.008 097	−0.003 193

discrepancies never exceed 1% for the states considered here.

In the second case, i.e., $\omega > 1/2n^2$, ionization can take place and the amplitude $\tau_{nlm}^{(2)}(\omega)$, Eq. (24), acquires an imaginary part connected via the optical theorem to the photoionization cross section.[55] More precisely the argument of the Coulomb Green's function in the reduced amplitude $T_{\lambda}^{(2)}(+\omega)$, Eq. (26), becomes positive and accordingly the parameter p_0^+ is imaginary: $p_0^+ = -i(\,|n^2 - 2\omega|\,)^{1/2}$. As a consequence $|\zeta_+| = 1$ and one observes that (34) converges very slowly or even diverges. In order to sum the series, we have used the ϵ algorithm, closely connected to the Padé approximants.[49–51] The results we have obtained in that way are in excellent agreement with those of Gavrila (see Table I). Moreover, since the series (34) can always be expressed in terms of Gauss hypergeometric functions ${}_2F_1(1,b;c;z)$, one can prove the convergence of the ϵ-algorithm scheme (see Appendix A and Ref. 26). In Table III we have compared our results for the excited states with those obtained by Zon *et al.*[22] As expected, the discrepancies are sometimes noticeable, the mixing of states becoming of increasing importance in this case. Note, however, that the order of magnitude still remains correct.

TABLE II. Second-order contribution to the light shift, expressed in cm^{-1}, of hydrogenic states $|nlm\rangle$ irradiated with a linearly polarized neodymium laser ($\omega = 9440\ \mathrm{cm}^{-1}$, $I = 1\ \mathrm{MW/cm}^2$). Comparison is made with Dubreuil's results obtained by a direct summation method (Ref. 25) and the data given by Zon *et al.* (Ref. 22) who took into account the degeneracy of the atomic states, in the presence of the laser field. These results show that the introduction of the quasienergy formalism leads to corrections smaller than 1% in the case considered here.

nlm	This work	Dubreuil	Zon
200	−2.841(−4)	−2.839(−4)	−2.844(−4)
210	−5.264(−4)	−5.262(−4)	−5.266(−4)
211	−3.787(−4)	−3.786(−4)	−3.788(−4)
310	2.191(−4)	2.195(−4)	2.185(−3)
311	1.287(−3)	1.303(−3)	1.302(−3)
321	2.689(−3)	2.711(−3)	2.703(−3)

B. Fourth-order level shift

The main difficulty encountered in the computation is to sum the triple series, Eq. (37). Other contributions, arising from the improper diagrams, always reduce to products of two simple series, with structures similar to those we have discussed above. Now, the convergence of the triple sum (37) depends essentially on the respective values of the variables ζ_i, $i = 1,2,3$. One can verify that, if the laser frequency is such that $2\omega < 1/2n^2$, the parameters p_{0i} are real positive and $|\zeta_i| < 1$, which ensures a good convergence of the summation process. Note, however, that we have routinely used the ϵ algorithm, which has permitted us to significantly improve the convergence rate of the nested sums. We present in Table IV the resulting values of the fourth-order amplitude (or dynamic dipole polarizability) $\tau_{1S}^{(4)}(\omega)$, for the ground state, at several selected values of the laser frequency ω.

In the absence of previous theoretical calculations

TABLE III. Two-photon hydrogenic amplitudes $\tau^{(2)}_{nlm}(\omega)$ in atomic units for a linearly polarized neodymium laser ($\omega = 4.30 \times 10^{-2}$ a.u.). The columns headed (a) and (b) are, respectively, the real and imaginary parts of $\tau^{(2)}_{nlm}(\omega)$. By comparing with the results obtained by Zon *et al.* (Ref. 22) one can check that the quasienergy formalism leads to significant corrections only for excited states (see Sec. IV A).

