PHYSICAL REVIEW A VOLUME 50, NUMBER 3 SEPTEMBER 1994

Multiphoton processes in the velocity gauge

Yves Gontier and Michel Trahin
Service des Photons, Atomes et Molecules, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette Cedex, France

(Received 8 April 1994)

The role played by the quadratic term of the interaction Hamiltonian $-\alpha\,\mathbf{A}(t)\cdot\mathbf{p}+\frac{1}{2}\alpha^2\,\mathbf{A}^2(t)$ in the velocity gauge is examined in both the semiclassical and the quantized theory. It is shown that this term cannot be eliminated in every case via standard contact transformations. For second-order processes a special transformation is proposed that eliminates the noncontributing parts of $\mathbf{A}^2(t)$, and leads to the right expressions for the Stark shift and the resonance-fluorescence probability. The generalization to arbitrary order of these results is done in a fully quantized theory by resorting to diagram summation. One finds that the contributions of diagrams displaying double-photon absorptions and emissions sum to zero. The only effect of the quadratic term is to shift the whole atomic spectrum by an intensity-dependent amount.

PACS number(s): 32.80.Rm, 32.60.+i

I. INTRODUCTION

The calculations in multiphoton theory and, more generally, those dealing with the interaction of intense radiation fields with atomic systems, are done either in the length gauge or in the velocity gauge. In the former case, the interaction Hamiltonian is $\mathbf{E}(t)\cdot\mathbf{r}$ (in a.u.), $\mathbf{E}(t)$ being the electric field, while in the last case the interaction is expressed as

$$-\alpha\,\mathbf{A}(t)\cdot\mathbf{p}+(\alpha^2/2)\,\mathbf{A}^2(t)$$

in terms of the vector potential $\mathbf{A}(t)$, the electron momentum $\mathbf{p}$, and the fine-structure constant α.

The choice of the gauge depends on the problem one has to solve. In this respect, it has been shown [1] that the difficulties arising in the calculation of the continuum-continuum matrix elements of the shift operator were more easily solved in the velocity gauge. In this case the $\mathbf{A}^2(t)$ term of the interaction Hamiltonian plays a central role. The two parts of this Hamiltonian have different effects on the states of the system atom-plus-field. The $\mathbf{A}(t)\cdot\mathbf{p}$ changes both the electron and the photon states, while the $\mathbf{A}^2(t)$ term operators only on the photon states. Since this term does not induce any atomic transition, one could be tempted to eliminate it by means of suitable transformations. One of them is discussed in Sec. II. By expressing the solution of the transformed equation in the form of a perturbative series, it is shown that the second-order approximation does not lead to the right expression for the resonance-fluorescence probability. This result proves that the arbitrary cancellation of the whole quadratic part of the interaction is not justified. In contrast, we show that there exists a transformation which eliminates most of the terms displayed by the expansion of $\mathbf{A}^2(t)$ and leads to the exact formula for the second-ordr Stark shift and the Kramers-Heisenberg formula.

In Sec. III the generalization of the discussion is done within a fully quantized calculation scheme. We evaluate up to arbitrary orders the contributions of $\mathbf{A}^2(t)$. By using a conventional diagrammatic representation, we show that this term provides three kinds of noncommuting interactions taking place at the same vertex: (i) undistinguishable absorption emission and emission absorption of a photon, (ii) absorption of two photons, and (iii) emission of two photons. The first kind of process is treated on the same footing as the free-field Hamiltonian. In contrast, the double absorptions and the double emissions together with the single-photon interactions coming from the $\mathbf{A}(t)\cdot\mathbf{p}$ term reduce the problem arising from gauge transform to that of performing the summation of a perturbation series involving four operators. To lowest order, it was found [2–4] that, in multiphoton transitions, the operator $\mathbf{A}^2(t)$ could be discarded for processes involving more than two net absorptions or emissions of photons. But this result cannot be extended to higher-order processes. The reason lies in the difficulty arising from divergent irreducible diagrams, i.e,. the diagrams accounting for transitions leaving the electron in its atomic initial or final state. This problem has motivated the present revisitation of multiphoton theory. In Sec. IV the diagrams representing high-order interactions are classified in four groups containing (i) only single-photon interactions, (ii) only double-photon interactions, (iii) a mixing of equal numbers of double absorptions and double emissions, and (iv) a mixing of an unequal number of double absorptions and double emissions. In doing so, one finds that some diagrams belonging to the third and the fourth class lead to divergences and provide overcontributions. The difficulty is solved in Sec. (V) where it is shown that among the double interactions supplied by $\mathbf{A}^2(t)$, only one can be straightforwardly treated. It comes from absorption-emission processes occurring at the same vertex, which produce an intensity-dependent shift of the whole spectrum which may be included in the free Hamiltonian. In Sec. (VI) nonperturbative expressions of the shift operator $R(z)$ depending only on $\mathbf{p}$ and $\mathbf{A}(t)$ operators are derived.

