tions may be important in ionic rare-earth compounds (and perhaps also in other systems). It is found that there are a number of competing effects which may or may not point to a relative enhancement of the higher-degree terms, but there is certainly no clear cut evidence that they are negligible compared with the usually considered electric quadrupole-quadrupole interactions. The effect of higher-degree terms on optical nonradiative energy-transfer processes is shown to be largely quantitative rather than qualitative, any observable effects arising principally from the less stringent selection rules resulting from the extra higher-degree terms. The effects on magnetic properties are also similar to those produced by the lower-degree quadrupole terms, except for the fact that the extra terms may produce appreciable contributions to components of the interaction and g-value tensors which are otherwise zero or very small. Unless it can be shown, therefore, that the higher-degree electric multipole terms are in fact negligible one must expect a completely general form for both the effective spin-spin interaction, $\mathbf{S}_i'\cdot\mathbf{K}\cdot\mathbf{S}_j'$ and the Zeeman interaction $\mu_B\mathbf{H}\cdot(\mathbf{g}_i\cdot\mathbf{S}_i'+\mathbf{g}_j\cdot\mathbf{S}_j')$ describing a pair of $S'=\frac{1}{2}$ ions. We have not considered situations of higher degeneracy or the case of non-Kramers ions but the same sort of complications will apply. The calculation of the components of the $\mathbf{K}$ and $\mathbf{g}$ tensors will generally be very difficult but in favorable cases they may be simplified appreciably by symmetry which, as always, remains as the only reliable guide to the possible anisotropy.

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Coherent Field Dynamics in First- and Second-Order Processes

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The problem of the interaction of a beam of radiation in a coherent state and an atomic system is investigated. A complete set of states of the field is introduced, called displaced stationary states, which are found to be particularly convenient in dealing with first- and second-order processes such as dispersion, Raman scattering, single and double emission, and absorption. In this formalism the resonant conditions for the occurrence of processes which have their counterpart in ordinary first- and second-order perturbation theory (where the state of the field is assumed to be a stationary state) are found in a natural way. In addition, a description is indicated of physical situations which are peculiar to coherent field dynamics and thus have no such counterpart. The coherence properties of the displaced stationary states are analyzed. It is shown under which experimental conditions (such as appropriate photon correlation methods) it would appear to be possible to verify whether the displaced stationary states indeed provide a good description of the underlying dynamics. The formalism can readily be extended to higher-order processes.

I. INTRODUCTION

IN quantum optics, with the advent of the laser, one faces the problem of describing elementary first- and second-order processes such as absorption and emission, dispersion and Raman effect, and double absorption and emission. If one treats the output of the laser as a coherent state (in the sense of Glauber[1]) and considers the interaction of such a state with a material system such as an atom or molecule, one is faced with the problem of the coherent state (unlike the stationary state with a definite number of photons) not being an eigenstate of the unperturbed Hamiltonian of the radiation field. The problem one faces is to find a representation of the Schrödinger equation suitable to convey a physically meaningful description of the atomic system interacting with the coherent field. In Sec. II, we introduce a complete set of states of the radiation field, the displaced stationary states, which we arrive at by carrying out a unitary transformation of the Schrödinger equation and representing the transformed equation in Fock space. In Sec. III, we deal with the problem of absorption and emission when the atomic system is interacting with a coherent field or with a displaced stationary state field. We find that in addition to a description bearing a close resemblance to the usual one prevailing when the initial

[1] R. J. Glauber, Phys. Rev. **131**, 2766 (1963).

state is a stationary field, a situation arises where the atomic system makes a transition and the final state of the field is the same as the initial one. In Sec. IV, we deal with second-order processes. We find that in addition to the processes of dispersion, Raman scattering, and double absorption and emission, which have their counterpart as ordinary second-order processes, there exists a description of processes having no such counterpart, which are thus peculiar to the initial coherent field. In Sec. V we discuss the coherence properties of the displaced stationary states. We find that states, for instance, of the form $V_k a_k^\dagger \mid 0\rangle$ do not have first- or higher-order coherence. They would thus exhibit photon correlations in appropriate photon-counting experiments. We make the prediction that if such states exist and interact with an atomic system, they can make transitions to states $V \mid 0\rangle = \mid \alpha\rangle$, which are coherent to all orders and thus do not exhibit photon correlations. An experimental way is thus indicated for testing the reality of the states $V_k a_k^\dagger \mid 0\rangle$.