State	Zon *et al.*		This work	
nlm	(a)	(b)	(a)	(b)
100	4.55		4.55	
200	181.6		181.7	
210	336.5		336.7	
300	−727.0		−753.0	
310	−1404.		−1388.	
320	−2036.		−1984.	
321	−1734.		−1719.	
400	−525.4	201.8	−527.5	174.5
410	−568.6	222.6	−568.3	226.7
420	−599.0	141.7	−596.0	168.3
421	−601.5	148.8	−601.0	148.4
500	−551.5	73.41	−522.5	92.70
510	−457.2	107.2	−452.7	115.1
520	−395.7	100.2	−424.4	80.47
521	−462.7	71.07	−462.7	70.84

and in order to check our computation, we have attempted to get estimated values of $\tau^{(4)}_{1S}(\omega)$. To this end, we performed a rough evaluation of the infinite sums, running over the (physical) spectrum, by including only a limited number of bound states in the summation. More precisely, we used a truncated representation of the infinite generalized sum entering the Coulomb Green's function expansion:

$$G_\lambda(E) \simeq \sum_{n=\lambda+1}^{n_{\max}} \frac{|n\lambda\rangle\langle n\lambda|}{E-E_n}$$

instead of

$$G_\lambda(E) = \sum_{n=\lambda+1}^{\infty} \frac{|n\lambda\rangle\langle n\lambda|}{E-E_n} + \int_0^\infty dk \frac{|k\lambda\rangle\langle k\lambda|}{E-E_k} , \tag{43}$$

where the integral represents the continuous spectrum contribution. Since in the electric dipole

TABLE IV. Second-order, $\tau^{(2)}_{1S}(\omega)$, and fourth-order, $\tau^{(4)}_{1S}(\omega)$, dynamic dipole polarizability of hydrogen in the ground state, in atomic units. (a) CO_2 laser; (b) neodymium; (c) ruby; (d) neodymium second harmonic.

ω a.u.	$-\tau^{(2)}_{1S}(\omega)$	$-\tau^{(4)}_{1S}(\omega)$
2.00(−4)	4.500 00	333.285
5.00(−4)	4.500 00	333.289
1.00(−3)	4.500 03	333.301
4.30(−3)(a)	4.500 49	333.5
5.00(−3)	4.500 66	333.6
1.00(−2)	4.502 66	334.6
4.30(−2)(b)	4.549 76	358.6
5.00(−2)	4.567 55	368.2
6.56(−2)(c)	4.617 90	397.0
8.60(−2)(d)	4.706 60	454.6
1.00(−1)	4.784 30	513.1

TABLE V. Comparison of the exact computation with a crude estimation of the value of a typical fourth-order amplitude

$$T^{(4)}_{101}(\omega) = \langle 10 | rG_1(E_1+\omega) rG_0(E_1+2\omega) \times rG_1(E_1+\omega) r | 10\rangle ,$$

obtained by replacing

$$G_\lambda(E) \sim \sum_{n=\lambda+1}^{n_{\max}} |n\lambda\rangle\langle n\lambda| / (E-E_n) .$$

As $n_{\max}$ increases, the estimated value of $T^{(4)}_{101}(\omega)$ does not become stable and departs more and more from the exact one.

$n_{\max}$	$\omega = 0.1425$	$\omega = 0.086$
5	−0.7503(+4)	−0.2322(+4)
10	−0.8769(+4)	−0.3012(+4)
15	−0.1028(+5)	−0.3766(+4)
20	−0.1179(+5)	−0.4516(+4)
⋮	⋮	⋮
Exact value	−0.3598(+4)	−0.6855(+3)

gauge this latter contribution is usually small with respect to the discrete spectrum contribution,[27,53] such an approximation provides reliable estimates in two-photon (second-order) calculations.[27] Unfortunately, this *a priori* favorable circumstance does not hold anymore in fourth-order computations as one can convince oneself by consulting Table V: If one increases $n_{\max}$, the obtained values for a typical amplitude $T^{(4)}_{\lambda_1\lambda_2\lambda_3}$ do not tend towards a stable limit and, on the contrary, increase steadily. This rather surprising behavior may be qualitatively explained by the fact that the main contributions to the multiple sums come from matrix elements connecting atomic states with the same principal quantum number [Ref. 53, formula (63.5); note the sign is erroneous in this reference]:

$$\langle n,l \mid r \mid n,l\pm 1\rangle = -\tfrac{3}{2} n (n^2 - l_>^2)^{1/2} . \tag{44}$$

Such matrix elements *grow* like n^2, unlike those connecting states with different quantum numbers n and n' which *decrease* like $(n_>)^{-3/2}$, where $n_> = \sup(n,n')$.[53] The more we include states in the triple summation, the larger these contributions, which do not compensate one to the other and are not counterbalanced by the continuous spectrum as they should be in a correct calculation, become. This very peculiar behavior illustrates the difficulties encountered when trying to get sensible estimates in high-order perturbative computations.

