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Exact summation of higher-order terms in multiphoton processes

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The summation of all the higher-order contributions to multiphoton absorption in atoms is presented. The nonperturbative expressions for the transition amplitudes are expressed in terms of continued fractions. They account for the whole perturbation originated by the radiation field on the atom but do not exhibit explicit expression for the level shift. In the case of radiation fields of high intensity the only reliable determination of the level shift involves a careful location of the poles of the resolvent operator. The theory has been tested in considering a two-level atom. An extremely rapid convergence of the continued fractions has been observed, and accurate results concerning high-order resonance shifts have been obtained. The expressions for the transition amplitudes being nonperturbative with regard to the field strength, they can be considered as analytic continuations of the perturbation series. This provides a useful approach to the problem of ultraintense fields.

I. INTRODUCTION

The domain of physics concerned with the interaction of high-intensity radiation fields with atomic systems has provided a great amount of theoretical works,[1-10] principally during the past decade. Such a renewal of interest finds its origin in the recent advent of high-power lasers which make it possible to observe highly nonlinear absorption processes.

Most of quantitative analysis of the phenomena originated by this interaction make intensive use of perturbation techniques. One usually considers the situation where the dominant contribution to a determined process is provided by the lowest-order nonvanishing term of the perturbation series. In this case a rapid convergence of the series is needed. Such a requirement implies the smallness of the interaction energy compared to the total energy of the system. Within this approximation the higher-order terms of the perturbation series are neglected.

For an Nth-order process, these terms are obtained in considering all possible combinations of arbitrary number of absorption and emission of photons in such a way that the net number of absorptions remains equal to N. There are two circumstances where the higher-order terms must not be ignored. First, there is the possibility of resonances in the intermediate states. In that case, even for low-intensity radiation field, the lowest-order term is not sufficient to reproduce the experimental results. Such a problem was investigated in calculating a renormalized Green's function through a Schwinger-Dyson equation.[9,10] The mass operator appearing in this equation was chosen in order to account for single or multiple forward scattering of photons of the radiation beam. This method, which finds its inspiration in the quantum electrodynamics method of Low,[11] is not satisfactory when real photons are involved. For example, diagrammatically, it is straightforward to see that the presence of two such adjacent (on both sides of a vertex) renormalized internal electron lines gives rise to multiple counting of diagrams. On the other hand, there is undercounting, i.e., certain classes of diagrams never appear. Nevertheless, the merit of such calculations was to point out the central role played by the higher-order terms in resonant process.

Secondly, with the increase of laser intensity, physicists will be faced, in the near future, with the problem of handling radiation fields whose electric component will be comparable to or larger than the interatomic Coulomb field. This situation outlines the other case for which the use of the lowest-order term of the perturbation series cannot give a reliable description of absorption phenomena since the perturbation series may not converge rapidly or even diverges.

As the amplitude of the external field increases the contribution of the higher-order terms becomes less and less negligible. With ultrastrong fields one may reside outside the radius of convergence of the series and a transition amplitude cannot be expressed in terms of such a series.

The aim of this paper is to propose a solution to this problem. To this end we present a method used to obtain the exact expression of the transition amplitudes. This method is based on the exact summation of the whole series of the higher-order terms in a region where the convergence is good (low intensity). This permits analytic continuation to the domain of very intense fields where the series is not convergent. This would be another approach to the problem of ionization of atoms by very intense fields. The idea of analytic continuation of transition amplitudes via the summation of the series of higher-order terms was proposed few years ago for the investigation of Compton scattering in intense field.[12,13]

The problem of bound electrons we consider is somewhat more complicated and requires a more-general treatment. The reason is that, in contrast to the free-electron case, the sums over the intermediate atomic states occur. The continued fractions representing the transition amplitudes are expressed in terms of noncommuting operators. This precludes a simple representation of these fractions in terms of transcendental functions.

On the other hand, for absorption processes involving more than two photons the use of the exact electron renormalized Green's function,[14] which takes into account all orders of forward scattering of real photons is not suitable. The reason can be found from the reading of this paper. One of the salient features of the work presented here is that the renormalized propagation function to be considered in a N-photon absorption ($N > 2$), is different from the one which could be determined through a Schwinger-Dyson equation involving the exact mass operator, i.e., the mass operator representing the sum of all proper diagrams of forward scattering.

No numerical result concerning atoms with more than two levels are presented here, firstly, to keep the paper focused to the technique utilized, and secondly, to have ample space and opportunity to discuss numerical results in future publications. Some idea of the numerical results, however, may be sampled in the previously published accounts.[15]

II. THEORETICAL PRELIMINARIES

A. General considerations

Some of the definitions adopted throughout this paper have been already employed in the case of free electrons.[12] Therefore, we shall avoid an unnecessary dispersion of notations concerning the problem of interaction of an electron (bound or free) with a radiation field.

Since we deal with radiation fields of optical frequencies the dipole approximation is made. In addition we suppose that the optical electron of the atom is spinless and is described by the Schrödinger equation in the presence of an external field

$$[H_A + H_F - \vec{E}(t)\cdot\vec{r}]\,\psi(t) = i\dot{\psi}(t)\,, \tag{2.1}$$

where H_A and H_F are the Hamiltonians of the free atom and of the field, respectively, and $\vec{E}(t)$ is the electric field. We note in passing that the interaction Hamiltonians $-\alpha\vec{A}(t)\cdot\vec{P} + \frac{1}{2}\alpha^2 A^2(t)$ and $-\vec{E}(t)\cdot\vec{r}$ differ, in the dipole approximation, by a gauge transformation. The radiation field is a single-mode field and has an occupation-number representation.

Since the problem of summation we consider has been handled by the use of the resolvent operator, Sec. II B is devoted to a brief summary of some properties regarding this operator.

B. Resolvent

Let $H = H^0 + V$ be the total Hamiltonian in the Schrödinger picture. H^0 is the sum of Hamiltonians of the free field and the atom, and V is the time-independent interaction Hamiltonian, given by

$$V = -\vec{E}(0)\cdot\vec{r}\,, \tag{2.2}$$

where $\vec{E}(0)$ the electric field, can be written

$$\vec{E}(0) = i[(2\pi/L^3)E_p]^{1/2}(\vec{\epsilon}a - \vec{\epsilon}^{\,*}a^\dagger)\,. \tag{2.3}$$

In Eq. (2.3), E_p is the photon energy, $\vec{\epsilon}$ is the complex polarization vector of the field, $a^\dagger$ and a are the creation and destruction operators for photons, respectively, and L^3 is the quantization volume.

It is convenient to write V as

$$V = V^+ + V^-\,, \tag{2.4}$$

where V^+ and V^- account, respectively, for the emission and the absorption of a photon. These operators can be determined from Eqs. (2.2) and (2.3). At the time origin, the interaction of the radiation field with the atom is supposed to be switched off. The state of the system is then eigenstate of H^0 and can be written

$$|a\rangle = |g\rangle \otimes |n\rangle\,, \tag{2.5}$$

where $|g\rangle$ is the atomic state and $|n\rangle$ is the state of the field characterized by the presence of n photons in the single mode under consideration.

At a later time, when the interaction is switched on, the state of the system is determined from the evolution operator $U(t)$. From the matrix element of this operator, one determines the transition amplitude through the equation

$$A_{fi}(t) = \langle f|U(t)|i\rangle\,, \tag{2.6}$$

where $|i\rangle$ and $|f\rangle$, the initial and final states of the system, are the eigenstates of H^0 shown in Eq. (2.5).

The problem concerning the calculation of this time-evolution operator can be formulated by the use of the resolvent.[16] The resolvent $G(E)$ of the total Hamiltonian H is by definition

$$G(E) = 1/(E - H). \tag{2.7}$$

It can be shown that the following simple relationship exists between the time-evolution operator of the Schrödinger picture and the resolvent:

$$U(t)=\frac{1}{2\pi i}\oint G(E)e^{-iEt}dE\,. \tag{2.8}$$

The contours of integration for positive and negative time are shown in Fig. 1.

From Eqs. (2.7) and (2.8) it seems to be straightforward to calculate a matrix element of $U(t)$. To this end one has to perform the inversion of the matrix $(E-H)$. Unfortunately, even for low-intensity radiation fields, the prohibitive size of the matrix to be inverted precludes such a procedure. The aim of this paper is to present a feasible method for the exact calculations of the matrix elements of $G(E)$. This method is based on the summation of all the contributions to a definite ionization process. First we write the resolvent operator in the equivalent form

$$G(E)=G^0(E)+G^0(E)VG(E)\,, \tag{2.9}$$

where

$$G^0(E)=1/(E-H^0)\,. \tag{2.10}$$

From Eq. (2.9) it is clear that $G(E)$ can be expressed on the form of an infinite series in V, the interaction,

$$G(E)=G^0(E)+G^0(E)VG^0(E)+G^0(E)VG^0(E)VG^0(E)+\cdots\,. \tag{2.11}$$

Taking the inner product of both sides of Eq. (2.11) one obtains the probability amplitude for a transition which takes place between the states $|a\rangle$ and $|b\rangle$. For well-defined initial and final states, only a certain class of terms contained in the series shown in Eq. (2.11) is relevant. The energy conservation in Eq. (2.8) implies such a selection among the terms. Starting from a definite state of the field, only the terms leading to the correct final state do not vanish, i.e., only channels accounting for a definite depletion of photons of the beam do not vanish. For example, in the case of one-photon ionization the series of Eq. (2.11) reduces to

$$G(E)=G^0V^-G^0+G^0V^+G^0V^-G^0V^-G^0+G^0V^-G^0V^+G^0V^-G^0+G^0V^-G^0V^-G^0V^+G^0+\cdots\,. \tag{2.12}$$

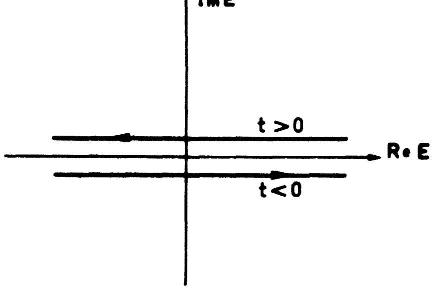

FIG. 1. Contours of integration for the integral of Eq. (2.8).