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II. SEMICLASSICAL THEORY

A. Contact transformation

Before looking at the problems arising in the velocity gauge within a fully quantized formalism, we examine the effect of a contact transition [6,7] on the semiclassical solution of the Schrödinger equation. The equation one has to solve is (in a.u.)

$$[H_0(\mathbf{r})+H_I'(\mathbf{r},t)+H_I''(t)]\Psi(\mathbf{r},t)=i\dot{\Psi}(\mathbf{r},t)\ , \tag{2.1a}$$

where

$$H_0(\mathbf{r})=\frac{\mathbf{p}^2}{2}+V(r)\ , \tag{2.1b}$$

$$H_I'(\mathbf{r},t)=-\alpha\,\mathbf{A}(t)\cdot\mathbf{p}\ , \tag{2.1c}$$

$$H_I''(t)=\frac{\alpha^2}{2}\,\mathbf{A}^2(t)\ , \tag{2.1d}$$

$$\mathbf{A}(t)=\frac{1}{\alpha}\left[\frac{F_1}{F_0}\right]^{1/2}\sum_k\frac{1}{\sqrt{\omega_k}}\boldsymbol{\varepsilon}_k(e^{i\omega_k t}+e^{-i\omega_k t})\ . \tag{2.1e}$$

In Eqs. (2.1), $V(r)$ is the static potential, $\boldsymbol{\varepsilon}_k$ is the photon polarization vector in the kth mode corresponding to energy ω_k, and F_1 is the flux corresponding to a single photon ($F=nF_1$).

We transform the state vector according to

$$\Psi(\mathbf{r},t)=e^{-i\int_0^t H_I''(t')dt'}\hat{\Psi}(\mathbf{r},t)\ . \tag{2.2}$$

The state $\hat{\Psi}(\mathbf{r},t)$ is the solution of the transformed equation

$$[H_0(\mathbf{r})+H_I'(\mathbf{r},t)]\hat{\Psi}(\mathbf{r},t)=i\dot{\hat{\Psi}}(\mathbf{r},t)\ , \tag{2.3}$$

which is obtained by using the independence property of the vector potential with respect to the space variables.

Equation (2.3) can be written in the interaction picture as

$$\overline{H}_I'(\mathbf{r},t)\overline{\Psi}(\mathbf{r},t)=i\dot{\overline{\Psi}}(\mathbf{r},t)\ , \tag{2.4}$$

where

$$\overline{\Psi}(\mathbf{r},t)=e^{iH_0t}\hat{\Psi}(\mathbf{r},t)\ , \tag{2.5a}$$

$$\overline{H}_I'(\mathbf{r},t)=e^{iH_0t}H_I'(\mathbf{r},t)e^{-iH_0t}\ . \tag{2.5b}$$

The solution of Eq. (2.4) is easily found to be

$$\overline{\Psi}(\mathbf{r},t)=\Phi(\mathbf{r})-i\int_0^t\overline{H}_I'(\mathbf{r},t')\overline{\Psi}(\mathbf{r},t')dt'\ . \tag{2.6}$$

The state $\Phi(\mathbf{r})$ is the solution of the Schrödinger equation in the absence of the radiation field. Due to Eqs. (2.2) and (2.5), the rate of the system atom plus field is obtained from the integral equation

$$\Psi(\mathbf{r},t)=e^{-iW(t)}\left[\Phi(\mathbf{r},t)-i\int_0^t e^{-iH_0(t-t')}H_I'(\mathbf{r},t')e^{iW(t')}\right.$$
$$\left.\times\Psi(\mathbf{r},t')dt'\right]\ , \tag{2.7}$$

where

$$\Phi(\mathbf{r},t)=\Phi(\mathbf{r})e^{-iEt} \tag{2.8a}$$

and

$$W(t)=\int_0^t H_I''(t')dt'\ . \tag{2.8b}$$

In Eq. (2.7) the effect of the quadratic term is to introduce a phase factor which can be dropped without affecting the value of the probability. The state $\Psi(\mathbf{r},t)$ can be expressed in the form of a perturbative series by iterating Eq. (2.7):

$$\Psi(\mathbf{r},t)=\Phi(\mathbf{r},t)-i\int_0^t dt'e^{-iH_0(t-t')}H_I'(\mathbf{r},t')\Phi(\mathbf{r}',t')$$
$$-\int_0^t dt'\int_0^{t'}dt''e^{-iH_0(t-t')}H_I'(\mathbf{r},t')e^{-iH_0(t'-t'')}H_i'(\mathbf{r},t'')\Phi(\mathbf{r},t'')+\cdots\ . \tag{2.9}$$

To check the relevance of the transformation of Eq. (2.2), we look at the resonance fluorescence. We calculate the probability for the absorption of a photon in the mode k and the emission of a photon in the mode k'. To this end, we consider the third term of Eq. (2.9) which displays two electron-field interactions. It is the only contribution to the second-order process we are concerned with.

By using Eqs. (2.1), the probability amplitude for the second-order absorption-emission process of Eq. (2.9) can be written as

$$M_{fg}^{(2)}(t)=\frac{F_1}{F_0}\frac{1}{\sqrt{\omega_k\omega_{k'}}}\sum_i\int_0^t dt'\int_0^{t'}dt''(\boldsymbol{\varepsilon}_{k'}\cdot\mathbf{p})_{fi}(\boldsymbol{\varepsilon}_k\cdot\mathbf{p})_{ig}e^{i(\omega_f+\omega_{k'}-\omega_i)t'}e^{i(\omega_i-\omega_k-\omega_g)t''}$$
$$+(\boldsymbol{\varepsilon}_k\cdot\mathbf{p})_{fi}(\boldsymbol{\varepsilon}_{k'}\cdot\mathbf{p})_{ig}e^{i(\omega_f-\omega_k-\omega_i)t'}e^{i(\omega_i+\omega_{k'}-\omega_g)t''}\ . \tag{2.10}$$

The probability per unit time which is obtained after little algebra reads

$$P_{fg}^{(t)}=\frac{F_1}{F_0}\left|\sum_i\left[\frac{(\boldsymbol{\varepsilon}_{k'}\cdot\mathbf{p})_{fi}(\boldsymbol{\varepsilon}_k\cdot\mathbf{p})_{ig}}{\omega_g-\omega_i+\omega_k}+\frac{(\boldsymbol{\varepsilon}_k\cdot\mathbf{p})_{fi}(\boldsymbol{\varepsilon}_{k'}\cdot\mathbf{p})_{ig}}{\omega_g-\omega_i-\omega_{k'}}\right]\right|^2 \times\delta(\omega_f-\omega_g+\omega_{k'}-\omega_k)\ . \qquad (2.11)$$

In comparison with the well-know formula of Kramers and Heisenberg [8], we see that the expression of Eq. (2.11) holds when the initial and the final states are different. In the case where $f=g$, the lack of any contribution coming from the $\mathbf{A}^2(t)$ term leads to a wrong result. This is the consequence of the transformation of Eq. (2.2) which is not appropriate.