II. GENERAL REPRESENTATION IN TERMS OF DISPLACED STATIONARY STATES

Unitary Transformation of the Equations of Motion

The dynamical problem consists, in principle, in solving a Schrödinger equation, which in the Dirac picture becomes

$$i\hbar(\partial/\partial t)\,U(t,t_0)\mid\psi(t_0)\rangle = H(t)\,U(t,t_0)\mid\psi(t_0)\rangle, \qquad (2.1)$$

where $U(t,t_0)$ is the usual time-displacement operator. The vector $\mid\psi(t_0)\rangle$ is the state of the combined material and radiation systems at time $t=0$. If the initial state of the radiation field is a stationary state, i.e., a state with a fixed number of photons, given by $\mid n\rangle_k = a_k^{\dagger n_k}/(n_k!)^{1/2}\mid 0\rangle$ (here $\mid 0\rangle$ is the vacuum state, $a_k^\dagger$, a_k are photon creation and annihilation operators, respectively, for the mode k), Eq. (2.1) is most appropriately represented in Fock space, and in the eigenstates of the unperturbed Hamiltonian of the material system. In this section we deal mainly with finding a suitable representation of Eq. (2.1) when the initial state of the radiation field is given by a so-called coherent state, described by

$$\mid\alpha\rangle_k = \exp(-\tfrac{1}{2}\mid\alpha_k\mid^2) \times \sum_{n_k=0}^{\infty}[\alpha_k^{n_k}/(n_k!)^{1/2}][a_k^{\dagger n_k}/(n_k!)^{1/2}]\mid 0\rangle, \qquad (2.2)$$

where α_k is a complex number. In this section we are only concerned with a single mode (and henceforth omit the subscript k), although our results can easily be generalized to the many-mode case. If we denote by $\mid i,\alpha\rangle$ the combined initial state of the atom (denoted by i) and of the field (denoted by α), we can rewrite Eq. (2.1) as

$$i\hbar(\partial/\partial t)\,U\mid i,\alpha\rangle = HU\mid i,\alpha\rangle. \qquad (2.3)$$

The displacement operator $V(\alpha)$, introduced by Glauber,[1] allows us to write the coherent state of Eq. (2.2) as

$$\mid\alpha\rangle = V\mid 0\rangle = \exp(\alpha a^\dagger - \alpha^* a)\mid 0\rangle. \qquad (2.4)$$

It will be remembered that V is a unitary operator. Because of this property, Eq. (2.3) can be written, after being multiplied to the left by V^{-1}, as

$$i\hbar(\partial/\partial t)V^{-1}UV\mid i,0\rangle = (V^{-1}HV)V^{-1}UV\mid i,0\rangle \qquad (2.5a)$$

or

$$i\hbar(\partial/\partial t)\,U'\mid i,0\rangle = H'U'\mid i,0\rangle. \qquad (2.5b)$$

Here the state $\mid i,0\rangle$ is an eigenstate of the original unperturbed Hamiltonian of the combined system. It would thus appear that a natural representation of Eq. (2.5a) be in a space spanned by the eigenvectors of the original unperturbed Hamiltonian, which is Fock space for the radiation field. We wish to evaluate the transformed Hamiltonian $H' \equiv V^{-1}HV$. We choose

$$H = -C^{-1}\exp[(i/\hbar)H_0 t]\int \mathbf{j}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})\,d^3r \times \exp[-(i/\hbar)H_0 t] = -C^{-1}\int \mathbf{j}(\mathbf{r},t)\cdot\mathbf{A}(\mathbf{r},t)\,d^3r \qquad (2.6)$$

as a typical interaction Hamiltonian in the Dirac picture, where H_0 is the unperturbed Hamiltonian of the combined material and radiation systems, and

$$H_{IS} = \int \mathbf{j}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})\,d^3r$$

is the interaction term of the total Hamiltonian in the Schrödinger picture.

We have the vector potential operator

$$\mathbf{A}(\mathbf{r}) = \mathbf{A}^{(+)}(\mathbf{r},a_{k_1}) + \mathbf{A}^{(-)}(\mathbf{r},a_{k_1}^\dagger), \qquad (2.7)$$

with

$$\mathbf{A}^{(+)}(\mathbf{r},a_{k_1}) = C\sum_{k_1}(\hbar/2\omega_{k_1}V_0)^{1/2}\mathbf{l}_{k_1}a_{k_1}\exp(i\mathbf{k}_1\cdot\mathbf{r}), \qquad (2.8a)$$

$$\mathbf{A}^{(-)}(\mathbf{r},a_{k_1}^\dagger) = C\sum_{k_1}(\hbar/2\omega_{k_1}V_0)^{1/2}\mathbf{l}_{k_1}a_{k_1}^\dagger\exp(-i\mathbf{k}_1\cdot\mathbf{r}), \qquad (2.8b)$$

and the following equalities:

$$V_k^{-1}\exp(ia_k^\dagger a_k t)\,a_k\exp(-ia_k^\dagger a_k t)\,V_k = V_k^{-1}a_k\exp(-i\omega_k t)\,V_k = (a_k+\alpha_k)\exp(-i\omega_k t), \qquad (2.9a)$$

$$V_k^{-1}\exp(ia_k^\dagger a_k t)\,a_k^\dagger\exp(-ia_k^\dagger a_k t)\,V_k = V_k^{-1}a_k^\dagger\exp(i\omega_k t)\,V_k = (a_k^\dagger+\alpha_k^*)\exp(i\omega_k t). \qquad (2.9b)$$