The first term in the right-hand side of Eq. (2.12) is the lowest-order term, the others are the higher-order terms previously mentioned.

The tractability of the problem we consider is greatly improved by a diagrammatic representation. Since intensive use of such representation will be made throughout this paper, Sec. IIC is devoted to the notations we use.

C. Diagrammatic representation

Even for a one-photon absorption, Eq. (2.12) shows that as the perturbation order increases the number and the size of the terms of the series representing $G(E)$ increase. Therefore, a graphical representation of the terms of this series will be very useful for a better understanding of our method of summation. In the diagrammatic representation we utilize, the electron line is vertical. Absorbed and emitted photons are represented by horizontal lines on the left- and right-hand side of the electron line, respectively.

The equivalence between a diagram and an analytic expression holds if the former is read from bottom upwards while the latter is read from right to left. The part of the electron line lying on both sides of any vertex represents the operator G^0. Utilizing these notations the four terms in the right-hand side of Eq. (2.12) can be represented by the diagrams shown in Fig. 2. A diagram is *proper* if it cannot be divided into two parts, one of them showing a net number of absorbed (or emitted) photons equals to zero; otherwise it is called improper. The *mirror* diagram of a diagram is obtained in replacing the incoming photon lines by outgoing photon lines and vice versa.[12]

For the sake of convenience the following operators will be intensively used:

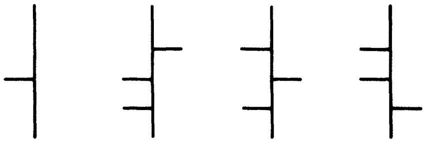

FIG. 2. Diagrammatic representation of the first four terms of the series of Eq. (2.12). The electron is described by vertical line, whereas absorbed and emitted photons are represented by horizontal lines on the left-hand side and on the right-hand side of the electron, respectively.

$$A = G^0 V^- , \tag{2.13a}$$

$$B = G^0 V^+ . \tag{2.13b}$$

A and B describe the absorption and the emission of a photon, respectively. In analogy with the case of a free electron they are called propagation-vertex operators. To end this subsection we give now the practical rules for writing down the analytic expression, in atomic units, for the transition amplitude related to a diagram. This amplitude can be obtained in considering the following factors: (a) a factor $(FE_p/F_0)^{1/2}\vec{r}\cdot\vec{\epsilon}$ for each absorption vertex; (b) a factor $(FE_p/F_0)^{1/2}\vec{r}\cdot\vec{\epsilon}^{\,*}$ for each emission vertex; for convenience F is the photon flux in cgs units, $F_0 = c/(2\pi a_0^3)$, a_0 being the Bohr radius and c is the light velocity; E_p is the photon energy (in a.u.), and $\vec{\epsilon}$ is the polarization vector of the field; (c) a factor

$$\sum_i \frac{|i\rangle\langle i|}{E_0 + nE_p - E_i}$$

for each internal line. E_0 is the energy of the initial atomic state and n is the net number of absorbed (or emitted) photons up to this line. The summation runs over the whole spectrum of the atom, discrete plus continuum; (d) finally, the inner product with the initial and the final atomic states of the expression so obtained is to be taken.

III. SUMMATION OF DIAGRAMS

In this section the method used for the summation of all the higher-order terms of the perturbation series is presented. It is based firstly on a systematic search of these terms and secondly on a suitable association of them which makes apparent the form of the generating function from which they can be derived up to an arbitrary order.

We begin by discussing the summation procedure for forward-scattering diagrams. Later it will be shown how the results of forward scattering can be incorporated into the scheme of calculation of photon-absorbing processes.

A. Forward-scattering diagrams

The forward-scattering diagrams can be divided into two classes: proper diagrams and improper diagrams (see Figs. 3 and 4). All of them are obtained in considering all possible combinations of arbitrary number of absorptions and emissions of photons in such a way that the number of absorptions and emissions are equal. In analogy with the free-electron case these diagrams will be also called self-energy diagrams.

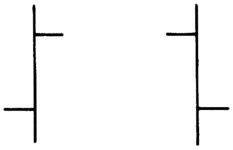

FIG. 3. Lowest-order proper forward-scattering diagrams.

Combining Eqs. (2.9) and (2.13) we find that the resolvent operator can be written

$$G(E) = G^0(E) + (A + B)G(E). \tag{3.1}$$

The iteration of this equation shows that $G(E)$ can be expressed on the form of an infinite series in terms of A and B,

$$G(E) = G^0(E) + (A + B)G^0 E + (A + B)(A + B)G^0(E) + \cdots . \tag{3.2}$$

Since $G(E)$ must account for no net absorption or emission of photons, the only relevant terms in this series are those that show an equal number of A and B operators. Thus $G(E)$ reduces to

$$G(E) = G^0(E) + [AB + BA]\, G^0(E) + [AABB + ABAB + ABBA + BAAB + BABA + BBAA]\, G^0(E) + \cdots . \tag{3.3}$$

On the other hand, most of the terms of this series represent improper diagrams. For example, only the first and the last term in the second square bracket on the right-hand side of Eq. (3.3) represent proper diagrams; the other four are improper. This is illustrated in Fig. 4 where it is shown that four of the six fourth-order diagrams can be cut into two lower-order self-energy diagrams.

It is a simple matter to show that all the (proper and improper) terms of the series of Eq. (3.3) can be obtained from the operator

$$\tau_{\square}(E) = 1/[1 - M^{\infty}(E)], \tag{3.4}$$

by expansion.

In analogy with the well-known theory of renormalization of quantum electrodynamics,[17] $M^{\infty}(E)$ is the sum of all the proper self-energy diagrams. For example, up to the fourth order all the terms

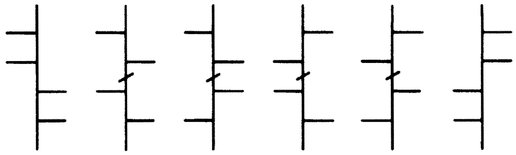

FIG. 4. Fourth-order forward-scattering diagrams. In each improper diagram the slash indicates where the cut must be made to obtain lower-order proper diagrams.

of the series of Eq. (3.3) are given by substituting in Eq. (3.4) the operator

$$M^{(4)}(E)=AB+BA+AABB+BBAA\,. \tag{3.5}$$

To have all the terms of Eq. (3.3) up to the sixth order, one must consider the additional contributions provided by the sixth-order proper self-energy diagrams symbolically represented by the four terms $AAABBB$, $AABABB$, $BBABAA$, and $BBBAAA$. Thus, in Eq. (3.4) the following operator is to be considered:

$$M^{(6)}=AB+BA+AABB+BBAA$$
$$+AAABBB+AABABB+BBBAAA+BBABAA\,. \tag{3.6}$$

In principle, the process can be continued up to arbitrary order. In practice, a serious limitation due to the increasing complexity of the operator $M^{(i)}(E)$ arises. The crux of the problem now is to determine the sum of all the proper self-energy diagrams. From the knowledge of $M^{\infty}(E)$ an expression for the electron propagator in the presence of the electromagnetic field can be found. Thus from Eqs. (3.3) and (3.4) one has

$$G(E)=\{1/[1-M^{\infty}(E)]\}\,G^0(E)=\tau_{\Box}(E)G^0(E)\,. \tag{3.7}$$

With a slight change in the notation, i.e., $M^{\infty}(E)=G^0(E)\Sigma(E)$, this expression for $G(E)$ becomes more familiar. It is widely encountered in the literature.

Now we focus our attention on the elaboration of a method which enables one to find in a convenient way all the proper self-energy diagrams. The labor is somewhat reduced by noting that there are two classes of such diagrams. One of them contains all the diagrams for which the first vertex depicts a photon absorption the other is concerned with all the diagrams whose first vertex depicts a photon emission. These two classes of diagrams can be investigated separately. It is straightforward to show that for each diagram belonging to a definite class, one can find the corresponding mirror diagram in the other class.