B. Modified contact transformation

In this section we consider another transformation which enables us to eliminate the noncontributing part of $\mathbf{A}^2(t)$ but preserves the right expression of the Kramers-Heisenberg formula and that of the second-order Stark shift calculated in the velocity gauge. In combining Eqs. (2.1d) and (2.1e), one finds that $H_I''(t)$ can be written as

$$H_I''(t)=H_I^{(1)}(t)+H_I^{(2)}(t)\ , \qquad (2.12)$$

where

$$H_I^{(1)}(t)=\frac{F_1}{F_0}\left[\frac{\boldsymbol{\varepsilon}_k^2}{\omega_k}[\cos(2\omega_k t)+1]+\frac{\boldsymbol{\varepsilon}_{k'}^2}{\omega_{k'}}[\cos(2\omega_{k'}t)+1]+2\frac{\boldsymbol{\varepsilon}_k\cdot\boldsymbol{\varepsilon}_{k'}}{\sqrt{\omega_k\omega_{k'}}}\cos[(\omega_k+\omega_{k'})t]\right] \qquad (2.13a)$$

and

$$H_I^{(2)}(t)=\frac{2F_1}{F_0}\frac{\boldsymbol{\varepsilon}_k\cdot\boldsymbol{\varepsilon}_{k'}}{\sqrt{\omega_k\omega_{k'}}}\cos[\omega_k-\omega_{k'})t]\ . \qquad (2.13b)$$

To solve Eq. (2.1a), we transform the wave vector according to

$$\Psi(\mathbf{r},t)=e^{i\int_0^t dt' H_I^{(1)}(t')}\tilde{\Psi}(\mathbf{r},t)\ , \qquad (2.14)$$

where $\tilde{\Psi}(\mathbf{r},t)$ is the solution of the transformed equation

$$[H_0(\mathbf{r})+H_I'(\mathbf{r},t)+H_I^{(2)}(t)]\tilde{\Psi}(\mathbf{r},t)=i\dot{\tilde{\Psi}}(\mathbf{r},t)\ . \qquad (2.15)$$

By following the same procedure as the one which led to Eq. (2.7), one finds that $\Psi(\mathbf{r},t)$ can be expressed in the form of the following perturbation series:

$$\Psi(\mathbf{r},t)=\Phi(\mathbf{r},t)-i\int_0^t dt' e^{-iH_0(t-t')}[H_I'(t')+H_I^{(2)}(t')]\Phi(\mathbf{r},t') -\int_0^t dt'\int_0^{t'}dt'' e^{-iH_0(t-t')}[H_I'(t')+H_I^{(2)}(t')]e^{-iH_0(t'-t'')}[H_I'(t'')+H_I^{(2)}(t'')]\Phi(\mathbf{r},t'')+\cdots\ , \qquad (2.16)$$

where, for brevity the space dependence of the operators has not been written explicitly.

In Eq. (2.16) the solution $\Psi(\mathbf{r},t)$ is determined up to the phase factor displayed in Eq. (2.14), i.e.,

$$\exp\left\{-i\int_0^t dt' H_I^{(1)}(t')\right\}\ .$$

The difference between the expansions of Eqs. (2.9) and (2.16) is the presence of $H_I^{(2)}(t)$ in the latter. As a result of straightforward algebra, one finds that the probability per unit time for the absorption of a photon in the mode k and the emission of a photon in the mode k' by a bound electron which makes a transition from the state $|g\rangle$ to the state $|f\rangle$ is

$$P_{fg}^{(t)}=\frac{F_1}{F_0}\frac{2\pi}{\sqrt{\omega_k\omega_{k'}}}\left|\boldsymbol{\varepsilon}_k\cdot\boldsymbol{\varepsilon}_{k'}\delta_{fg}+\sum_i\frac{(\boldsymbol{\varepsilon}_{k'}\cdot\mathbf{p})_{fi}(\boldsymbol{\varepsilon}_k\cdot\mathbf{p})_{ig}}{\omega_g-\omega_i+\omega_k}+\frac{(\boldsymbol{\varepsilon}_k\cdot\mathbf{p})_{fi}(\boldsymbol{\varepsilon}_{k'}\cdot\mathbf{p})_{ig}}{\omega_g-\omega_i-\omega_{k'}}\right|^2 \times\delta(\omega_f-\omega_g+\omega_{k'}-\omega_k)\ , \qquad (2.17)$$

where δ_{fg} is the Krönecker symbol.

By comparison with Eq. (2.11), one sees that the purely field-independent term is restored. Therefore, the expansion of Eq. (2.16) provides the right expression for the transition rate while the noncontributing parts of $\mathbf{A}^2(t)$ are eliminated by the transformation of Eq. (2.14) in the form of a phase factor. We note that the parts of $\mathbf{A}^2(t)$ which are eliminated by the transformation are characterized by oscillation frequencies which are the sum and twice the frequencies of each mode. The remaining contribution occurring in the expansion (2.16) is a low-frequency term which becomes independent of the frequency when $k\rightarrow k'$.