C. Connection with the dc-Stark effect

A test, which does not suffer such drawbacks, is provided by considering the limit $\omega \to 0$. As a matter of fact, one might expect that, if the frequency goes to zero, the expressions of $\Delta E_n^{(2)}$ and $\Delta E_n^{(4)}$ tend to the corresponding dc-Stark shift corrections. This is verified at once in the second-order case since

$$\lim_{\omega\to 0} \Delta E_n^{(2)} = \lim_{\omega\to 0} \frac{E^2}{2E_0^2} \tau_n^{(2)}(\omega) = \frac{E^2}{E_0^2} \sum_{n_1} \frac{|(z)_{nn_1}|^2}{E_n - E_{n_1}} , \tag{45}$$

which is exactly the first nonvanishing term in perturbation-theory calculation of the Stark effect.[39] One can verify in Table IV that, as $\omega \to 0$, the dynamic dipole polarizability $\tau_{1s}^{(2)}(\omega)$ tends towards $\frac{9}{2}$, in coincidence with the exact result for the static dipole polarizability.[65,66]

The verification is more intricate in the fourth-order case since the first terms contained in the expression of $\tau_n^{(4)}(\omega)$, Eq. (22), become singular when $\omega \to 0$. In these terms, which correspond to the proper diagrams I and II in Fig. 1(b), the initial state $|n\rangle$ *is not* excluded from the infinite sum over the states $|n_2\rangle$, and they can be rewritten accordingly[67]:

$$\tau_n^{(4)}(\omega)\Big|_{\mathrm{I+II}} = \sum_{n_3}\sum_{n_1} \frac{|(z)_{nn_3}|^2 |(z)_{nn_1}|^2}{2\omega} \left[\frac{1}{(E_n - E_{n_3} + \omega)(E_n - E_{n_1} + \omega)} + \frac{1}{(E_n - E_{n_3} - \omega)(E_n - E_{n_1} - \omega)} \right]$$
$$+ \sum_{n_3}\sum_{n_2\neq n}\sum_{n_1} (z)_{nn_3}(z)_{n_3n_2}(z)_{n_2n_1}(z)_{n_1n} \left[\frac{1}{(E_n - E_{n_3} + \omega)(E_n - E_{n_2} + 2\omega)(E_n - E_{n_1} + \omega)} \right.$$
$$\left. + \frac{1}{(E_n - E_{n_3} - \omega)(E_n - E_{n_2} - 2\omega)(E_n - E_{n_1} - \omega)} \right] . \tag{46}$$

Taking the limit $\omega \to 0$, one easily extracts the apparent singularity in the first term, and one obtains

$$\lim_{\omega\to 0} \tau_n^{(4)}(\omega)\Big|_{\mathrm{I+II}} = 2 \sum_{n_3}\sum_{n_2\neq n}\sum_{n_1} \frac{(z)_{nn_3}(z)_{n_3n_2}(z)_{n_2n_1}(z)_{n_1n}}{(E_n - E_{n_3})(E_n - E_{n_2})(E_n - E_{n_1})} - 2 \sum_{n_1}\sum_{n_3} \frac{|(z)_{nn_1}|^2 |(z)_{nn_3}|^2}{(E_n - E_{n_1})^2 (E_n - E_{n_3})} . \tag{47}$$