In the following, we consider only the proper diagrams of the first class, i.e., diagrams beginning with a photon absorption. All the terms of the perturbation series of Eq. (3.3) representing the proper self-energy diagrams up to the fourteenth-order are written in Table I. The sixteenth-order terms are shown on Table II in order to supply enough information for the elaboration of the general law from which these terms can be determined and then summed.

The three operators T, X, and Y are defined by

TABLE I. Terms beginning by a photon absorption contributing to forward scattering up to fourteenth order.

Order 2

AB

Order 4

XY

Order 6

XTY

Order 8

XT^2Y
$XXYY$

Order 10

XT^3Y
$XTXYY$
$XXTYY$
$XXYTY$

Order 12

XT^4Y
XT^2XYY
$XTXTYY$
$XTXYTY$
XXT^2YY
$XXTYTY$
$XXYT^2Y$
$XXXYYY$
$XXYXYY$

Order 14

XT^5Y
XT^3XYY
XT^2XTYY
XT^2XYTY
$XTXT^2YY$
$XTXTYTY$
$XTXYT^2Y$
XXT^3YY
XXT^2YTY
$XXTYT^2Y$
$XXYT^3Y$
$XTXXYYY$
$XXTXYYY$
$XXXTYYY$
$XXXYTYY$
$XXXYYTY$
$XTXYXYY$
$XXTYXYY$
$XXYTXYY$
$XXYXTYY$
$XXYXYTY$

$$T=AB+BA\,, \tag{3.8a}$$
$$X=AA\,, \tag{3.8b}$$
$$Y=BB\,; \tag{3.8c}$$

this reduced notation and principally the use of the operator T enables one to write all the contributions up to arbitrary order without a prohibitive use of space. Apart from the order two which is expressed only in terms of A and B, all the contributions are special combinations of the three operators defined in Eqs. (3.8). X, Y, and T are quadratic operators in the electric field and all the terms begin with an X and end in a Y. For each order the term containing the largest power of T is first considered. For example the highest power for T admissible for the sixth order is 1, that for the eighth order is 2, etc. In the next step, the power of T is decreased by two units and a couple of operators XY or YX is inserted. The remaining operators T are combined in all possible ways with the inner X and Y operators. This process is continued as many times as necessary in order to reach the lowest non-negative power of T. The last point to be examined is the way by which the couples of operators XY or YX appear as the power of T is decreased.

TABLE II. Sixteenth-order terms beginning by a photon absorption contributing to forward scattering.

Order 16	
XT^6Y	$XXXTYTYY$
XT^4XYY	$XXXTYYTY$
XT^3XTYY	$XXXYT^2YY$
XT^3XYTY	$XXXYTYTY$
XT^2XT^2YY	$XXXYYT^2Y$
XT^2XTYTY	XT^2XYXYY
XT^2XYT^2Y	$XTXTYXYY$
$XTXT^3YY$	$XTXYTXYY$
$XTXT^2YTY$	$XTXYXTYY$
$XTXTYT^2Y$	$XTXYXYTY$
$XTXYT^3Y$	XXT^2YXYY
XXT^4YY	$XXTYTXYY$
XXT^3YTY	$XXTYXTYY$
XXT^2YT^2Y	$XXTYXYTY$
$XXTYT^3Y$	$XXYT^2XYY$
$XXYT^4Y$	$XXYTXTYY$
XT^2XXYYY	$XXYTXYTY$
$XTXTXYYY$	$XXYXT^2YY$
$XTXXTYYY$	$XXYXTYTY$
$XTXXYTYY$	$XXYXYT^2Y$
$XTXXYYTY$	$XXYXXYYY$
XXT^2XYYY	$XXXYXYYY$
$XXTXTYYY$	$XXXXYYYY$
$XXTXYTYY$	$XXXYYXYY$
$XXTXYYTY$	$XXYXYXYY$
$XXXT^2YYY$	

It is easy to see that all the terms of order $2P$ ($P = 1, 2, \ldots$) with nonvanishing power of T are deduced from the terms of order $2(P-1)$ in considering all the permutations of the inner X and Y operators occurring in the terms of order $2(P-1)$ with a number of operators T increased by one unit. For example, tenth-order terms are obtained in considering all the inner permutations of T^3 with XY and of T with $XXYY$. This gives XT^3Y and $XTXYY$, and $XXTYY$ and $XXYTY$.

From this last remark it is straightforward to see how the couples of operators XY and YX occur in the T-dependent terms.

The remaining terms containing only the X and Y operators are easily determined if we define the following operators:

$$t = XY + YX\ , \tag{3.9a}$$

$$x = XX\ , \tag{3.9b}$$

$$y = YY\ . \tag{3.9c}$$

We observe that the twelfth-order and the sixteenth-order terms of this kind can be written xty and $xt^2y + xxyy$, respectively. Comparing these contributions to those of the sixth- and eighth-order we see in a simple way that the terms of order $2N$ containing no operator T can be determined from the terms of order N by performing the substitutions $T \to t$, $X \to x$, and $Y \to y$.

The above rules allow one to write down all the terms of arbitrary order. The last part of the work is concerned with the summation of these terms. The sum of all the proper forward-scattering diagrams is found from the observation of the structure of the terms shown in Tables I and II. It is clear that the sum of the terms up to fourth order is

$$M_-^{(4)} = AB + XY\ . \tag{3.10a}$$

The sum of the terms up to sixth order is given by

$$M_-^{(6)} = AB + X[1/(1-T)]\,Y\ , \tag{3.10b}$$

where an expansion of the fraction is to be made and only the terms of order less than or equal to six are retained. In considering the next order we find that the sum of the terms up to the eighth order is given by

$$M_-^{(8)} = AB + X[1-(T+XY)]^{-1}Y\ , \tag{3.10c}$$

where the expansion of the fraction is performed up to eighth order. Continuing in this fashion, we find that the sum of all the proper forward-scattering diagrams, beginning with a photon absorption, up to the tenth order is

$$M_-^{(10)} = AB + X\{1-[T+X(1-T)^{-1}Y]\}^{-1}Y\ . \tag{3.10d}$$

The sum of such diagrams up to the twelfth order is given by

$$M_-^{(12)} = AB + X\frac{1}{1-\{T+X[1-(T+XY)]^{-1}Y\}}\,Y\ , \tag{3.10e}$$

and for the fourteenth order one has

$$M_-^{(14)} = AB + X\frac{1}{1-(T+X\{1-[T+X(1-T)^{-1}Y]\}^{-1}Y)}\,Y\ . \tag{3.10f}$$

From Eqs. (3.10), the expression for M_-^∞ can be derived by noting a periodicity as the successive order of M_- are considered. One finds that

$$M_-^\infty = AB + X\Pi^- Y\ , \tag{3.11a}$$

where

$$\Pi^- = [1-(T+X\Pi^- Y)]^{-1}\ . \tag{3.12a}$$

The class of forward-scattering diagrams beginning with a photon emission can be treated by the same procedure. The sum of such diagrams is

$$M_+^\infty = BA + Y\Pi^+ X\ , \tag{3.11b}$$

with

$$\Pi^+ = [1-(T+Y\Pi^+ X)]^{-1}\ , \tag{3.12b}$$

and M^{∞} is given by

$$M^{\infty} = M^{\infty}_{-} + M^{\infty}_{+} . \tag{3.13}$$

It must be pointed out that the expression for M^{∞} can be written on an alternative form which permits a comparison with previous results concerning free electrons. As it is shown in the Appendix, M^{∞} can be written

$$M^{\infty} = A\tau_0 B + B\tau_{\phi} A , \tag{3.14}$$

where

$$\tau_0 = (1 - A\tau_0 B)^{-1} \tag{3.15a}$$

and

$$\tau_{\phi} = (1 - B\tau_{\phi} A)^{-1} . \tag{3.15b}$$

A comparison with the results obtained in Ref. 12 can be done if in each denominator the A and B operators are replaced by $x_n^{(\mathrm{abs})}$ and $x_n^{(\mathrm{em})}$, respectively. The value of the index n corresponding to a particular denominator is determined in considering that the A (B) operator increases (decreases) by one unit the number of photons involved up to this denominator. For example, with these notations, the operator M^{∞} for a free electron is

$$M^{\infty} = \cfrac{x_0 \bar{x}_1}{1 - \cfrac{x_1 \bar{x}_2}{1 - \cfrac{x_2 \bar{x}_3}{1 - \cdots}}} + \cfrac{\bar{x}_0 x_{-1}}{1 - \cfrac{\bar{x}_{-1} x_{-2}}{1 - \cfrac{\bar{x}_{-2} x_{-3}}{1 - \cdots}}} , \tag{3.16}$$

where use has been made of the fact that $x_n^{(\mathrm{em})} = \bar{x}_n^{(\mathrm{abs})} \equiv \bar{x}_n$. From Eq. (3.16) it is clear that the self-energy of a free electron calculated in Ref. 12 and the one deduced by our method are identical (remember that $\Sigma = G^0 M^{\infty}$). This agreement was expected since our derivation is general and holds for bound and for free electrons.