The equalities of the extreme right of Eqs. (2.9a), and (2.9b) follow from the displacement theorem.[1]

We define

$$L^{(+)}(\alpha_k)=\int \mathbf{j}(\mathbf{r})\cdot\mathbf{A}^{(+)}(\mathbf{r},\alpha_k)\,d^3r, \qquad (2.10a)$$

$$L^{(-)}(\alpha_k^*)=\int \mathbf{j}(\mathbf{r})\cdot\mathbf{A}^{(-)}(\mathbf{r},\alpha_k^*)\,d^3r, \qquad (2.10b)$$

where we have replaced a_k by α_k in $\mathbf{A}^{(+)}(\mathbf{r}, a_k)$ and $a_k^\dagger$ by α_k^* in $\mathbf{A}^{(-)}(\mathbf{r}, a_k^\dagger)$. Using Eqs. (2.6)–(2.10b), the transformed Hamiltonian is given by

$$\begin{aligned}
V_k^{-1}HV_k &= V_k^{-1}\exp[(i/\hbar)H_0t]\int \mathbf{j}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})\,d^3r\exp[(-i/\hbar)H_0t]V_k\\
&=\exp[(i/\hbar)H_0t]\int \mathbf{j}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})\,d^3\mathbf{r}\exp[(-i/\hbar)H_0t]\\
&\quad+\Big(\exp[(i/\hbar)H_mt]\int \mathbf{j}(r)\cdot\mathbf{A}^{(+)}(\mathbf{r},\alpha_k)\exp[-(i/\hbar)H_mt]\Big)\exp(-i\omega_kt)\\
&\quad+\Big(\exp[(i/\hbar)H_mt]\int \mathbf{j}(r)\cdot\mathbf{A}^{(-)}(\mathbf{r},\alpha_k^*)\exp[-(i/\hbar)H_mt]\Big)\exp(i\omega_kt)\\
&=H+\exp[(i/\hbar)H_mt][L^{(+)}(\alpha_k)\exp(-i\omega_kt)+L^{(-)}(\alpha_k^*)\exp(i\omega_kt)]\exp[-(i/\hbar)H_mt]\\
&\equiv H+Q(t,k), \qquad (2.11)
\end{aligned}$$

where we define H_m as the unperturbed Hamiltonian of the material system in the Schrödinger picture. In the case where the incident radiation consists of several modes $\prod_k V_k \mid 0\rangle$, we have, in general,

$$\begin{aligned}
\prod_k V_k^{-1}H\prod_k V_k &= H+\sum_k \exp[(i/\hbar)H_mt][L^{(+)}(\alpha_k)\\
&\quad\times\exp(-i\omega_kt)+L^{(-)}(\alpha_k^*)\exp(i\omega_kt)]\\
&\quad\times\exp[-(i/\hbar)H_mt]. \qquad (2.12)
\end{aligned}$$

The first term on the right of Eq. (2.12) is the original interaction Hamiltonian in the Dirac picture. The second term is an operator of the material system only, i.e., it is a c number in the field operators. It therefore follows that Eq. (2.12), taken between two stationary states $\mid n\rangle$ and $\langle m \mid$, of photon number n and m, respectively, becomes,

$$\langle m \mid V_k^{-1}HV_k \mid n\rangle=\langle m \mid H \mid n\rangle+Q(t,k)\delta_{mn}. \qquad (2.13)$$

Note that here the transformed Hamiltonian has matrix elements in Fock space between stationary states whose photon number differ by ±1, 0. In Eq. (2.13) this result follows from the choice of an interaction Hamiltonian given by Eq. (2.6), which is linear in the vector potential A. Had we added the nonrelativistic quadratic term in the vector potential, the coupling between stationary states of the transformed interaction Hamiltonian would still have been between states differing by a small number of photons. It therefore appears to be physically meaningful to represent the field part of Eq. (2.5a) or (2.5b) in Fock space (regarding $V^{-1}HV$ as the interaction Hamiltonian), or to represent Eqs. (2.5a) and (2.5b) in the space spanned by the eigenstates of the unperturbed Hamiltonian H_0. This means that we work with states of the field of the form $V \mid n\rangle$, which we call displaced stationary states.