After regrouping with the other, nonsingular terms, contained in the expression (22), one gets eventually

$$\lim_{\omega\to 0} \tau_n^{(4)}(\omega) = 6 \left[\sum_{n_3}\sum_{n_2\neq n}\sum_{n_1} \frac{(z)_{nn_3}(z)_{n_3n_2}(z)_{n_2n_1}(z)_{n_1n}}{(E_n - E_{n_3})(E_n - E_{n_2})(E_n - E_{n_1})} - \sum_{n_3}\sum_{n_1} \frac{|(z)_{nn_3}|^2 |(z)_{nn_1}|^2}{(E_n - E_{n_3})^2 (E_n - E_{n_1})} \right] , \tag{48}$$

which coincides, to within a factor 6, with the fourth-order contribution to the static dipole polarizability.[39] When specializing to the ground state (see Table IV), one can verify that, as ω goes towards zero, $\tau_{1S}^{(4)}(\omega) \to -333.285$, which is close to within 1 part in 10^6 from the exact value[65,66]:

$$\lim_{\omega\to 0}\tau_{1S}^{(4)}(\omega)=-6\left[\frac{3555}{2^5}\right].$$

Note that the factor $6=\binom{4}{2}$ arises from the number of combinations of four indistinguishable photons, taken two by two, as displayed in the Feynman diagrams, Fig. 1(b). This factor disappears when averaging the fourth power of the electric field operator, Eq. (21).

D. Generalization of the optical theorem

If the laser frequency ω increases such that $|E_n|<2\omega<2|E_n|$, two-photon ionization is possible and the amplitude corresponding to the diagram I in Fig. 1(b) (two-photon absorption followed by the stimulated emission of two identical photons) becomes complex. This may be shown by considering its expression

$$\tau_n^{(4)}(\omega)\Big|_{\rm I}=\lim_{\eta\to 0}\langle n\,|\,zG(E_n+\omega+i\eta)zG(E_n+2\omega+i\eta)zG(E_n+\omega+i\eta)z\,|\,n\rangle\ , \tag{49}$$

where we have displayed the small imaginary part $i\eta$. This latter part is of importance, in the case considered here, since the real part of the argument of the second Green's function is positive. One has explicitly

$$\lim_{\eta\to 0}G(E_n+2\omega+i\eta)=\sum_{n_2}\frac{|n_2\rangle\langle n_2|}{E_n+2\omega-E_{n_2}}+\lim_{\eta\to 0}\int_0^\infty dk_2\frac{|k_2\rangle\langle k_2|}{E_n+2\omega+i\eta-E_{k_2}}\ , \tag{50}$$

where the integral, corresponding to the continuous spectrum contribution, becomes

$$\lim_{\eta\to 0}\int_0^\infty dk_2\frac{|k_2\rangle\langle k_2|}{E_n+2\omega-E_{k_2}+i\eta}=P\int_0^\infty dk_2\frac{|k_2\rangle\langle k_2|}{E_n+2\omega-E_{k_2}}-i\pi\,|k_2\rangle\langle k_2|\,\Big|_{E_{k_2}=E_n+2\omega}\ . \tag{51}$$

As a consequence, the imaginary part of $\tau_n^{(4)}(\omega)$ is

$$\mathrm{Im}\tau_n^{(4)}(\omega)=-\pi\,|\,\langle k_2\,|\,zG(E_n+\omega)z\,|\,n\rangle\,|^2\,|_{E_{k_2}=E_n+2\omega}\ , \tag{52}$$

which is exactly, within a factor $-\pi$, the square of the two-photon ionization amplitude.[54] This may be rewritten in terms of the two-photon ionization cross section $\sigma_n^{(2)}$, extending in some sense the optical theorem to the case of four-photon forward scattering:

$$a_0^2\mathrm{Im}\tau_n^{(4)}(\omega)=-\frac{1}{4\pi}\frac{\sigma_n^{(2)}}{\alpha\omega}\frac{I_0}{I}\ . \tag{53}$$

This has provided us with another independent check of our computation; see Table VI. However, it is worth noting that, in this particular situation, the Sturmian triple series Eq. (37), associated with the amplitude (49), does not converge. As a matter of fact, when $2\omega>|E_n|$, the parameter $p_{02}=-i(2|E_n+2\omega|)^{1/2}$ and $|\xi_2|=1$: one observes then a strong divergence of the triple series (37). Again, we have been able to sum it by using the ϵ algorithm, which has permitted us to easily derive the corresponding Padé table.