If we note that the number of proper forward-scattering diagrams of order $2k$ is $\binom{2k}{k}/(2k-1)$, it is clear that the list of the 264 terms of the fourteenth order, that of the 858 terms of the sixteenth order, etc., is considerably shortened by the use of the operators X, Y, and T. In addition this reduced notation has permitted to find in a simple manner the laws for the determination of all the proper self-energy diagrams. The last remark we can formulate regarding the use of such a notation is that the direct determination of M^{∞} on the form shown in Eq. (3.14) is not straightforward. For these reasons the operators X, Y, and T will be used throughout this paper in spite of the fact that the resulting formulas can be reduced to a more compact form.

B. One-photon ionization

The procedure previously used for the summation of the self-energy diagrams naturally holds for the determination of the multiphoton ionization amplitudes. As before we could search and associate in a suitable way the relevant terms of the perturbation series but this method is not the most suitable in the present case. The reason is that no reliable expression for the sum can be obtained without considering sufficient number of forward-scatttering processes in each internal electron line. This implies handling more and more terms of increasing orders as the number of absorbed photons increases. We would be rapidly faced with a tedious problem of diagram classification. Here it is instructive to note that for a net absorption of N photons the number of diagrams of order $N+2k$ is given by the binomial coefficient $\binom{N+2k}{k}$.

The alternate method we discuss now makes intensive use of the results of Sec. III A. It is first applied to one-photon ionization. Later it will be generalized to multiphoton absorption. The basic idea is that the operator $G(E)$ of Eq. (3.7), symbolically represented in Fig. 5(b) by a square inserted into an electron line, accounts for all the proper and improper forward-scattering diagrams. Next we assume that such an operator acts on both sides of the single absorption operator A.

The corresponding transition amplitude can be written

$$\mathfrak{A}_{ab}^{(1)*}(E) = \frac{1}{2\pi i} \oint e^{-iEt} \langle a | \tau_{\square} A \tau_{\square} G_0 | b \rangle \, dE , \tag{3.17}$$

where it is understood that all the operators in the integrand depend on E.

The asterisk in the left-hand side of Eq. (3.17) indicates that the transition amplitude so defined contains multiple contributions of the same diagram. It is straightforward to check that this multiple counting occurs in the third-order contributions where the term $\langle a | ABAG_0 | b \rangle$ is counted twice.

Our aim now is to deduce from Eq. (3.17) the exact transition amplitude by eliminating in $\mathfrak{A}^{(1)*}$ all the extra contributions to the one-photon process. To this end we consider the following operator occurring in Eq. (3.17):

$$\Omega^{(1)*} = \tau_{\square} A \tau_{\square} . \tag{3.18}$$

For convenience we keep one of the $\tau_{\square}$ operators unchanged in Eq. (3.18) and eliminate from the other one the processes giving rise to multiple counting. This can be done without any loss of generality. The operator $\tau_{\square}$ located on the left-

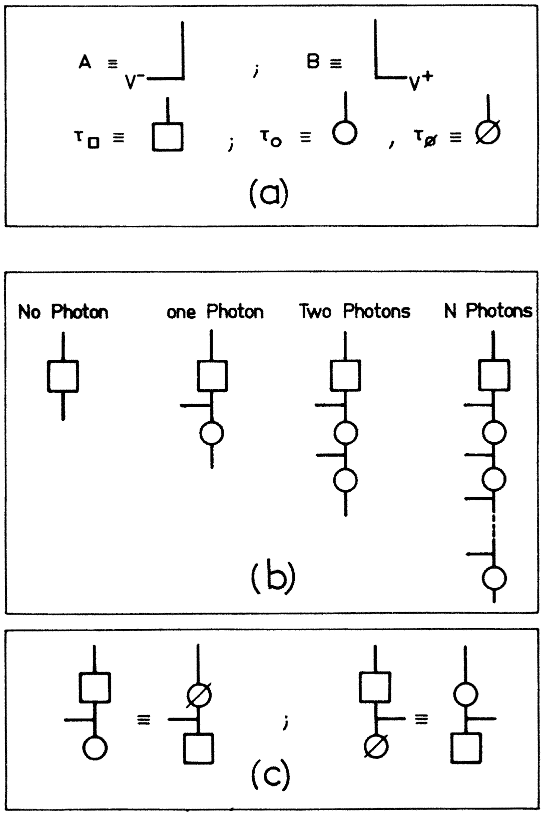

FIG. 5. (a) Diagrammatic representation of the operators A, B, $\tau_{\Box}$, τ_0, and τ_{ϕ}. The operator $G_0(E)$ appears everywhere on the form of a vertical line. (b) Graphical representation of $G(E)$ for multiple-photon absorption. (c) Graphical representation of identities of Eqs. (3.29a) and (3.29b), respectively.

hand side of A is chosen to be unchanged (in other words, the expression of $\Omega^{(1)*}$ will show, on the left-hand side of A the full expansion of $\tau_{\Box}$).

Our elimination procedure involves the knowledge of all the terms appearing multiple times in $\Omega^{(1)*}$. The search of such terms is a difficult task. In contrast to what was encountered for the summation of the self-energy diagrams, it is not straightforward to derive a law which enables one to find all the processes under consideration. For this reason another point of view has been adopted. Our approach to this problem is based on the use of a criterion for multiple counting. This criterion is easily derived by noting that in Eq. (3.18), multiple counting occurs every time the operator $\Omega^{(1)*}$ can be written in the form

$$\begin{aligned} &\Omega^{(1)*}=\tau_{\Box}(A\tau+\tau_i A\tau_j)\,, \\ &A\tau \not\Rightarrow \tau' A\,, \end{aligned} \tag{3.19}$$

where τ, τ', τ_i, and τ_j represent particular sets of forward scattering and τ_i excludes the identity operator (i.e., no absorbed and emitted photons).

The important point to be noted is that the last term in the right-hand side of Eq. (3.19) contains all the additional multiple contributions. Therefore, it is clear that its elimination leads to the correct operator $\Omega^{(1)}$ to be used for the calculation of the transition amplitude.

As a general rule, the terms of $\tau_{\Box}$ to be eliminated are the ones which can be partially or completely crossed from left to right by the operator A. This criterion can be easily derived by expanding the operators $\tau_{\Box}$ in Eq. (3.18). One obtains

$$\Omega^{(1)*}=(1+AB+BA+\cdots)A(1+AB+BA+\cdots)\,, \tag{3.20}$$

where only the terms up to second order have been retained. Remembering our previous convention regarding the invariance of the operator $\tau_{\Box}$ on the left-hand side of A, a little rearrangement of the terms can be done in Eq. (3.20) to give

$$\begin{aligned} \Omega^{(1)*}=&(1+AB+BA+\cdots) \\ &\times[A(1+AB+\cdots)+(AB+\cdots)A(1+\cdots)+\cdots]\,, \end{aligned} \tag{3.21}$$

which is a particular case of Eq. (3.19). One observes that ABA, the only third-order term which is counted twice, can be written

$$A(BA)\equiv(AB)A\,.$$

This illustrates what is meant by crossing: within the same term the operator A, which accounts for the net absorption of a photon, can be considered as acting prior or in succession to forward-scattering operators. It must be pointed out that only the terms giving rise to multiple counting possess this property.

More generally, from Eqs. (3.18), (3.4), and (3.13) one has

$$\Omega^{(1)*}=\tau_{\Box}A[1-(M^{\infty}_{+}+M^{\infty}_{-})]^{-1}\,. \tag{3.22}$$

By the use of the Feynman identity, Eq. (3.22) yields

$$\Omega^{(1)*}=\tau_{\Box}A\left(\frac{1}{1-M^{\infty}_{-}}+\frac{1}{1-M^{\infty}_{-}}M^{\infty}_{+}\frac{1}{1-(M^{\infty}_{-}+M^{\infty}_{+})}\right). \tag{3.23}$$

Owing to the obvious relations

$$M^{\infty}_{+}=B[1/(1-M^{\infty}_{+})]A\,, \tag{3.24a}$$

$$M^{\infty}_{-}=A[1/(1-M^{\infty}_{-})]B\,, \tag{3.24b}$$

Eq. (3.23) becomes

$$\Omega^{(1)*}=\tau_{\Box}\left(A\frac{1}{1-M^{\infty}_{-}}+M^{\infty}_{-}\frac{1}{1-M^{\infty}_{+}}A\frac{1}{1-(M^{\infty}_{+}+M^{\infty}_{-})}\right). \tag{3.25}$$

Further iterations are not necessary. Now we see that this expression for $\Omega^{(1)*}$ has the same form as the one shown in Eq. (3.19). More precisely, only the first term in parentheses has not been crossed by the operator A. From the criterion previously discussed only this term is relevant all the others are to be dropped since they give rise to multiple counting.

In comparing Eqs. (3.19) and (3.25) one finds that

$$\tau \equiv 1/(1-M_-^\infty) \equiv \tau_0 \tag{3.26}$$

and

$$\Omega^{(1)*} \to \Omega^{(1)} = \tau_\Box A \tau_0 . \tag{3.27}$$

Therefore, the exact one-photon absorption amplitude is

$$\mathfrak{A}_{ab}^{(1)}(t) = \frac{1}{2\pi i} \oint e^{-iEt} \langle a | \tau_\Box A \tau_0 G^0 | b \rangle \, dE . \tag{3.28a}$$

It can be checked that this expression for the transition amplitude accounts for all the contributions to one-photon ionization process, each of them being counted once time. To this end, all the terms up to the ninth order are shown in Tables III and IV.