We have checked the rightness of the expansion in the case of a well-known second-order process. In semiclassical theory, many transformations like those of Eq. (2.2) or Eq. (2.14) can be done. Their effect is to put some terms of the Hamiltonian into a phase factor which is canceled by taking the square of the transition amplitude. Such a procedure is fully justified if one knows that the terms involved do not bring any contribution, but it is not always the case. The example discussed in this section shows that there exists a transformation where the low-frequency contribution of the $\mathbf{A}^2(t)$ plays an essential role while the high-frequency ones can be discarded. From this particular second-order process, one sees that it is not always justified to ignore the quadratic term of the interaction Hamiltonian as is often done in the literature. The problem we must solve now concerns the generalization of the preceding discussion to processes of arbitrary orders provided by the series (2.16). More precisely, we have to examine to what extent the high-frequency contributions can be neglected in terms of arbitrary orders. To this end, we resort to a fully quantized calculation scheme where the interactions at frequencies $2\omega_k$, $2\omega_{k'}$, and $\omega_k+\omega_{k'}$ correspond to simultaneous absorptions and emissions of two photons of the same mode and of two different modes.

III. QUANTIZED THEORY

In this section we consider a quantized monomode radiation field. The formalism of the resolvent operator is adopted and extensive use of the diagrammatic representation is made. The quantized energy which appears in the expression of the free Hamiltonian H_0 of the system precludes the commutation of $\mathbf{A}$ or $\mathbf{A}^2$ with H_0. Thus, all the diagrams are topologically different. In the Schrödinger picture, the Hamiltonian of the system atom plus field is (in a.u.)

$$H=H_0-\alpha\,\mathbf{A}\cdot\mathbf{p}+\frac{\alpha^2}{2}\mathbf{A}^2\ , \tag{3.1}$$

where

$$H_0=-\frac{1}{2}\nabla^2-\frac{1}{r}+\frac{1}{2}\omega(a^\dagger a+aa^\dagger) \tag{3.2}$$

and

$$\mathbf{A}=\frac{1}{\alpha}\frac{1}{\omega}\left[\frac{I_1}{I_0}\right]^{1/2}(\boldsymbol{\varepsilon}a+\boldsymbol{\varepsilon}^*a^\dagger)\ . \tag{3.3}$$

In Eq. (3.3), I_1 is the intensity (in W/cm^2) corresponding to one photon, i.e., $I=nI_1$;

$$I_0=1.404\times10^{17}\ \mathrm{W/cm^2}\ ,$$

a and $a^\dagger$ are the destruction and the creation operators of a photon of energy ω, respectively; and $\boldsymbol{\varepsilon}$ is the polarization vector of the field. For brevity, the time and the spatial dependences of the operators will not be written explicitly. Substitution of $\mathbf{A}$ as given by Eq. (2.3) into Eq. (3.1) gives the different components of the Hamiltonian explicitly. One has

$$H=H_0+H_I\ , \tag{3.4a}$$

$$H_I=K^-+K^++V^-+V^++W^-+W^+\ , \tag{3.4b}$$

where

$$V^-=(V^+)^*=-\frac{1}{\omega}\left[\frac{I_1}{I_0}\right]^{1/2}\boldsymbol{\varepsilon}\cdot\mathbf{p}\,a\ , \tag{3.5a}$$

$$K^-=(K^+)^*=\frac{1}{2\omega^2}\frac{I_1}{I_0}aa^\dagger\ , \tag{3.5b}$$

and

$$W^-=(W^+)^*=\frac{1}{2\omega^2}\frac{I_1}{I_0}aa\ . \tag{3.5c}$$

In Eqs. (3.5), $V^\pm$ are the single-photon interactions (absorptions or emissions), while $K^\pm$ and $W^\pm$ are the operators corresponding to processes where two photons simultaneously interact with the electron. Thus, K^- and K^+ account indifferently for the absorption-emission or the emission-absorption of a photon; W^- and W^+ correspond to a double-photon absorption and emission, respectively. We note that $K^\pm$ and $W^\pm$ are the quantum operators corresponding to $H_I^{(2)}(t)$ and $H_I^{(1)}(t)$ of Eqs. (2.13), respectively.

By defining $K=K^++K^-$, we note that

$$K=\frac{1}{2}\frac{I_1}{I_0\omega^2}[a,a^\dagger]_+ \tag{3.6}$$

is of the same form as that of the free radiation field. By including K into H_0, we define the intensity-dependent Hamiltonian

$$H_0'=-\tfrac{1}{2}\nabla^2-\frac{1}{r}+\frac{1}{2}\left[\omega+\frac{I_1}{I_0\omega^2}\right][a,a^\dagger]_+\ . \tag{3.7}$$

Equation (3.7) shows that the terms $aa^\dagger+a^\dagger a$ of $\mathbf{A}^2$ accounting for the absorption and the emission of a photon at the same vertex simply produce an intensity-dependent shift of the whole spectrum of the atom. Such a shift is taken into account by using H_0' instead of H_0.