The power of this technique is illustrated in Table VII, where we compare a typical (divergent) sequence of partial sums obtained from a direct summation of Eq. (37), and a (convergent) sequence of Padé approximants obtained via the ϵ algorithm. Note, however, that the convergence of the Padé sequences cannot be assessed on the grounds of general theorems in the case of such multiple series. In spite of the lack of general proof, the accuracy of our numerical results gives yet further support to our method.

TABLE VI. Comparison of our results for the imaginary part of the fourth-order dynamic dipole polarizability $\mathrm{Im}\tau_{1S}^{(4)}(\omega)$ of the ground state with the square of the corresponding two-photon-ionization amplitude (Ref. 54). This comparison validates our numerical approach for summing the (divergent) triple series Eq. (37).

ω (a.u.)	$\tau_{1S}^{(4)}(\omega)$	$-\pi\,\lvert\langle k_2\,\rvert zGz\,\rvert 1S\rangle\rvert^2$
0.3799 ($\lambda=1200$ Å)	−0.8350(+4)	−0.8350(+4)
0.2849 ($\lambda=1600$ Å)	−0.4164(+4)	−0.4158 (+4)

TABLE VII. Comparison of a divergent sequence of partial sums

$$S_n=\sum_{r_1=0}^{n}\sum_{r_2}^{\infty}\sum_{r_3}^{\infty}a_{r_1r_2r_3}\xi_1^{r_1}\xi_2^{r_2}\xi_3^{r_3}\ ,$$

with the corresponding diagonal sequence of Padé approximants obtained from the ϵ-algorithm scheme. The sum considered here corresponds to a typical complex amplitude $T^{(4)}_{101}(\omega)$, Eq. (37), associated to the diagram I at $\omega=0.3799$ ($\lambda=1200$ Å), i.e., when two-photon ionization of the ground state can take place.

n	S_n	$[n/n]$
1	$0.963(+7)-i.222(+7)$	$-0.130(+8)-i.265(+7)$
2	$-0.682(+8)-i.142(+9)$	$+0.256(+7)+i.135(+8)$
3	$-0.726(+9)+i.692(+9)$	$+0.988(+7)+i.248(+7)$
4	$0.381(+10)+i.142(+10)$	$+0.650(+7)-i.526(+6)$
⋮	⋮	⋮
7	$0.768(+11)+i.330(+10)$	$+0.578(+7)-i.139(+5)$
8	$-0.996(+11)-i.125(+12)$	$+0.578(+7)-i.148(+5)$
9	$-0.502(+11)+i.302(+12)$	$+0.578(+7)-i.151(+5)$
⋮	⋮	
17	$-0.333(+13)+i.114(+14)$	
18	$-0.144(+14)-i.815(+13)$	
⋮	⋮	

V. CONCLUSION

In this paper, we have presented an efficient and reliable method, based on a Sturmian expansion of the Coulomb Green's function, for computing high-order perturbative amplitudes in hydrogen. The stimulated radiative shift of a hydrogenic state may be obtained, up to and including fourth order, from the formula

$$\Delta E_n(\omega)\simeq\frac{I}{2I_0}\tau_n^{(2)}(\omega)+\left[\frac{I}{2I_0}\right]^2\tau_n^{(4)}(\omega)+\cdots\ . \tag{54}$$

It is interesting to note that, for the ground state, at laser frequencies $\omega<\frac{3}{8}$, the shift is negative, i.e., the $1S$ state is lowered, in similarity with the dc-Stark effect. As a consequence, at low frequencies, the stimulated radiative corrections in S states are opposite to the Lamb shift which is positive (see Fig. 2). One can check that this latter shift can be compensated in the $1S$ state by irradiating a H atom with a neodymium laser working at $I\sim10^{11}$ W/cm^2.

Another interesting fact which may be deduced from our results listed in Table IV is that, still at lower frequencies, the dc-Stark results provide a fair approximation of the order of magnitude of the corrections. Moreover, this similarity indicates that the perturbative series for the ac-Stark shift is likely to be asymptotic.[12] There is no doubt that this point would deserve further investigations in connection with the other expansions obtained, for instance, in terms of operator continued fractions.