Displaced Stationary States

Considering, as before, only one mode, the displaced stationary state vectors are of the form

$$V[a^{\dagger n}/(n!)^{1/2}] \mid 0\rangle\equiv V \mid n\rangle \qquad n=0, 1, \cdots, \infty. \qquad (2.14)$$

It is readily verified that the states given by expression (2.14) represent a complete set, satisfying the completeness relation

$$\sum_{n=0}^{\infty} V\frac{a^{\dagger n}}{(n!)^{1/2}} \mid 0\rangle\langle 0 \mid \frac{a^n}{(n!)^{1/2}}V^{-1}=1. \qquad (2.15)$$

Clearly, Eq. (2.15) can be written

$$V\left(\sum_{n=0}^{\infty}\frac{a^{\dagger n}}{(n!)^{1/2}} \mid 0\rangle\langle 0 \mid \frac{a^n}{(n!)^{1/2}}\right)V^{-1}=VV^{-1}=1, \qquad (2.16)$$

because of the unitarity property of the operator V. It might be of interest to be able to write the states (2.14) in a form in which the operator V acts on $\mid 0\rangle$ first, i.e., in terms of the state $\mid \alpha\rangle$. We write expression (2.14) as

$$[(Va^\dagger V^{-1})^n/(n!)^{1/2}]V \mid 0\rangle \qquad n=0, 1, \cdots, \infty, \qquad (2.17)$$

and evaluate $Va^\dagger V^{-1}$. By the displacement theorem,[1] we have

$$\begin{aligned} a^\dagger &= V[V^{-1}a^\dagger V]V^{-1} \\ &= V[a^\dagger+\alpha^*]V^{-1} \\ &= Va^\dagger V^{-1}+\alpha^* VV^{-1} \\ &= Va^\dagger V^{-1}+\alpha^*. \end{aligned} \tag{2.18}$$

Hence

$$Va^\dagger V^{-1}=a^\dagger-\alpha^*. \tag{2.19}$$

Using Eq. (2.19) and the expression (2.17), we obtain for expression (2.14) the desired form

$$\begin{aligned} V \mid n\rangle &= [(a^\dagger-\alpha^*)^n/(n!)^{1/2}]V \mid 0\rangle \\ &= [(a^\dagger-\alpha^*)^n/(n!)^{1/2}] \mid \alpha\rangle \qquad n=0, 1, \cdots, \infty. \end{aligned} \tag{2.20}$$

The completeness relation (2.15) can thus also be written as

$$\sum_{n=0}^{\infty} \frac{(a^\dagger-\alpha^*)^n}{(n!)^{1/2}} V \mid 0\rangle\langle 0 \mid V^{-1} \frac{(a-\alpha)^n}{(n!)^{1/2}} = 1. \tag{2.21}$$

III. FIRST-ORDER PROCESSES

From Eq. (2.13) it is apparent that $V_k^{-1}HV_k$ has matrix elements either of the form $\langle m \mid H \mid n\rangle$ or Q, depending upon whether m differs from n by ± 1 or by 0, respectively. This would indicate, that in first order, for an initial state $V \mid n\rangle$ the final state can be either $\langle n \mid V^{-1}$ or $\langle n\pm 1 \mid V^{-1}$. We wish to calculate the matrix elements in first-order perturbation theory of the operator $U(t)$, which we designate by $U_{(1)}(t)$. We assume that we have a two-level atomic system for the material system, with energy levels E_i, (energy of the initial state), and E_f (energy level of the final state), and with states $\mid i\rangle$ and $\mid f\rangle$, respectively. We consider the following situations:

(1) Initial state E_i, excited state. Final state E_f, ground state. State of the field $V_k \mid n_k\rangle$.

In the light of Eq. (2.13) we then have two cases. The first case is

$$\begin{aligned} &\langle 0, f \mid \{a_k^{n_k+1}/[(n_k+1)!]^{1/2}\} V^{-1}U_{(1)}(t)\, V[a_k^{\dagger n_k}/(n_k!)^{1/2}] \mid i, 0\rangle \\ &\quad = (i\hbar)^{-1} \int_0^t \langle 0, f \mid \frac{a_k^{n_k+1}}{[(n_k+1)!]^{1/2}} V_k^{-1}H(t')\, V_k \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle dt' \\ &\quad = (i\hbar)^{-1} \int_0^t \langle 0, f \mid \frac{a_k^{n_k+1}}{[(n_k+1)!]^{1/2}} H(t') \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle\, dt' \\ &\quad = (i\hbar)^{-1} \int_0^t dt' \exp[i(\omega_f-\omega_i)t']\langle f \mid L_k^{(-)}(1) \mid i\rangle (n_k+1)^{1/2} \exp(i\omega_k t'). \end{aligned} \tag{3.1}$$

This is the well-known expression for induced and spontaneous emission. The atomic system makes a transition to the ground state and the final state of the field is $\langle 0 \mid \{a_k^{n_k+1}/[(n_k+1)!]^{1/2}\} V_k^{-1}$. The process takes place under the resonant condition

$$\omega_k=\omega_i-\omega_f. \tag{3.2}$$

If the initial state of the field were $V \mid 0\rangle = \mid \alpha\rangle$, we would have a final state $\langle 0 \mid a_{k'}V_k^{-1}$, with $k'=k$ or $k'\neq k$. This would correspond to spontaneous emission.