The well-known formalism of the resolvent operator is used here and will not be repeated. The readers can look them up in the literature [9,10]. One defines the resolvent operator $G'(z)$ as

$$G'(z)=\frac{1}{z-H_0'-H_I'}\ , \tag{3.8}$$

where H_0' is given by Eq. (3.7) and

$$H_I'=V^-+V^++W^-+W^+\ . \tag{3.9}$$

By expanding $G'(z)$ with respect to H_I', one has

$$G'(z)=G_0'(z)+G_0'(z)H_I'G_0'(z)+G_0'(z)H_I'G_0'(z)H_I'G_0'(z)+\cdots\ , \tag{3.10}$$

with

$$G_0'(z)=\frac{1}{z-H_0'}\ . \tag{3.11}$$

The serial expression of $G'(z)$ can be put into the following compact form:

$$G'(z)=G_0'(z)+G_0'(z)S'(z)G_0'(z)\ , \tag{3.12}$$

where the operator $S'(z)$ is expressed in a power series of the interaction as

$$S'(z)=H_I'+H_I'G_0'(z)H_I'+H_I'G_0'(z)H_I'G_0'(z)H_I'+\cdots\ . \tag{3.13}$$

The difference between $S'(z)$ and the shift operator $R(z)$ of the standard resolvent theory lies in the fact that no state is excluded from the basis involved by the representation of the Green functions $G_0'(z)$.

The study of multiphoton processes requires the knowledge of the diagonal and the nondiagonal matrix elements of $S'(z)$ [11]. To warrant the relevance of the theory at high intensity, nonperturbative expressions of the matrix elements of this operator must be derived from Eq. (3.13) and then calculated to all orders in the intensity parameter. To this end, one must sum the whole perturbation series of Eq. (3.13). The expressions thus obtained display continued fractions involving, for linearly polarized light, the four interaction operators of Eq. (3.14). They hold at arbitrary high intensity provided that enough iterations of the continued fractions are taken into account.

IV. DIAGRAMMATIC REPRESENTATION

A. Classification of the diagrams

We discuss the procedure of summation by considering the diagonal part of $S'(z)$. If one substitutes H_I' as given by Eq. (3.9) into Eq. (3.13), we are led to a perturbation series involving four noncommuting operators. They account for all the possible combinations of single- and/or double-photon absorptions and emissions so that the number of absorptions is equal to the number of emissions. The summation of such a series is made easier by representing diagrammatically the processes we are concerned with. As is shown in Fig. 1, the photons correspond to horizontal lines which meet the vertical lines at the vertices. The absorbed (emitted) photons are represented by incoming (outgoing) lines at the left (right) side of the vertices. The double vertical lines represent the electron propagator built up from the eigenenergies of H_0'. The analytical expression of ordered operators can be illustrated by means of diagrams like those of Fig. 2 which are read upwards.

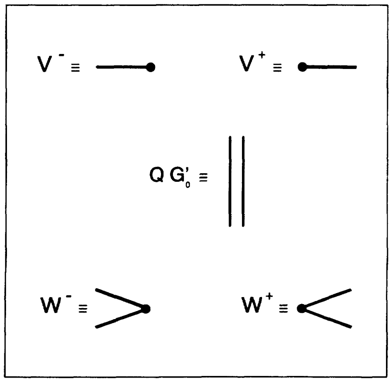

FIG. 1. Correspondence between the operators occurring in the formulas and the symbols used in the diagrammatic representation.

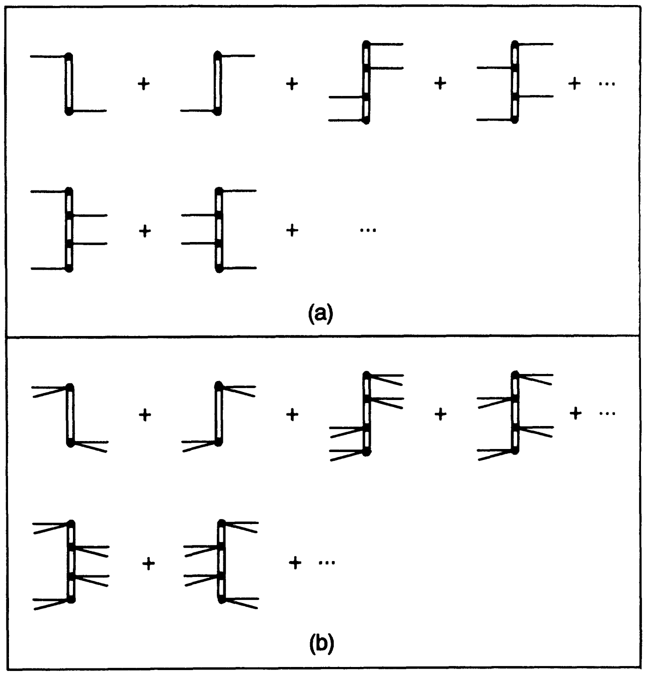

FIG. 2. (a) Diagrams representing second- and fourth-order contributions to the shift of an atomic level involving only single-photon interactions. (b) Fourth- and eighth-order forward-scattering diagrams involving double absorptions and emissions.

It is useful to adopt the following definitions concerning the topology of diagrams: (i) the forward scattering diagrams [12] are those displaying the same number of absorptions and emissions of photons between the same initial and final states; (ii) the reducible diagrams are those which can be split, at least, into two parts, one of them being a forward-scattering diagram, and (iii) to each direct diagram, there corresponds a mirror diagram obtained by interchanging the absorption and the emission lines. For brevity, only forward-scattering diagrams, as illustrated in Figs. 2 and 3, are considered in our analysis. In doing so, we do not restrict the generality of our results since the same discussion holds for processes involving the net absorption or emission of one or several photons and leads to the same conclusion.

Each diagram belongs to one of the four classes displayed in Figs. 2 and 3. The diagrams 2(a) and 2(b) contain only single and double interactions with equal and unequal numbers of double absorptions and double emissions, respectively. We discard the diagrams 2(a) from our discussion because they are identical to those encountered in the length gauge. Such diagrams have been studied in detail in previous accounts [11]. The only difference is that the energies of the electron lines are shifted by the operator K. For each class, the number of diagrams can be found in Table I up to eighth order. Apart from the class 2(a) where the number of diagrams of order $2k$ is given by the binomial coefficient $\binom{2k}{k}$, it is difficult to set up a general rule for the other classes.