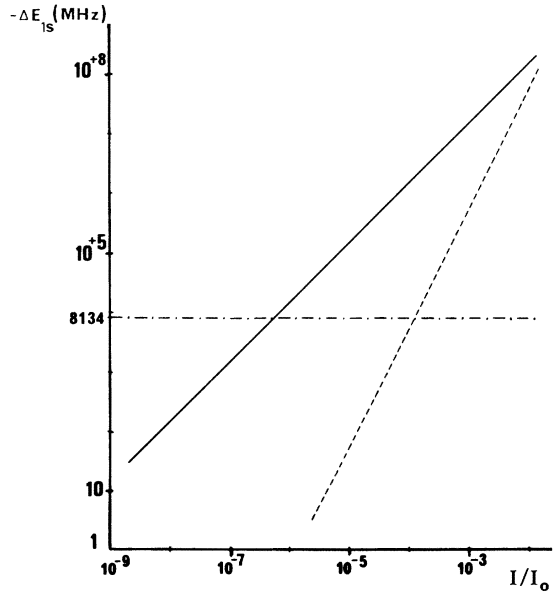

FIG. 2. Second-order (solid line) and fourth-order (dashed line) contributions to the level shift of the ground state in the presence of a monomode neodymium laser ($\omega=4.3\times10^{-2}$ a.u.) of intensity I. Here $I_0=7.016\times10^{16}$ W/cm^2. The horizontal dot-dashed line represents the Lamb shift $\simeq+8134$ MHz of the ground state.

We have also shown the usefulness of using Padé-related techniques for summing poorly convergent or even divergent multiple series. The question of generalizing the concept of Padé approximants to the case of multiple series is by no means a trivial one.[68] However, the success of our approach (see Table VII) shows that it is not hopeless.

As a by-product, we have given a Sturmian representation of the reduced Coulomb Green's function which should be useful in stationary perturbation-theory computations in hydrogen. Another interesting result was to show that approximate calculations, based on a partial summation of the discrete spectrum contribution, can lead to misleading estimations of the magnitude of the fourth-order amplitudes. We have also obtained an extension of the optical theorem to the case of multiphoton elastic scattering by relating the two-photon-ionization cross section to the imaginary part of the amplitude of four-photon forward scattering.

ACKNOWLEDGMENTS

We have greatly benefited from discussions with Y. Heno and S. Klarsfeld to whom we express our thanks. We are indebted also to B. Dubreuil and P. Pignolet who kindly checked several of our calculations.

APPENDIX A: CLOSED-FORM EXPRESSIONS FOR SECOND-ORDER RADIAL AMPLITUDES

Any second-order radial transition amplitude of the general form

$$T_\lambda^{(2)}(\omega)=\langle nl \mid rG_\lambda(E_n+\omega)r \mid nl\rangle, \quad \lambda=l\pm 1 \tag{A1}$$

may be rewritten as a series expansion in the variable

$$\zeta=\left[\frac{p_0-1/n}{p_0+1/n}\right]^2, \quad p_0=[-2(E_n+\omega)]^{1/2} .$$

We have

$$T_\lambda^{(2)}(\omega)=\sum_{r=0}^{\infty} a_r\zeta^r , \tag{A2}$$

where a_r depends in a complicated way on the parameters ω, n, l, and λ [see Eqs. (30)–(34)], and on the index r. The important point is to note that a_r contains polynomials of order $p\le(n-l)^2$ *in the summation index* r, which implies that after some algebra, $T_\lambda^{(2)}(\omega)$ can always be expressed in terms of a linear combination of sums of the following general form:

$$S(p,q;\alpha;\zeta)=\sum_{r=0}^{\infty} r^p\frac{(r+q)!}{(r+\alpha)}\frac{\zeta^r}{r!} , \tag{A3}$$

where p and q are positive integers and α and ζ are complex numbers. These series obey the recurrence relation