The second case is

$$\begin{aligned} &\langle 0, f \mid [a_k^{n_k}/(n_k!)^{1/2}] V_k^{-1}U_{(1)}(t)\, V_k[a_k^{\dagger n_k}/(n_k!)^{1/2}] \mid i, 0\rangle \\ &\quad = (i\hbar)^{-1} \int_0^t \langle 0, f \mid \frac{a_k^{n_k}}{(n_k!)^{1/2}} V_k^{-1}H(t')\, V_k \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle\, dt' \\ &\quad = (i\hbar)^{-1} \int_0^t \langle 0, f \mid Q(t') \mid i, 0\rangle\, dt' \\ &\quad = (i\hbar)^{-1} \int_0^t dt' \exp[i(\omega_f-\omega_i)t']\langle f \mid L_k^{(-)}(\alpha_k^*) \mid i\rangle \exp(i\omega_k t'), \end{aligned} \tag{3.3}$$

with again the condition (3.2). Here the atomic system makes a transition to the ground state but the final state of the field is the same as the initial state $V_k \mid n_k\rangle$.

(2) Initial state E_i, ground state, final state E_f, excited state. State of the field $V_k \mid n_k\rangle$.

Here again, two cases. The first case is

$$\langle 0,f\,|\,\{a_k{}^{n_k-1}/[(n_k-1)!]\}V_k{}^{-1}U_{(1)}(t)\,V_k[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,i,0\rangle$$

$$=(i\hbar)^{-1}\int_0^t\langle 0,f\,|\,\frac{a_k{}^{n_k-1}}{[(n_k-1)!]^{1/2}}\,V_k{}^{-1}H(t')\,V_k\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\,dt'$$

$$=(i\hbar)^{-1}\int_0^t\langle 0,f\,|\,\frac{a_k{}^{n_k-1}}{[(n_k-1)!]^{1/2}}\,H(t')\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\,dt'$$

$$=(i\hbar)^{-1}\int_0^t dt'\exp[i(\omega_f-\omega_i)t']\langle f\,|\,L_k{}^{(+)}(1)\,|\,i\rangle(n_k)^{1/2}\exp(-i\omega_k t'). \quad (3.4)$$

This is the well-known expression for absorption. The atomic system makes a transition from the ground state to the excited state. The final state of the field is $\langle 0\,|\,\{a^{n_k-1}/[(n_k-1)!]^{1/2}\}V_k{}^{-1}$. The process takes place under the resonant condition

$$\omega_k=\omega_f-\omega_i. \quad (3.5)$$

The second case is

$$\langle 0,f\,|\,[a_k{}^{n_k}/(n_k!)^{1/2}]V_k{}^{-1}U_{(1)}(t)\,V_k[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,i,0\rangle$$

$$=(i\hbar)^{-1}\int_0^t\langle 0,f\,|\,\frac{a_k{}^{n_k}}{(n_k!)^{1/2}}\,V_k{}^{-1}H(t')\,V_k\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\,dt'$$

$$=(i\hbar)^{-1}\int_0^t\langle 0,f\,|\,Q(t')\,|\,i,0\rangle\,dt'$$

$$=(i\hbar)^{-1}\int_0^t dt'\exp[i(\omega_f-\omega_i)t']\langle f\,|\,L_k{}^{(+)}(\alpha_k)\,|\,i\rangle\exp(-i\omega_k t'), \quad (3.6)$$

with the condition (3.5). The atomic system makes a transition to the excited state and the initial and final states of the field remain unchanged, so that the new physical feature of the states $V_k\,|\,n_k\rangle$ is that, in addition to the usual process of absorption and emission given by Eqs. (3.1) and (3.4), we now also have, under the same conditions (3.2) and (3.5), the processes described by Eqs. (3.3) and (3.6), whereby the atomic system makes the up or down transition leaving the state of the field unchanged.

IV. SECOND-ORDER PROCESSES

For a description of second-order processes we seek the appropriate matrix elements in second-order perturbation theory of the operator $U(t)$, which we designate by $U_{(2)}(t)$, i.e.,

$$U_{(2)}(t)=[(i\hbar)^{-1}]^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2 H(\tau_1)H(\tau_2). \quad (4.1)$$

The representation of $U_{(2)}(t)$ is in a space given by Eq. (2.15). We can therefore choose for example a matrix element of the form

$$\langle 0,f\,|\,\prod_k[a_k{}^{m_k}/(m_k!)^{1/2}]V_{k_1}{}^{-1}V_{k_2}{}^{-1}U_{(2)}(t)\,V_{k_2}V_{k_1}\prod_k[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,i,0\rangle, \quad (4.2)$$

where, as before, $|\,i\rangle$ and $|\,f\rangle$ designate the initial and final states, respectively, of the material system with corresponding energy levels $E_i=\hbar\omega_i$ and $E_f=\hbar\omega_f$. Substituting Eq. (4.1) into (4.2) we have

$$\langle 0,f\,|\,\prod_k[a_k{}^{m_k}/(m_k!)^{1/2}]V_{k_1}{}^{-1}V_{k_2}{}^{-1}U_{(2)}(t)\,V_{k_2}V_{k_1}\prod_k[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,i,0\rangle$$

$$=[(i\hbar)^{-1}]^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\prod_k\frac{a_k{}^{m_k}}{(m_k!)^{1/2}}\,V_{k_1}{}^{-1}V_{k_2}{}^{-1}H(\tau_1)H(\tau_2)V_{k_2}V_{k_1}\prod_k\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle. \quad (4.3)$$