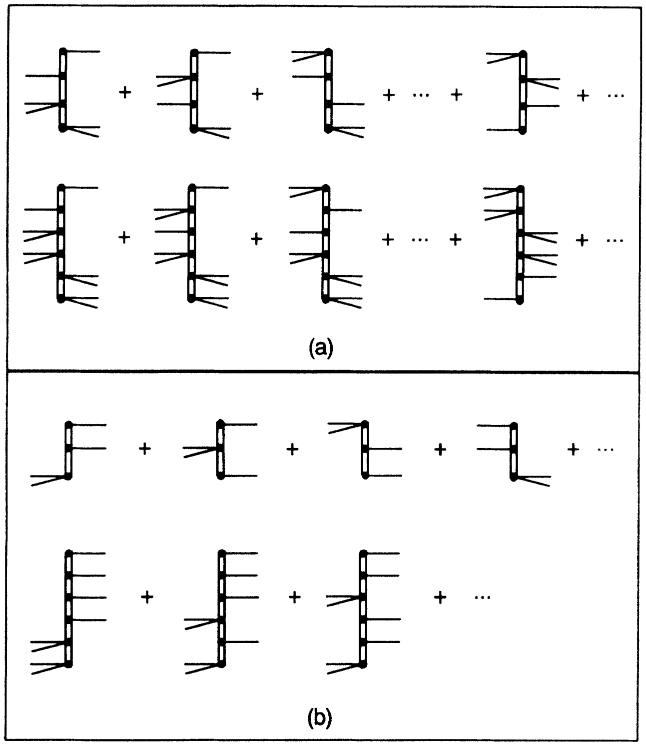

FIG. 3. (a) Sixth- and tenth-order corrections to a level shift involving an equal number of double absorptions and emissions. (b) Same as (a) except that the double absorptions and the double- photon emissions are unequal in number.

B. Calculation of the diagrams

The diagrams corresponding to the four classes previously mentioned are calculated order by order. The value of the diagrams belonging to the same class is obtained up to a common factor. In this way one sees what classes sum to zero and must be discarded. As was previously noted, the diagrams 2(a) are topologically identical to the corresponding diagrams in the length gauge. Therefore, the resulting resummed expression are straightforwardly derived from the preceding ones by shifting the energies of the continued fractions by the quiver energy K.

The diagrams 2(b) are obtained from the diagrams 2(a) by replacing the single interactions by double-photon absorptions and emissions. The number of diagrams of order $4k$ is also given by the binomial coefficient $\binom{2k}{k}$. This number comes from the contribution of $\frac{1}{2}\binom{2k}{k}$ pairs of diagrams (direct diagram and its mirror). After a little algebra, one finds that the contribution of any direct diagram is given, up to a constant factor, by $(1/2\omega)^{2k-1}$, while that of the corresponding mirror diagrams is $-(1/2\omega)^{2k-1}$. Therefore, all the graphs 2(b) of order $4k$ sum to zero.

TABLE I. The number of topologically different diagrams of second-, fourth-, sixth- and eighth-order contained in the classes displayed in Figs. 2(a), 2(b), 3(a), and 3(b).

Class / Order	2(a)	2(b)	3(a)	3(b)	Total
2	2	0	0	0	2
4	6	2	0	6	14
6	20	0	24	60	104
8	70	6	180	210	466

The reducible diagrams which appear in the ensemble of Fig. 3 lead to an overestimation of the sum. For example, the first and the fourth diagrams of Fig. 3(a) are topologically different from each other and their contributions, calculated for the state $|g\rangle$, are

$$D_1=\lim_{\eta\to 0}\sum_i \frac{(\boldsymbol{\varepsilon}\cdot\mathbf{p})_{gi}(\boldsymbol{\varepsilon}\cdot\mathbf{p})_{ig}}{(-2\omega+\eta)(\eta)(E_g+\omega-E_i+\eta)} \tag{4.1a}$$

and

$$D_2=\lim_{\eta\to 0}\sum_i \frac{(\boldsymbol{\varepsilon}\cdot\mathbf{p})_{gi}(\boldsymbol{\varepsilon}\cdot\mathbf{p})_{ig}}{(E_g+\omega-E_i+\eta)(\eta)(-2\omega+\eta)}\ , \tag{4.1b}$$

respectively.

Obviously, the quantities of Eqs. (4.1) are equal and counted twice. The origin of this overestimation can be found when one considers the product of the field operators. The sequence of the creation and destruction operators corresponding to D_1 and D_2 are $a^\dagger a^\dagger aaaa^\dagger$ and $aa^\dagger a^\dagger a^\dagger aa$, respectively. From the well-known boson commutation relation

$$[a,a^\dagger]_-=1\ , \tag{4.2}$$

one easily shows that

$$a^\dagger a^\dagger aaaa^\dagger=aa^\dagger a^\dagger a^\dagger aa\ . \tag{4.3}$$

The equality of the contributions of D_1 and D_2 is not accidental. It comes from the commutation of some parts of diagrams containing blocks of operators. In the example we are considering, one has from Eq. (4.3),

$$[a^\dagger a^\dagger aa,aa^\dagger]_-=0\ , \tag{4.4}$$

which means that, for these reducible diagrams, the double emission-absorption processes can indifferently occur before or after the single absorption-emission one. We note that the splitting of the operator blocks takes place at the junction of two forward-scattering graphs. This rule is general and applies to every higher-order reducible diagram. Furthermore, this overestimation comes from the classes of diagrams containing single- and double-photon interactions with an equal number of double absorptions and double emissions. Thus, in addition to the elimination of multiple counting which consists in keeping only topologically different diagrams, one has to avoid overcontributions provided by the reducible diagrams.