$$S(p,q;\alpha;\zeta)=R(p-1,q;\zeta)-\alpha S(p-1,q;\alpha;\zeta) , \tag{A4}$$

where

$$R(p',q;\zeta)=\sum_{r=0}^{\infty} r^{p'}\frac{(r+q)!}{r!}\zeta^r , \tag{A5}$$

which, in turn, obey another simple recurrence relation:

$$R(p',q;\zeta)=R(p'-1,q+1;\zeta) -(q+1)R(p'-1,q;\zeta) \tag{A6}$$

with

$$R(0,q;\zeta)=\sum_{r=0}^{\infty}(r+q)!\zeta^r/r! =q!(1-\zeta)^{-(q+1)} . \tag{A7}$$

Thus the series $S(p,q;\alpha;\zeta)$ can be recurrently expressed as a sum of algebraic terms of the form (A7), plus *one* term proportional to $S(0,p;\alpha;\zeta)$:

$$S(0,p;\alpha;\zeta)=\sum_{r=0}^{\infty}\frac{(r+q)!}{(r+\alpha)}\frac{\zeta^r}{r!} =\frac{q!}{\alpha}{}_2F_1(\alpha,q+1;\alpha+1;\zeta) \tag{A8}$$

$$=\frac{q!}{\alpha}(1-\zeta)^{-q}{}_2F_1(1,\alpha-q;\alpha+1;\zeta) . \tag{A9}$$

Consequently, the amplitude $T_\lambda^{(2)}(\omega)$ may be eventually expressed as a linear combination of algebraic factors, plus one term containing a Gauss hypergeometric function.

In the particular case of a second-order amplitude involving the ground state, i.e.,

$$T_1(\omega)=\langle 10 \mid rG_1(\omega-\tfrac{1}{2})r \mid 10\rangle ,$$

$$\zeta=\left[\frac{p_0-1}{p_0+1}\right]^2, \quad p_0=(1-2\omega)^{1/2} ,$$

one gets, for instance,

$$T_1(\omega)=\frac{12}{(1-p_0^2)^2}\left[\frac{p_0^2}{2}-1+\frac{(p_0+1)^{-8}}{(2p_0-1)}\,{}_2F_1\left[2-\frac{1}{p_0},4;3-\frac{1}{p_0};\zeta\right]\right], \tag{A10}$$

which corresponds to Gavrila's result.[55]

Generalization to any second-order bound-bound transition amplitude between states $|n,l\rangle$ and $|n',l'\rangle$ is straightforward, though leading to cumbersome calculations. It should be stressed, however, that it is by far more convenient, from a practical point of view, to directly compute the sum, Eq. (A2), which usually converges very well. When it is not so (see Sec. IV), one can safely use the ϵ algorithm for constructing convergent sequences of Padé approximants (converging faster towards the exact result) which represent the analytical continuation of the series (A2). This is one of the main advantages we have encountered in using the Sturmian representation of the Coulomb Green's function in such computations.

APPENDIX B: STURMIAN EXPANSION FOR THE REDUCED COULOMB GREEN'S FUNCTION

We start from the definition[62,63]

$$G_\lambda^{(n)}(E_n)=\frac{\partial}{\partial E}(E-E_n)G_\lambda E\Big|_{E=E_n}=\left[G_\lambda(E)+(E-E_n)\frac{\partial}{\partial E}G_\lambda(E)\right]_{E=E_n}. \tag{B1}$$

On using the orthogonality properties of the eigenfunctions of $H_{\rm at}$, one has

$$\frac{\partial}{\partial E}G_\lambda(E)=\sum_{n'}-\frac{|n'\lambda\rangle\langle n'\lambda|}{(E-E_{n'})^2}=-[G_\lambda(E)]^2 \tag{B2}$$

and replacing $G_\lambda(E)$ by its Sturmian expansion, Eq. (28), one gets

$$\frac{\partial}{\partial E}G_\lambda(E)=-\sum_{\nu=\lambda+1}^{\infty}\sum_{\nu'=\lambda+1}^{\infty}\frac{|S_{\nu,\lambda}(p_0)\rangle\langle S_{\nu,\lambda}(p_0)|\,S_{\nu',\lambda}(p_0)\rangle\langle S_{\nu',\lambda}(p_0)|}{(1-\nu p_0)(1-\nu' p_0)}. \tag{B3}$$