We wish to evaluate

$$V_{k_1}{}^{-1}V_{k_2}{}^{-1}H(\tau_1)H(\tau_2)\,V_{k_2}V_{k_1}=V_{k_1}{}^{-1}V_{k_2}{}^{-1}H(\tau_1)\,V_{k_1}V_{k_2}V_{k_1}{}^{-1}V_{k_2}{}^{-1}H(\tau_1)\,V_{k_2}V_{k_1}. \quad (4.4)$$

Now, we know from Eq. (2.12) that

$$V_{k_1}^{-1}V_{k_2}^{-1}H(\tau_1)\,V_{k_2}V_{k_1}=H(\tau_1)+Q(\tau_1,k_1)+Q(\tau_1,k_2), \tag{4.5}$$

where we define as before in Eq. (2.11),

$$Q(\tau,k)\equiv M^{(-)}(\tau)[L_k^{(+)}(\alpha_k)\,\exp(-i\omega_k\tau)+L_k^{(-)}(\alpha_k^*)\,\exp(i\omega_k\tau)]M^{(+)}(\tau), \tag{4.6}$$

$$M^{(-)}(\tau)\equiv\exp[(i/\hbar)H_m\tau], \tag{4.7a}$$

$$M^{(+)}(\tau)\equiv\exp[-(i/\hbar)H_m\tau]. \tag{4.7b}$$

By further defining

$$P(\tau)\equiv Q(\tau,k_1)+Q(\tau,k_2), \tag{4.8}$$

we have for Eq. (4.4), using Eq. (4.5),

$$V_{k_1}^{-1}V_{k_2}^{-1}H(\tau_1)H(\tau_2)\,V_{k_2}V_{k_1}=H(\tau_1)H(\tau_2)+H(\tau_1)P(\tau_2)+P(\tau_1)H(\tau_2)+P(\tau_1)P(\tau_2). \tag{4.9}$$

Since the first term on the right of Eq. (4.9) is quadratic, the second and third term are linear in H, and the last term is independent of H, they will contribute to Eq. (4.3) in different ways. For a given initial state $V_{k_2}V_{k_1}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}][a_{k'}{}^{\dagger n_{k'}}/(n_{k'}!)^{1/2}]\,|\,0\rangle$ the first and fourth term on the right of Eq. (4.9) will couple to different final states. The second and third term of Eq. (4.9) will couple to different final states than either the first or fourth term. In other words, Eq. (4.9) is the second-order counterpart of Eq. (2.12). We discuss the contribution of each term of Eq. (4.9) one by one. For a given initial state $V_{k_2}V_{k_1}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}][a^{\dagger n_{k'}}/(n_{k'}!)^{1/2}]\,|\,0\rangle$ with arbitrary k and k', Eq. (4.3) becomes

$$\begin{aligned}
&\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}+1}}{[(n_{k'}+1)!]^{1/2}}\frac{a_k{}^{n_k-1}}{[(n_k-1)!]^{1/2}}\,V_{k_1}^{-1}V_{k_2}^{-1}U_{(2)}(t)\,V_{k_2}V_{k_1}\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}+1}}{[(n_{k'}+1)!]^{1/2}}\frac{a_k{}^{n_k-1}}{[(n_k-1)!]^{1/2}}\,V_{k_1}^{-1}V_{k_2}^{-1}H(\tau_1)H(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}+1}}{[(n_{k'}+1)!]^{1/2}}\frac{a_k{}^{n_k-1}}{[(n_k-1)!]^{1/2}}\,H(\tau_1)H(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle.
\end{aligned} \tag{4.10}$$

The extreme right of Eq. (4.10) results from Eq. (4.9), since of the four terms on the right of the latter equation, only the term $H(\tau_1)H(\tau_2)$ contributes with final states indicated on the left of Eq. (4.10). This is not surprising, of course, since $H(\tau_1)H(\tau_2)$ is the familiar second-order term in perturbation theory. In fact Eq. (4.10) describes dispersion and Raman scattering. In a similar vein we can describe double photon emission and absorption, respectively, by

$$\begin{aligned}
&\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}+1}}{[(n_{k'}+1)!]^{1/2}}\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,V_{k_1}^{-1}V_{k_2}^{-1}U_{(2)}(t)\,V_{k_2}V_{k_1}\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}+1}}{[(n_{k'}+1)!]^{1/2}}\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,V_{k_1}^{-1}V_{k_2}^{-1}H(\tau_1)H(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}+1}}{[(n_{k'}+1)!]^{1/2}}\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,H(\tau_1)H(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle
\end{aligned} \tag{4.11}$$