C. Mixing of single and double photointeractions

We calculate the contributions of the diagrams belonging to the classes displayed in Figs. 3(a) and 3(b). Due to the summations coming from the electron propagators, these contributions result from the sum of many terms which an be arranged in groups characterized by a set of atomic states. For each order and inside each group, the diagrams only differ from each other by the energy of their electron lines, not by their vertex factors (matrix elements). As an example, the analytical expression corresponding to a diagram where p interactions $V^{\pm}$ are mixed with q interactions $W^{\pm}$ is written, up to a common factor, in the form of a product of $p+q-1$ energy denominators. For this discussions, the diagrams are designated through ensembles of numbers representing the processes taking place at the vertices. In front of the number of photons, the plus and the minus signs denote the photon absorptions and emissions, respectively. Thus, the first and the second diagrams of Fig. 3(a) will be labeled $(-2,+2,-1,+1)$ and $(-2,+1,+2,-1)$, respectively. In examining the sixth-order processes of Fig. 3(a), we find 24 topologically different diagrams obtained by considering all the possible combinations of the four kinds of absorption and emission operators. There are four pairs of identical reducible diagrams. By using the result of Sec. IV B, the overabundant terms may be eliminated. Four reducible diagrams remain which can be labelled as $(-2,+2,-1,+1)$, $(-2,+2,+1,-1)$, $(+2,-2,-1,+1)$, and $(+2,-2,+1,-1)$, while the four diagrams which are to be dropped are denoted by $(-1,+1,-2,+2)$, $(+1,-1,-2,+2)$, $(+2,-2,-1,+1)$, and $(+2,-2,+1,-1)$. The analytical expressions corresponding to the four remaining diagrams are

$$D_1=\beta \lim_{\eta\to 0} \sum_i \frac{d_{gi}d_{ig}}{\eta(-2\omega+\eta)(\omega_g-\omega_i-\omega+\eta)} , \tag{4.5a}$$

$$D_2=\beta \lim_{\eta\to 0} \sum_i \frac{d_{gi}d_{ig}}{\eta(-2\omega+\eta)(\omega_g-\omega_i+\omega+\eta)} , \tag{4.5b}$$

$$D_3=\beta \lim_{\eta\to 0} \sum_i \frac{d_{gi}d_{ig}}{\eta(2\omega+\eta)(\omega_g-\omega_i-\omega+\eta)} , \tag{4.5c}$$

$$D_4=\beta \lim_{\eta\to 0} \sum_i \frac{d_{gi}d_{ig}}{\eta(2\omega+\eta)(\omega_g-\omega_i+\omega+\eta)} . \tag{4.5d}$$

In Eqs. (4.5), β is the intensity parameter derived from Eqs. (3.5) $\{\beta=[1/(4\omega^6)](I/I_0)^3\}$ and $d=(\varepsilon\cdot\mathbf{p})$. The summations run over the whole atomic spectrum. After elementary algebra, $D=D_1+D_2+D_3+D_4$ can be put in the form

$$D=\beta\sum_i d_{gi}d_{ig}\frac{\omega_{gi}}{\omega^2(\omega_{gi}^2-\omega^2)} . \tag{4.6}$$

In spite of the large value that each of the four divergent diagrams of Eqs. (4.5) can reach, their sum is finite and cancels exactly the contribution of the 16 remaining ones. Such a property is verified numerically to any order in the present work.

The same discussions holds for the eight-order graphs of Fig. 3(a). Here, the problem is a little more complicated because among the 180 diagrams, 156 are irreducible, while 24 are reducible. An algorithm has been found to classify, eliminate overabundant terms, and calculate the contribution of each diagram. By using the commutation rules of Sec. IV B, one finds that the sum of the 12 remaining diagrams (i) is finite, (ii) is independent of the parameter η, and (iii) cancels the sum of the 156 irreducible diagrams exactly.

The difficulty arising from the diagrams of Fig. 3(b) is solved by using the previous method. A group of 15 diagrams involving two double absorptions can be isolated from the 210 eighth-order graphs of Fig. 3(b). Among them, one finds nine reducible diagrams. In contrast to the preceding case, none of them is identical to any of the six remaining ones by changing the order of the propagation-vertex operators. This is the consequence of the asymmetry between the number of double absorptions and double emissions. For example, the third eighth-order diagram of Fig. 3(b) is reducible and can be split into a double absorption followed by two single emissions. The commutation of these blocks does not change the topology of the graph. Therefore, it is unique and its contribution is counted once. Such a property holds for all the diagrams of class 3(b). The main points of the discussion can be summarized through the following general rules: (a) When the number of double absorptions and double emission are equal, the contribution of the reducible diagrams is large and compensates that of the irreducible ones; (b) in the case of unequal numbers of double interactions, the corresponding contributions are different from each other and both of them vanish separately; and (c) by generalizing the analysis to any order, one finds that all the diagrams involving double absorptions and/or double emission of photons are to be discarded from the computation of the operator $S'(z)$.

V. OPERATOR EXPRESSIONS

It is worthwhile to note that in any all-order theory, one is faced with divergences due to reducible diagrams. The previous investigations allow us to avoid those contained in diagrams involving double-photon interactions [Figs. 3(a) and 3(b)]. Nevertheless, divergences still remain in the reducible diagrams of Fig. 2(a) involving single-photon interactions. They come from the renormalization of external electron lines. As has been shown in the preceding numerical analysis, these divergences can be avoided by summing the perturbation series. such a property is widely utilized in theories resorting to projector techniques [9–11].