It must be pointed out that an equivalent expression for $\mathfrak{A}_{ab}^{(1)}$ could be obtained in leaving unchanged the operator lying on the right-hand side of A in Eq. (3.18) i.e.,

$$\mathfrak{A}_{ab}^{(1)}(t) = \frac{1}{2\pi i} \oint e^{-iEt} \langle a | \tau_\phi A \tau_\Box G^0 | b \rangle \, dE . \tag{3.28b}$$

From Eqs. (3.28) it is straightforward to derive the useful relation

$$\tau_\Box A \tau_0 = \tau_\phi A \tau_\Box , \tag{3.29a}$$

and the one obtained in changing $A \to B$ and $B \to A$,

$$\tau_\Box B \tau_\phi = \tau_0 B \tau_\Box . \tag{3.29b}$$

TABLE III. Terms contributing to one-photon absorption up to seventh order, $T_1 \equiv AB$.

Order 1	Order 7
A	$T^3 A$
Order 3	$TXY\,A$
	$XTY\,A$
$T\,A$	$XYT\,A$
$A\,T_1$	$TYX\,A$
Order 5	$YTX\,A$
	$YXT\,A$
$T^2 A$	$T^2 A\,T_1$
$XY\,A$	$XY\,A\,T_1$
$YX\,A$	$YX\,A\,T_1$
$T\,A\,T_1$	$T\,A\,T_1^2$
$A\,T_1^2$	$T\,A\,XY$
$A\,XY$	$A\,T_1^3$
	$A\,T_1XY$
	$A\,XT_1Y$
	$A\,XYT_1$

TABLE IV. Nine-order terms contributing to one photon absorption, $T_1 \equiv AB$.

Order 9	
$T^4 A$	$T^2 A\,XY$
$T^2XY\,A$	$XY\,A\,XY$
$TXTY\,A$	$YX\,A\,XY$
$TXYT\,A$	$T\,A\,T_1^3$
$XT^2Y\,A$	$T\,A\,T_1XY$
$XTYT\,A$	$T\,A\,XTY$
$XYT^2 A$	$T\,A\,XYT_1$
$T^2YX\,A$	$A\,T_1^4$
$TYTX\,A$	$A\,T_1^2XY$
$TYXT\,A$	$A\,T_1XTY$
$YT^2X\,A$	$A\,T_1XYT_1$
$YTXT\,A$	$A\,XT^2Y$
$YXT^2 A$	$A\,XTYT_1$
$T^3 A\,T_1$	$A\,XYT_1^2$
$TXY\,A\,T_1$	$YYXX\,A$
$XTY\,A\,T_1$	$YXYX\,A$
$XYT\,A\,T_1$	$YXXY\,A$
$TYX\,A\,T_1$	$XYYX\,A$
$YTX\,A\,T_1$	$XYXY\,A$
$YXT\,A\,T_1$	$XXYY\,A$
$T^2 A\,T_1^2$	$A\,XYXY$
$XY\,A\,T_1^2$	$A\,XXYY$
$YX\,A\,T_1^2$	

In Eqs. (3.28) the effect of τ_0, τ_ϕ, and $\tau_\Box$ is to shift the poles exhibited by the lowest-order contribution to $\Omega^{(1)}$ obtained from Eq. (3.27) by expansion. Nevertheless the important point to be noted is that these operators cannot be interpreted as shift operators. The level shifts can only be determined through a consistent calculation of the poles of $\Omega^{(1)}(E)$. On the other hand, the asymmetry of this operator gives rise to serious difficulties if we want to define a renormalized state since in this case the renormalization would be different for the initial and the final states. For this reason, the transition amplitudes will be calculated in considering the matrix element of the operator $\Omega^{(i)}$ with the eigenstates of H_0.

Before concluding this subsection we note that the problem of the emission of a photon of the radiation field can be treated in the same way. One finds that the expression for the corresponding transition amplitude is now

$$\mathfrak{A}_{ab}^{(1)}(t) = \frac{1}{2\pi i} \oint e^{-iEt} \langle a | \tau_0 B \tau_\Box G^0 | b \rangle \, dE , \tag{3.30}$$

as is expected from time reversal invariance considerations.

C. *N*-photon ionization

The method discussed in Sec. III B can be used in the case of N-photon ionization. To show this, the absorption of two photons is first considered. It will be shown, at the end of this subsection, how the results obtained in this simple case can be generalized.

For this two-photon process we suppose (as was done previously) that all the electron lines are fully perturbed, i.e., we consider the following expression for the transition amplitude:

$$\mathcal{A}_{ab}^{(2)*}(t)=\frac{1}{2\pi i}\oint e^{-iEt}\langle a|\tau_{\square}A\tau_{\square}A\tau_{\square}G^0\,|b\rangle\,dE\,. \tag{3.31}$$

Just like in Eq. (3.17), the asterisk indicates that some contributions occur multiple times. As was already pointed out, this multiple counting arises from the presence of two $\tau_{\square}$ operators on both sides of each A. As before we have to eliminate all these additional contributions. Our task is considerably simplified by the use of our criterion for multiple counting. Let us consider the operator

$$\Omega^{(2)*}=\tau_{\square}A\tau_{\square}A\tau_{\square}\,. \tag{3.32}$$

The presence in Eq. (3.32) of two operators A provides two sources of multiple counting which will be eliminated successively. To this end, use is made of Eq. (3.4) to write

$$\Omega^{(2)*}=\tau_{\square}A\tau_{\square}A[1-(M_{+}^{\infty}+M_{-}^{\infty})]^{-1}\,, \tag{3.33}$$

which by virtue of Eqs. (3.24) and (3.26) reads

$$\Omega^{(2)*}=\tau_{\square}A\tau_{\square}\left(A\tau_0+M_{-}^{\infty}\frac{1}{1-M_{+}^{\infty}}A\,\frac{1}{1-(M_{+}^{\infty}+M_{-}^{\infty})}\right). \tag{3.34}$$

From our criterion, it is clear that the second term in the right-hand side of Eq. (3.34) contains all the additional contributions which have been already counted in $\tau_{\square}A\tau_{\square}A\tau_0$. Therefore, it will be dropped. In doing so, one of the sources of multiple counting is eliminated. Due to the presence of two operators $\tau_{\square}$ on both sides of A, some contributions occur multiple times in the expansion of the remaining operator $\tau_{\square}A\tau_{\square}A\tau_0$. By the same procedure all these additional contributions are eliminated. The operator Ω, which has been previously defined by the operator Ω^* in which all the multiple contributions are canceled, is given for this two-photon process by

$$\Omega^{(2)}=\tau_{\square}A\tau_0A\tau_0\,. \tag{3.35}$$

It is now a simple matter to write the exact expression for the two photon ionization amplitude. One finds

$$\mathcal{A}_{ab}^{(2)}(t)=\frac{1}{2\pi i}\oint e^{-iEt}\langle a|\tau_{\square}A\tau_0A\tau_0G^0|b\rangle\,dE\,. \tag{3.36}$$

A straightforward generalization of the above discussion enables one to determine the form of the N-photon ionization amplitude. One obtains

$$\mathcal{A}_{ab}^{(N)}(t)=\frac{1}{2\pi i}\oint e^{-iEt}\langle a|\tau_{\square}A\tau_0A\tau_0\cdots\tau_0A\tau_0G^0|b\rangle\,dE\,, \tag{3.37}$$

where N operators A must be considered in the integrand. From Eqs. (3.28), (3.36), and (3.37) it is clear that the exact expressions for multiphoton absorption amplitudes are not expressible with the only electron propagator of Eq. (3.7). We observe that the elimination of multiple counting and undercounting of diagrams outlines the role played by the propagation operators

$$G'(E)=\tau_0(E)G^0(E)\,, \tag{3.38a}$$

$$G''(E)=\tau_{\phi}(E)G^0(E)\,, \tag{3.38b}$$

according to Eq. (3.29) which generalizes into

$$\tau_{\square}A\tau_0A\tau_0\cdots\tau_0A\tau_0G^0=\tau_{\phi}A\tau_{\phi}A\tau_{\phi}\cdots\tau_{\phi}A\tau_{\square}G^0\,. \tag{3.39}$$

Since the two representations of Eqs. (3.38) for the electron propagator depend on the way according to which the diagrams are associated, it seems rather puzzling to give a general expression for the level shifts. Only the numerical determination of the pole of $\Omega(E)$ will give the values of the level shifts.

To conclude this section, a diagrammatic representation of the ensemble of the results is given in Fig. 5.

IV. TWO-LEVEL ATOMS

It is instructive to apply the theory so far discussed to the case of a two-level atom. More precisely, we will be concerned, in this section, with the calculation of the light shifts induced by the absorption of one, three and five photons. A comparison with the results previously obtained by other authors will provide a test for our theory. Of course no conclusion will be extrapolated to an atom for which the continuous part of the spectrum play a significant role in any multiabsorption process.