and

$$\begin{aligned}
&\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}-1}}{V[(n_{k'}-1)!]^{1/2}}\frac{a_k{}^{n_k-1}}{[(n_k-1)!]^{1/2}}\,V_{k_1}^{-1}V_{k_2}^{-1}H(\tau_1)H(\tau_2)\,V_{k_1}V_{k_2}\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_{k'}{}^{n_{k'}-1}}{[(n_{k'}-1)!]^{1/2}}\frac{a_k{}^{n_k-1}}{[(n_k-1)!]^{1/2}}\,H(\tau_1)H(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{\dagger n_{k'}}}{(n_{k'}!)^{1/2}}\,|\,i,0\rangle.
\end{aligned} \tag{4.12}$$

The conditions under which, after the integrals are done, the processes of dispersion, Raman scattering, double quantum emission, and absorption take place are well known and have been widely discussed in the literature.[2]

[2] M. Goeppert-Mayer, Ann. Physik **9**, 273 (1931).

However, in this paper we wish to draw attention to the properties of the final state of the radiation field as will be discussed in Sec. V. We now consider the terms $H(\tau_1)P(\tau_2)$ and $P(\tau_1)H(\tau_2)$ in Eq. (4.9). In the case of emission, we have

$$\begin{aligned}
&\langle 0,f\,|\,\{a_k{}^{n_k+1}/[(n_k+1)!]^{1/2}\}V_{k_1}{}^{-1}V_{k_2}{}^{-1}U_{(2)}(t)\,V_{k_2}V_{k_1}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,i,0\rangle\\
&\quad=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,V_{k_1}{}^{-1}V_{k_2}{}^{-1}H(\tau_1)P(\tau_2)\,V_{k_2}V_{k_1}\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\\
&\quad\quad+\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,V_{k_1}{}^{-1}V_{k_2}{}^{-1}P(\tau_1)H(\tau_2)\,V_{k_2}V_{k_1}\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,H(\tau_1)P(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\\
&\quad\quad+\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,P(\tau_1)H(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle. \qquad (4.13)
\end{aligned}$$

Consider the term $H(\tau_1)P(\tau_2)$ on the right of the last equation. By inserting the unity operator for the complete set of atomic states $|\,j\rangle$ we have

$$\begin{aligned}
&\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,H(\tau_1)P(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\sum_j\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,H(\tau_1)\,|\,j\rangle\langle j\,|\,P(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\sum_j\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_2}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\exp(i/\hbar H_0\tau_1)H_{IS}\exp(-i/\hbar H_0\tau_1)\,|\,j\rangle\langle j\,|\,P(\tau_2)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\sum_j\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\exp[i(\omega_f-\omega_j)\tau_1+i\omega_k\tau_1]\langle f\,|\,L_k{}^{(-)}(1)\,|\,j\rangle\langle j\,|\,P(\tau_2)\,|\,i\rangle(n_k+1)^{1/2}. \qquad (4.14)
\end{aligned}$$

It will be remembered that, whereas H is both an atomic and field operator, the operator $P(\tau_2)$ is an atomic operator only. This state of affairs has no counterpart in ordinary second-order perturbation theory where one only deals with the form $H(\tau_1)H(\tau_2)$. By considering only the term $Q(\tau_2, k_1)$ in $P(\tau_2)$ we get

$$\begin{aligned}
&\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\langle 0,f\,|\,\frac{a_k{}^{n_k+1}}{[(n_k+1)!]^{1/2}}\,H(\tau_1)Q(\tau_2,k_1)\,\frac{a_k{}^{\dagger n_k}}{(n_k!)^{1/2}}\,|\,i,0\rangle\\
&\quad=\sum_j(1/i\hbar)^2\langle f\,|\,L_k{}^{(-)}(1)\,|\,j\rangle\langle j\,|\,L_k{}^{(-)}(\alpha_{k_1}{}^*)\,|\,i\rangle(n_k+1)^{1/2}\\
&\quad\quad\times\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\exp[i(\omega_f-\omega_j)\tau_1+i\omega_k\tau_1]\exp[i(\omega_j-\omega_i)\tau_2+i\omega_{k_1}\tau_2]\\
&\quad\quad+\sum_j(1/i\hbar)^2\langle f\,|\,L_k{}^{(-)}(1)\,|\,j\rangle\langle j\,|\,L_{k_1}{}^{(+)}(\alpha_{k_1})\,|\,i\rangle(n_k+1)^{1/2}\\
&\quad\quad\times\int_0^t d\tau_1\int_0^{\tau_1}d\tau_2\exp[i(\omega_f-\omega_j)\tau_1+i\omega_k\tau_1]\exp[i(\omega_j-\omega_i)\tau_2-i\omega_{k_1}\tau_2]. \qquad (4.15)
\end{aligned}$$