According to what is done in the length gauge, one defines the operators P and Q which are the projectors onto and outside the subspace (ε) spanned by particular dressed states of the system atom plus field. In general, these states are those which are degenerated by the presence of resonances. In this respect, we recall that two states are degenerated when their energies differ from each other by an integer number of photons. Such an event can also happen in nonresonant processes when the initial or the final sate is found as an intermediate state in the expansion of the Green function. By introducing the projectors P and Q, the shift operator $S'(z)$ given by Eq. (3.13) transforms into

$$\begin{aligned} R'(z)={}&H_I'+H_I'QG_0'(z)H_I' \\ &+H_I'QG_0'(z)H_I'QG_0'(z)H_I'+\cdots , \end{aligned} \tag{5.1}$$

with the obvious relations

$$P+Q=1\ , \tag{5.2a}$$

$$PQ=0\ . \tag{5.2b}$$

From the preceding lines, the resummed expressions in the velocity gauge can be easily derived from those already obtained in the length gauge [5]. One has to replace H_0 everywhere by H_0' [the intensity-dependent Hamiltonian given by Eq. (3.7)] and to use the interaction operators $V^{\pm}$ of Eq. (3.5a). In doing so, one finds that the diagonal part of the shift operator $R'(z)$ sums to

$$R'^{(D)}(z)=[V^{+}\bar{\tau}'_{\square}\bar{V}^{-}\bar{\tau}'_{\bigcirc}+V^{-}\bar{\tau}'_{\square}\bar{V}^{+}\bar{\tau}'_{\varnothing}[(z-H_0')\ , \tag{5.3}$$

where $\bar{\tau}'_{\square}$, $\bar{\tau}'_{\bigcirc}$, and $\bar{\tau}'_{\varnothing}$ are continued fractions of $\bar{V}^{\pm}=QV^{\pm}$.

The resummed part of $R'(z)$ accounting for the effective absorption and emission of N photons are

$$R'^{(ND)}(z)=[V^{+}\bar{\tau}'_{\square}(\bar{V}^{-}\bar{\tau}'_{\bigcirc})^{N+1} + V^{-}\bar{\tau}'_{\square}(\bar{V}^{-}\bar{\tau}'_{\bigcirc})^{N-1}](z-H_0') \tag{5.4a}$$

and

$$R'^{(ND)}(z)=[V^{-}\bar{\tau}'_{\square}(\bar{V}^{+}\bar{\tau}'_{\varnothing})^{N+1} + V^{+}\bar{\tau}'_{\square}(\bar{V}^{+}\bar{\tau}'_{\varnothing})^{N-1}](z-H_0')\ , \tag{5.4b}$$

respectively.

The τ operators are the following continued fractions of the interaction $V^{\pm}$:

$$\bar{\tau}'_{\bigcirc}=\frac{1}{z-H_0'-\bar{V}^{-}\bar{\tau}'_{\bigcirc}\bar{V}^{+}}\ , \tag{5.5a}$$

$$\bar{\tau}'_{\varnothing}=\frac{1}{z-H_0'-\bar{V}^{+}\bar{\tau}'_{\varnothing}\bar{V}^{-}}\ , \tag{5.5b}$$

$$\bar{\tau}'_{\square}=\frac{1}{z-H_0'-\bar{V}^{-}\bar{\tau}'_{\bigcirc}\bar{V}^{+}-\bar{V}^{+}\bar{\tau}'_{\varnothing}\bar{V}^{-}}\ . \tag{5.5c}$$

These expressions are formally identical to the ones derived in the length gauge. The only, but important, difference is that the interaction is now $\alpha\,\mathbf{A}\cdot\mathbf{p}$ instead of being $\mathbf{E}\cdot\mathbf{r}$, whereas the whole spectrum is shifted upward. We note that in most cases, the transition amplitudes involve energy differences. Therefore, the shift due to the $\mathbf{A}^2$ term, which is independent of the atomic operators, is canceled and does not influence the final result.

VI. CONCLUSION

The problem concerning the calculation of level shifts and transition probabilities in the velocity gauge has been examined within standard semiclassical and quantized models. It has been shown that a contact transformation which cancels the whole contribution of the quadratic term of the interaction Hamiltonian is not suitable to set up the correct expressions of the second-order Stark shift and the resonance-fluorescence probability. An alternate transformation has been proposed which restores the lacking term that is lacking in the corresponding formulas. Its implications have been studied on second-order processes by resorting to a semiclassical model. Within this framework, it has been shown that the interactions involving twice the frequency of the radiation field could be discarded. The generalization of this property to arbitrary order has been examined by using a fully quantized theory. A suitable resummation of diagrams involving single- and double-photon absorptions and emissions shows the importance of calculating carefully the reducible diagrams which account for the corrections of the external lines. These diagrams play an important role concerning the gauge invariance of the theory [13]. By computing order by order the contributions of all the diagrams belonging to the same class displaying at least one double-photon interaction, we have found that the sum of these diagrams did not have to be considered. Such a law, which has been already enunciated for lowest-order processes [3,4], is confirmed and can be generalized to arbitrary order from the present work.

Thus, in the velocity gauge, the double absorptions or emissions coming from $(\alpha^2/2)\,\mathbf{A}^2(t)$ (aa and $a^{\dagger}a^{\dagger}$ terms) do not contribute to any order. Only the processes of absorption emission of a photon at the same vertex ($aa^{\dagger}$ and $a^{\dagger}a$ terms) need to be retained because they induce an intensity-dependent shift of the whole spectrum. This rule has permitted us to write resummed operator expressions which will serve to make nonperturbative calculations in the case of radiation-matter interaction at very high intensity.

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