Before going further it is to be noted that for a transition which takes place between the states $|a\rangle$ and $|b\rangle$, all the contributions involving these states as intermediate states can be isolated and summed separately. This technique has been used in Ref. 18 to calculate the Bloch-Siegert shift and will be utilized here when series representation for the shift will be needed. It will be shown how the general results of Sec. III can be straightforwardly expressed within this formalism. We give below some basic formulas elaborated in this framework. For further developments the reader is referred to the papers of Ref. 19.

The matrix element of $G(E)$ from which the transition amplitude can been calculated is

$$G_{ab}(E) = \frac{R_{ab}(E)}{(E-E_a-R_{aa}(E))(E-E_b-R_{bb}(E))-|R_{ab}(E)|^2}\,, \tag{4.1}$$

where $R(E)$ operates in the two dimensional subspace ϵ spanned by $|a\rangle$ and $|b\rangle$, the states of the system atom plus field:

$$R(E) = V + V\frac{Q}{E-H_0}V + V\frac{Q}{E-H_0}V\frac{Q}{E-H_0}V + \cdots\,. \tag{4.2}$$

In Eq. (4.2), Q is the projection operator outside ϵ. The problem now is to calculate exactly the matrix elements of $R(E)$ occurring in Eq. (4.1). For this, we apply our summation techniques of Sec. III. We begin with a slight change in the notations. We define the new absorption and emission operators as

$$\overline{A} = [Q/(E-H_0)]V^-\,, \tag{4.3a}$$

$$\overline{B} = [Q/(E-H_0)]V^+\,, \tag{4.3b}$$

respectively. Thus Eq. (4.2) reads

$$R(E) = (V^+ + V^-)\sum_{q=0}^{\infty}(\overline{A}+\overline{B})^q\,. \tag{4.4}$$

The terms of the infinite series of Eq. (4.4) to be summed for the calculation of R_{aa} and R_{bb} are those which contain equal number of creation and destruction operators of photons. There are two kinds of terms: those beginning with V^+ and those beginning with V^-. It is clear that the first class contains one extra operators $\overline{A}$ and the last class of terms contains one extra operators $\overline{B}$. Regarding only the operators $\overline{A}$ and $\overline{B}$ one has to sum in the first case all the higher order terms contributing to one-photon absorption whereas in the second case we are concerned with one-photon emission. Invoking Eq. (3.27) and the corresponding expression obtained from the substitution $A\rightarrow B, B\rightarrow A$, one can write the form of the operator $R(E)$ to be used for the calculation of $R_{aa}(E)$ and $R_{bb}(E)$. It is

$$R(E) = V^+\overline{\tau}_\square\overline{A}\overline{\tau}_0 + V^-\overline{\tau}_\square\overline{B}\overline{\tau}_\phi\,, \tag{4.5}$$

where

$$\overline{\tau}_\square = (1-\overline{A}\overline{\tau}_0\overline{B}-\overline{B}\overline{\tau}_\phi\overline{A})^{-1}\,, \tag{4.6a}$$

$$\overline{\tau}_0 = (1-\overline{A}\overline{\tau}_0\overline{B})^{-1}\,, \tag{4.6b}$$

$$\overline{\tau}_\phi = (1-\overline{B}\overline{\tau}_\phi\overline{A})^{-1}\,. \tag{4.6c}$$

It is perhaps worthwhile to point out that the following relations hold:

$$\overline{\tau}_\square\overline{A}\overline{\tau}_0 = \overline{\tau}_\phi A\overline{\tau}_\square\,, \tag{4.7a}$$

$$\overline{\tau}_\square\overline{B}\overline{\tau}_\phi = \overline{\tau}_0\overline{B}\overline{\tau}_\square\,. \tag{4.7b}$$

A similar procedure is used for the calculation of R_{ab} and R_{ba}, the nondiagonal matrix elements of $R(E)$. Since the transition can involve the absorption of several photons, one must find the relevant expression of the operator $R(E)$ for each particular case. We begin with one-photon absorption. The terms of the series of Eq. (4.4) to be summed are the ones characterized by one net absorption of photon. As before, there are two classes of terms according to the nature of the first operator, on the left-hand side, which is either V^+ or V^-. Considering the first class, the terms to be considered can be obtained from all the combinations of arbitrary number of $\overline{A}$ and $\overline{B}$ operators in such a way that the net number of operators $\overline{A}$ is two (one of them annihilates the effect of the emission operator V^+). Thus, concerning the operators $\overline{A}$ and $\overline{B}$, we are led to a two-photon absorption problem. Owing to Eq. (3.35), the contribution to $R(E)$ provided by the terms of the first class is $V^+\overline{\tau}_\square\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0$. By the same way one finds that the contribution to $R(E)$ arising from the terms of the second class is $V^-\overline{\tau}_\square$ since in this case V^- accounts for the single absorption. Collecting our results one finds that the form of the operator $R(E)$ to be used in the calculation of the nondiagonal matrix elements of $R(E)$ for one-photon absorption is

$$R(E) = V^+\overline{\tau}_\square\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0 + V^-\overline{\tau}_\square\,. \tag{4.8}$$

The method can be generalized to higher-order absorption processes. Thus, the expression of $R(E)$ to be used for the calculation of the relevant matrix elements in the case of a two-photon absorption is

$$R(E) = V^+\overline{\tau}_\square\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0 + V^-\overline{\tau}_\square\overline{A}\overline{\tau}_0\,, \tag{4.9}$$

and for a three-photon absorption one gets

$$R(E) = V^+\overline{\tau}_\square\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0 + V^-\overline{\tau}_\square\overline{A}\overline{\tau}_0\overline{A}\overline{\tau}_0\,. \tag{4.10}$$

From Eqs. (4.8), (4.9), and (4.10), the generalization to arbitrary order is straightforward. For a N-photon absorption one has

$$R(E)=V^{+}\bar{\tau}_{\Box}\bar{A}\bar{\tau}_{0}\bar{A}\bar{\tau}_{0}\cdots\bar{\tau}_{0}\bar{A}\bar{\tau}_{0}+V^{-}\bar{\tau}_{\Box}\bar{A}\bar{\tau}_{0}\cdots\bar{\tau}_{0}\bar{A}\bar{\tau}_{0}\,, \tag{4.11}$$

where the first term in the right-hand side contains $N+1$ operators $\bar{A}$ and the last contains $N-1$ such operators. It is clear that from Eq. (4.7a) that the above expressions for $R(E)$ may also be written in other equivalent forms involving $\bar{\tau}_{\Box}$ and $\bar{\tau}_{\phi}$. They are not given here to limit the expense of space.

We consider now, in more detail, the problem of one photon transition. The states of the system atom plus field will be denoted $|am\rangle=|a\rangle|m\rangle$ or $|bm'\rangle=|b\rangle|m'\rangle$, $|a\rangle$ and $|b\rangle$ being the two atomic states and m and m' are the occupation number of the field. The corresponding energies are $E_{am}=E_a+m\omega$ and $E_{bm'}=E_b+m'\omega$, E_a and E_b are the energies of the atomic states $|a\rangle$ and $|b\rangle$, respectively and ω is the photon energy (in a.u.). Let us express V^- and V^+ in terms of d the dipole operator. One has

$$V^{-}=da\,, \tag{4.12a}$$

$$V^{+}=d^{*}a^{\dagger}\,, \tag{4.12b}$$

a and $a^{\dagger}$ being the destruction and creation operators of a photon, respectively. Finally, for the sake of brevity we define β as

$$\beta=d_{ab}\,, \tag{4.13}$$

where in contrast to Eq. (4.1) the indices a and b stand for the atomic states only.

For one-photon transition which takes place between the states $|an\rangle$ and $|bn+1\rangle$, one finds from Eqs. (4.1), (4.5), and (4.8) that

$$G_{an\,bn+1}(E)=\frac{R^{(1)}_{an\,bn+1}(E)}{(E-E_{an}-R^{(1)}_{an\,an}(E))(E-E_{bn+1}-R^{(1)}_{bn+1\,bn+1}(E))-|R^{(1)}_{an\,bn+1}(E)|^{2}}\,, \tag{4.14}$$

with

$$R^{(1)}_{an\,bn+1}(E)=(n+1)^{1/2}\beta\,, \tag{4.15a}$$

$$R^{(1)}_{an\,an}(E)=\cfrac{n|\beta|^{2}}{E-E_{bn-1}-\cfrac{(n-1)|\beta|^{2}}{E-E_{an-2}-\cfrac{(n-2)|\beta|^{2}}{E-E_{bn-3}-\cdots}}}\,, \tag{4.15b}$$

$$R^{(1)}_{bn+1\,bn+1}=\cfrac{(n+2)|\beta|^{2}}{E-E_{an+2}-\cfrac{(n+3)|\beta|^{2}}{E-E_{bn+3}-\cfrac{(n+4)|\beta|^{2}}{E-E_{an+4}-\cdots}}}\,. \tag{4.15c}$$

Provided the average number of photons is large enough, all the matrix elements of the operator V appearing in the denominators of the continued fractions reduce to $b=(n)^{1/2}\beta$. In using the method utilized in Ref. 18 the first-order resonance shift (Bloch-Siegert shift) can be obtained from Eqs. (4.15) in the form of a power series of the intensity. Complete agreement is found with the results presented in that paper.