The next step is to note that the Sturmian functions *are not orthogonal,* and that the scalar product reads[69]

$$\langle S_{\nu,\lambda}(p_0)\,|\,S_{\nu',\lambda}(p_0)\rangle=p_0^{-1}[\nu\delta_{\nu\nu'}-\tfrac{1}{2}[(\nu-\lambda-1)(\nu+\lambda)]^{1/2}\delta_{\nu',\nu-1}-\tfrac{1}{2}[(\nu-\lambda)(\nu+\lambda+1)]^{1/2}\delta_{\nu',\nu+1}]. \tag{B4}$$

Then one gets easily

$$\frac{\partial}{\partial E}G_\lambda(E)=-p_0^{-1}\sum_{\nu=\lambda+1}^{\infty}\left[\nu\frac{|S_{\nu,\lambda}(p_0)\rangle\langle S_{\nu,\lambda}(p_0)|}{(1-\nu p_0)^2}-\frac{1}{2}\frac{(\nu-\lambda-1)(\nu+\lambda)\,|S_{\nu,\lambda}(p_0)\rangle\langle S_{\nu-1,\lambda}(p_0)|}{(1-\nu p_0)[1-(\nu-1)p_0]}\right.$$
$$\left.-\frac{1}{2}\frac{(\nu-\lambda)(\nu+\lambda+1)\,|S_{\nu,\lambda}(p_0)\rangle\langle S_{\nu+1,\lambda}(p_0)|}{(1-\nu p_0)[1-(\nu+1)p_0]}\right]. \tag{B5}$$

This is the form we have used for expressing the second-order amplitude entering the expression of $(\partial/\partial E)\Delta E_n^{(2)}$, Eqs. (41). The reduced Coulomb Green's function is then obtained by inserting (B5) into (B1) and taking the limit $E=E_n$, i.e., $p_0=1/n$. The remaining finite result is given in Eq. (40). Note that, if we consider the special case of the ground state, the reduced function becomes

$$G_0^{(1)}(-\tfrac{1}{2})=\sum_{\nu=2}^{\infty}\frac{S_{\nu,0}(r)S_{\nu,0}(r')}{1-\nu}-2e^{-(r+r')}[5-2(r+r')]. \tag{B6}$$

As the Sturmian functions can be expressed in terms of Laguerre polynomials, this may be rewritten as follows:

$$G_0^{(1)}(-\tfrac{1}{2})=-2e^{-(r+r')}\left[2\sum_{\nu=1}^{\infty}\frac{L_\nu^1(2r)L_\nu^1(2r)}{\nu(\nu+1)}+5-2(r+r')\right], \tag{B7}$$

where $L_\nu^\mu(r)$ are Laguerre polynomials. By comparing with the known representation[62,63]

$$G_0^{(1)}(-\tfrac{1}{2})=2e^{-(r+r')}\left[2\ln(4rr')+2(r+r')-7-\left[\frac{1}{r}+\frac{1}{r'}\right]+4\gamma+\frac{e^{2r_<}}{r_<}-2\,\mathrm{Ei}(2r_<)\right], \tag{B8}$$

one gets the nice summation formula

$$\sum_{\nu=1}^{\infty}\frac{L_\nu^1(2r)L_\nu^1(2r')}{\nu(\nu+1)}=1-2\gamma-\ln(4rr')+\frac{1}{2r}+\frac{1}{2r'}-\frac{e^{2r_<}}{2r_<}+\mathrm{Ei}(2r_<), \tag{B9}$$

where $r_<$ is $\inf(r,r')$, $\gamma=0.5772\ldots$ denotes Euler's constant and $\mathrm{Ei}(2r_<)$ is the exponential integral function

$$\mathrm{Ei}(-x)=-\int_x^{+\infty}e^{-t}t^{-1}dt\ .$$

By considering reduced Green's functions for other atomic states, one could obtain more general formulas, similar to those given by Erdelyi and others.[70]

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