A more-accurate determination of the resonance shift involves a numerical analysis of Eq. (4.14) together with Eqs. (4.15). This has been done and the results shown in Fig. 6 are in excellent agreement with those of Ref. 20. A good test regarding the accuracy of our method is provided by the exact solution which can be found when $\omega_0/\omega=0$ $(\omega_0=E_a-E_b)$.[21-23] Since in this case b/ω satisfies $J_0(4b/\omega)=0$, one can compare our result concerning b/ω with the exact value calculated from the zeros of the Bessel function. Examination of Table V shows that the agreement is highly satisfactory.

The third-order resonance shift for a transition taking place between the states $|an\rangle$ and $|bn+3\rangle$ can be treated in the same way. In contrast to what happens for the first-order resonance, two equivalent expressions for the nondiagonal matrix element of $G(E)$ are obtained depending on whether the states $|an\rangle$ and $|bn+3\rangle$ are contained in the ensemble of the intermediate states or not. In the former case the matrix element $G_{an\,bn+3}(E)$ is conveniently written by using the functions S_{an}, S_{bn}, T_{an}, and T_{bn} defined as

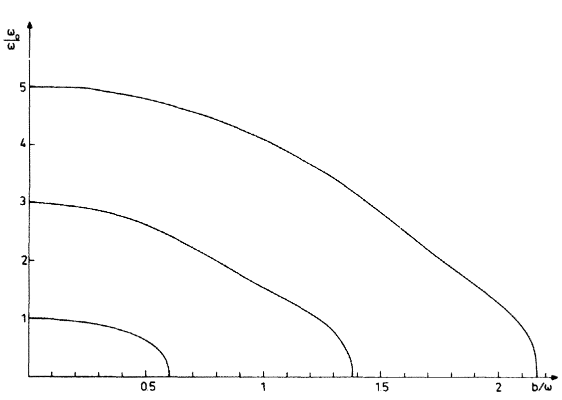

FIG. 6. Bloch-Siegert shifts corresponding to one-, three-, and five-photon resonance as functions of b/ω.

$$S_{(a,b)n}=\frac{n|\beta|^2}{E-E_{(a,b)n}-\dfrac{(n+1)|\beta|^2}{(E-E_{(b,a)n+1}-S_{(a,b)n+2})}}\,, \tag{4.16a}$$

$$T_{(a,b)n}=\frac{n|\beta|^2}{E-E_{(a,b)n-1}-\dfrac{(n-1)|\beta|^2}{(E-E_{(b,a)n-2}-T_{(a,b)n-2})}}\,, \tag{4.16b}$$

where the functions S_a and S_b are simply related by

$$S_{an}=n|\beta|^2/(E-E_{an}-S_{bn+1}) \tag{4.16c}$$

(a similar relation could be found for T_a and T_b). One has

$$G_{an\;bn+3}(E)=\frac{[(n+1)(n+2)(n+3)]^{1/2}\beta^3}{(E-E_{an}-S_{bn+1}-T_{bn})(E-E_{bn+1}-S_{an+2})(E-E_{an+2}-S_{bn+3})(E-E_{bn+3}-S_{an+4})}\,. \tag{4.17}$$

We consider now the second case where the initial and final states never appear as intermediate states. One finds that $G_{an\;bn+3}(E)$ can be written in terms of the matrix elements of the operator $R(E)$ as

$$G_{an\;bn+3}(E)=\frac{R^{(3)}_{an\;bn+3}}{[E-E_{an}-R^{(3)}_{an\;an}(E)][E-E_{bn+3}-R^{(3)}_{bn+3\;bn+3}(E)]-[R^{(3)}_{an\;bn+3}(E)]^2}\,, \tag{4.18}$$

with

$$R^{(3)}_{an\;an}=T_{bn}+\frac{(n+1)|\beta|^2}{E-E_{bn+1}-\dfrac{(n+2)|\beta|^2}{(E-E_{an+2})}}\,, \tag{4.19a}$$

$$R^{(3)}_{bn+3\;bn+3}=S_{an+4}+\frac{(n+3)|\beta|^2}{E-E_{an+2}-\dfrac{(n+2)|\beta|^2}{(E-E_{bn+1})}}\,, \tag{4.19b}$$

$$R^{(3)}_{an\;bn+3}=\frac{[(n+1)(n+2)(n+3)]^{1/2}\beta^3}{(E-E_{bn+1})(E-E_{an+2})-(n+2)|\beta|^2}\,. \tag{4.19c}$$

Naturally, the two equivalent expressions for $G_{an\;bn+3}$ as given by Eqs. (4.17) and (4.18) can be used for the investigation of the resonance shift. The form of Eq. (4.18) is suitable for a perturbative treatment of the shift, whereas the factorized expression of Eq. (4.17), which is obtained with a little bit of algebraic manipulations is used for computational analysis. The numerical results obtained for this third-order resonance shift are shown in Fig. 6 where the excellent agreement with those of Ref. 20 is to be noted. As before the accuracy of our calculation is tested in considering the exact solution which can be found for $\omega_0/\omega=0$. Table V shows that our results are obtained with extremely good precision.

The aim of the above quantitative analysis concerning a two-level atom was not to deal with a problem which has been intensively discussed in the literature. This simple case has provided a test for the accuracy of a calculation involving the general formulas of Sec. III. The rapid convergence of the continued fractions which has been observed for the treatment of this problem seems promising for further application of the method to more complicated atoms.

TABLE V. Values of ω_0/ω determined in using the parameter b/ω calculated from the zeroes of $J_0(4b/\omega)$, for few low-order resonances. The numbers in parentheses indicate the powers of 10.

order	1	3	5
b/ω	0.601 206 389 3	1.380 019 528	2.163 431 978
ω_0/ω	3.90(−10)	1.97(−10)	2.30(−10)

V. RAMAN-LIKE PROCESSES

So far we have only envisaged the multiple absorption of photons induced by a single radiation field but the method we used can be generalized to more-complicated problems. To see this we consider, in this section, the case of an electron interacting with two electromagnetic (em) fields. Among the processes which this interaction can originate, we limit ourselves to the discussion of Raman-like processes. The two em fields are chosen to behave unlikely with regard to the electron. One of them is the radiation field considered in the preceding sections. It has been shown how this field can give rise to multiphoton absorptions and radiative shifts. The additional quantized field surrounding the atomic electron is the radiation field of arbitrary small occupation number originated by the Raman emission. Such a field cannot give rise to significant perturbation on the atom. Thus the creation operators of this field will be only considered.

The resolvent operator for a two-field problem is given by

$$G(E)=G_0(E)+G_0(E)(V+V')G(E)\,, \tag{5.1}$$

where

$$G_0(E)=1/(E-H_0)\,. \tag{5.2}$$

H_0 is the free Hamiltonian of the system, V and V' describe the interaction of the electron with the fields. Remembering Eqs. (2.4) and (2.13) we can rewrite Eq. (5.1) as

$$G(E) = G_0(E) + (A + B + Z)G(E), \tag{5.3}$$

where Z is the emission operator of the Raman photon, i.e., $Z \equiv G_0 V'^{+}$. The operator Z is illustrated in Fig. 7 by a horizontal waved line. Since Z does not contain a destruction operator, any absorption of photon and forward-scattering process is referred unambiguously to the laser field whose destruction and creation operators are denoted by A and B, respectively. Equation (5.3) can be solved by iteration. One obtains $G(E)$ on the form of an infinite series in terms of A, B, and Z. For example up to first order in Z and up to third order in A and B the expression of $G(E)$ to be used for one-photon absorption is

$$\begin{aligned} G(E) = {} & AG_0 + (BAA + ABA + AAB)G_0 + (ZA + AZ)G_0 \\ & + (ZBAA + BZAA + BAZA + BAAZ)G_0 \\ & + (ZABA + AZBA + ABZA + ABAZ)G_0 \\ & + (ZAAB + AZAB + AAZB + AABZ)G_0 \,. \end{aligned} \tag{5.4}$$

One observes that to zeroth order in Z, one obtains the series already found for a one-photon absorption induced by a single field. To first order in A and in Z one has the two terms representing the lowest-order contribution to the Raman effect. The other terms in Eq. (5.4) are the higher-order corrections. One can find many other types of series involving arbitrary number of absorbed photons and more than one interaction with the additional em field. It is to be noted that in contrast to what happened in the case of a single field, the number of terms to be summed rapidly increases and one must handle considerable amount of contributions even for low perturbation orders. In using the close analogy between the problem we consider and the one we have solved in Sec. III, the summation can be performed straightforwardly. To see this we treat the most general case where arbitrary number of interactions of the electron with the two em fields occur. It is convenient to write $G(E)$ as

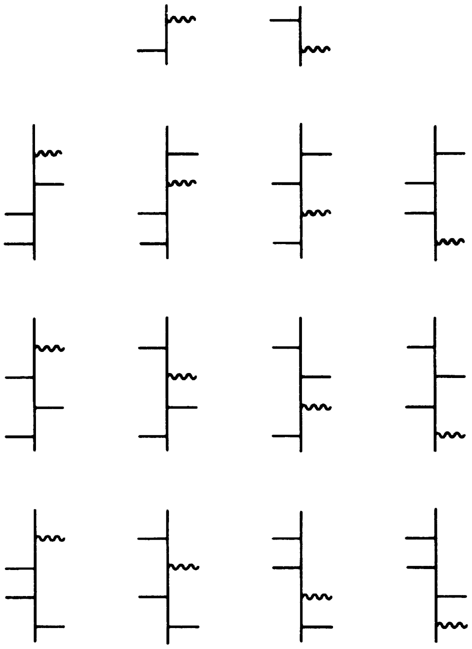

FIG. 7. Second- and fourth-order diagrams contributing to Raman scattering. The Raman photon is represented by a waved line.

$$G(E) = \tilde{\Omega}^{(N)}(E) G_0(E), \tag{5.5}$$

where the superscript N indicates the net number of absorbed photons.

Limiting ourselves to one photon absorption we have written in Table VI all the contributions to $\tilde{\Omega}^{(1)}$ up to the fourth order in the field strengths regardless the nature of these fields. In making the substitutions $X \rightarrow A$, $Y \rightarrow B$, $T_1 = T \rightarrow Z$, we note a one to one correspondence between the 25 terms shown in Tables III and VI. This analogy holds for every perturbation orders. Therefore, from Eq. (3.27) together with Eqs. (3.4), (3.11), (3.12), and (3.13), it is a simple matter to derive the exact expression for $\tilde{\Omega}^{(1)}(E)$. One has

$$\tilde{\Omega}^{(1)} = \tilde{\tau}_{\square} A \tilde{\tau}_0 \,, \tag{5.6}$$

TABLE VI. Contributions to one-photon absorption up to fourth order in the presence of the radiation field whose creation operator is represented by Z.

Order 1	Order 4
A	$Z^3 A$
Order 2	$ZAB\ A$
	$AZB\ A$
$Z\ A$	$ABZ\ A$
$A\ Z$	$ZBA\ A$
Order 3	$BZA\ A$
	$BAZ\ A$
$Z^2 A$	$Z^2 A\ Z$
$AB\ A$	$AB\ A\ Z$
$BA\ A$	$BA\ A\ Z$
$Z\ A\ Z$	$Z\ A\ Z^2$
$A\ Z^2$	$Z\ A\ AB$
$A\ AB$	$A\ Z^3$
	$A\ ZAB$
	$A\ AZB$
	$A\ ABZ$

where

$$\tilde{\tau}_{\Box} = 1/[1 - (Z + A\Lambda^{-}B + B\Lambda^{+}A)], \tag{5.7a}$$

$$\tilde{\tau}_{0} = 1/[1 - (Z + A\Lambda^{-}B)], \tag{5.7b}$$

$$\Lambda^{-} = 1/[1 - (Z + A\Lambda^{-}B)], \tag{5.8a}$$

$$\Lambda^{+} = 1/[1 - (Z + B\Lambda^{+}A)]. \tag{5.8b}$$

For shortness the dependence on E is not written explicitly. A straightforward generalization of the above discussion allows one to determine the expression of $\tilde{\Omega}^{(N)}$; For N net absorptions of photons one finds

$$\tilde{\Omega}^{(N)} = \tilde{\tau}_{\Box} A \tilde{\tau}_{0} A \tilde{\tau}_{0} \cdots \tilde{\tau}_{0} A \tilde{\tau}_{0}. \tag{5.9}$$

Any specific process can be described in considering a special case of the above formulas. For example, returning to one-photon absorption we make the assumption that the laser field is weak enough to neglect all forward-scattering processes. On the other hand, let Z operating in the two-dimensional space be spanned by the (Raman) photon states $|0\rangle$ and $|1\rangle$. The matrix element of $\tilde{\Omega}^{(1)}$ with these states is

$$\tilde{\Omega}_{10}^{(1)} = \langle 1|\Omega^{(1)}|0\rangle \simeq \langle 1|\frac{1}{1-Z} A \frac{1}{1-Z}|0\rangle, \tag{5.10}$$

since

$$Z = \begin{pmatrix} 0 & 0 \\ Z_{10} & 0 \end{pmatrix}, \tag{5.11}$$

one gets

$$\tilde{\Omega}_{10}^{(1)} = Z_{10}A + AZ_{10}. \tag{5.12}$$

Equation (5.12) shows the two familiar lowest-order contributions to Raman scattering, i.e., absorption (emission) of a single photon of the laser field followed by the emission (absorption) of a single photon of different mode.

This simple example illustrates the manner by which the required expression for a specific process can be extracted from our general formulas. For computational analysis the above approximation regarding the higher-order effects induced by the field is not necessary and any problem can be envisaged in its generality. This outlines the usefulness of having compact formulas accounting for the ensemble of processes occurring when two fields of quite different intensities interact with an atom.

As a concluding remark one must note that the same discussion holds for free electrons. Thus, multiple absorption or emission of photons by an electron in the presence of a static potential can be treated in the same way. The only difference is in the operator Z which must describe the interaction of the electron with the static field. Thus Eqs. (5.7), (5.8), (5.9), and the ones obtained in changing $A \to B, B \to A$ can be used for the calculation of higher-order corrections to bremsstrahlung and inverse bremsstrahlung problems. Such applications of the theory is devoted to subsequent publications.

VI. CONCLUSION

The summation of the whole perturbation series representing all the higher-order contributions to definite absorption processes has led to compact expressions for the transition amplitudes. The way by which they have been elaborated is quantum electrodynamically consistent i.e.; our technique avoids any kind of multiple counting or undercounting of diagrams. In that sense, the expressions presented in this paper are a representation of the exact transition amplitudes. Some important remarks regarding the structure of the results can be formulated. Firstly we observe that in any multiphoton process the perturbed Green's function joining two adjacent vertices is not the exact electron Green's function which can be calculated separately. As a consequence no explicit form for the radiative shifts can be put forward. Only a numerical investigation of the poles of the resolvent operator will give the value of the atomic level shifts. The summation of diagrams renders everything into close expressions which must be considered in their entirety. Secondly, the fact that the transition amplitudes are expressed in nonperturbative form with regard to the field strength, suggests an important application of our results to the case of very intense fields.

The transition amplitudes we obtain are bounded for every value of the photon flux. Such a property indicates that the corresponding expressions may be considered as analytic continuation of the perturbation series. The availability of expressions as ours facilitate rigorous and accurate studies of the problem of ultraintense fields. This aspect of the problem of the interaction of the radiation with an atom is not discussed here but will be subsequently examined.

The calculation of the matrix elements of operators in terms of the continued fractions is not easy. The origin of the difficulty lies in the presence of the continuum whose contribution increases as the intensity is varied towards large values. A numerical method allowing the accurate calculation of this contribution is in progress. Quantitative investigation dealing with finite number of discrete levels have been done. In this respect the situation is very encouraging. With the standard iteration procedure for evaluating the continued fractions, convergence is easily reached. This has been observed in the two-level atom considered in Sec. IV and also in more complicated atoms. For example, with a system of 28 levels and up to the intensity of 10^{17} W/cm^2, there have been no problem regarding convergence of the continued fractions in a trial calculation, for a case that we have examined.[14]

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APPENDIX

It is shown here how the mass operator M^∞ as given by Eq. (3.14) can be obtained from Eqs. (3.11), (3.12), and (3.13). To this end some algebraic manipulations must be done in the expressions of $M^\infty_\pm$. Let us consider M^∞_- which can be written from Eqs. (3.8) and (3.11a) as

$$M^\infty_- = A(1 + A\Pi^- B)B . \tag{A1}$$

Using Eq. (3.12a) one has

$$M^\infty_- = A\left(1 + A \frac{1}{1 - [BA + A(1 + A\Pi^- B)B]} B\right) B , \tag{A2}$$

which from Eq. (A1) gives

$$M^\infty_- = A\left(1 + A \frac{1}{1 - (BA + M^\infty_-)} B\right) B . \tag{A3}$$

Utilizing the Feynman identity, the fraction inside the large parentheses can be expanded and one obtains

$$M^\infty_- = A\left[1 + A\left(\frac{1}{K} + \frac{1}{K} BA \frac{1}{K} + \frac{1}{K} BA \frac{1}{K} BA \frac{1}{K} + \cdots\right) B\right] B , \tag{A4}$$

where

$$K = 1 - M^\infty_- . \tag{A5}$$

The series in the right-hand side of Eq. (A4) can be immediately summed and M^∞_- can be written

$$M^\infty_- = A[1 - A(1/K)B]^{-1} B , \tag{A6}$$

which by virtue of Eq. (A5) reads

$$M^\infty_- = A[1 - A(1 - M^\infty_-)^{-1} B]^{-1} B . \tag{A7}$$

Finally, by iteration, one finds that M^∞_- can be expressed in terms of the operator τ_0, defined in Eq. (3.15a), as

$$M^\infty_- = A\tau_0 B . \tag{A8}$$

Similar arguments hold for M^∞_+. One obtains

$$M^\infty_+ = B\tau_\phi A , \tag{A9}$$

where τ_ϕ is given by Eq. (3.15b).

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