By doing the integrals in Eq. (4.15), the resonant conditions under which linear variation with time is achieved are

$$\omega_i-\omega_f=\omega_k+\omega_{k_1} \qquad (4.16)$$

and

$$\omega_i-\omega_f=\omega_k-\omega_{k_1}. \qquad (4.17)$$

Equation (4.16) corresponds to the case where the initial atomic state is in the excited state. In the final state, the atomic state is in the ground state. The frequency ω_{k_1} of the coherent wave has to satisfy Eq. (4.16), which is analogous to two-photon emission; Eq. (4.17) corresponds to dispersion for $\omega_i=\omega_f$, and to Raman scattering for

$\omega_i \neq \omega_f$ (Stokes for $\omega_i - \omega_f < 0$, anti-Stokes for $\omega_i - \omega_f > 0$). It is interesting to note that for $n_k = 0$, we have essentially scattering of a coherent wave leading to a final state $\langle 0 \mid a_k V_{k_1}^{-1}$ of the radiation field, with k arbitrary.

In the case of absorption we have

$$\begin{aligned}
&\langle 0, f \mid \{a_k^{n_k-1}/[(n_k-1)!]^{1/2}\} V_{k_1}^{-1} V_{k_2}^{-1} U_{(2)}(t)\, V_{k_2} V_{k_1} [a_k^{\dagger n_k}/(n_k!)^{1/2}] \mid i, 0\rangle \\
&\quad = \left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} V_{k_1}^{-1} V_{k_2}^{-1} H(\tau_1) P(\tau_2) V_{k_2} V_{k_1} \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&\qquad + \left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} V_{k_1}^{-1} V_{k_2}^{-1} P(\tau_1) H(\tau_2) V_{k_2} V_{k_1} \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&\quad = \left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} H(\tau_1) P(\tau_2) \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&\qquad + \left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} P(\tau_1) H(\tau_2) \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle. \qquad (4.18)
\end{aligned}$$

Again, as in the case of emission, if we only consider the term $H(\tau_1)P(\tau_2)$ on the right of Eq. (4.18) and insert the unity operator for the complete set of atomic states $\mid j\rangle$, we have

$$\begin{aligned}
&\left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} H(\tau_1) P(\tau_2) \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&\quad = \sum_j \left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} H(\tau_1) \mid j\rangle\langle j \mid P(\tau_2) \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&\quad = \sum_j \left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{t_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} \exp(i/\hbar H_0 \tau_1) H_{IS} \exp(-i/\hbar H_0 \tau_1) \mid j\rangle\langle j \mid P(\tau_2) \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&\quad = \sum_j \left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \exp[i(\omega_f - \omega_j)\tau_1 - i\omega_k \tau_1] \langle f \mid L_k^{(+)}(1) \mid j\rangle\langle j \mid P(\tau_2) \mid i\rangle (n_k)^{1/2}. \qquad (4.19)
\end{aligned}$$

By considering only the term $Q(\tau_2, k_1)$ in $P(\tau_2)$ we get

$$\begin{aligned}
&\left(\frac{1}{i\hbar}\right)^2 \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle 0, f \mid \frac{a_k^{n_k-1}}{[(n_k-1)!]^{1/2}} H(\tau_1) Q(\tau_2, k_1) \frac{a_k^{\dagger n_k}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&\qquad = \sum_j [(i\hbar)^{-1}]^2 \langle f \mid L_k^{(+)}(1) \mid j\rangle\langle j \mid L_{k_1}^{(-)}(\alpha_{k_1}{}^*) \mid i\rangle (n_k)^{1/2} \\
&\qquad\quad \times \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \exp[i(\omega_f - \omega_j)\tau_1 - i\omega_k \tau_1] \exp[i(\omega_j - \omega_i)\tau_2 + i\omega_{k_1}\tau_2] \\
&\qquad + \sum_j [(i\hbar)^{-1}]^2 \langle f \mid L_k^{(+)}(1) \mid j\rangle\langle j \mid L_{k_1}^{(+)}(\alpha_{k_1}) \mid i\rangle (n_k)^{1/2} \\
&\qquad\quad \times \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \exp[i(\omega_f - \omega_j)\tau_1 - i\omega_k \tau_1] \exp[i(\omega_j - \omega_i)\tau_2 - i\omega_{k_1}\tau_2]. \qquad (4.20)
\end{aligned}$$

By doing the integrals of the last equation, the conditions under which linear variation with time is achieved (the resonant conditions) are

$$\omega_f - \omega_i = \omega_k + \omega_{k_1}, \qquad (4.21)$$

$$\omega_f - \omega_i = \omega_k - \omega_{k_1}. \qquad (4.22)$$

Equation (4.21) corresponds to the case where the initial atomic state is in the ground state. In the final state the atomic state is in the excited state. Equation (4.21) is the analogue of two-photon absorption. Equation (4.22) is the absorption counterpart of dispersion and Raman scattering. If the initial atomic state is the ground state, the final state can be either the ground state (for $\omega_k = \omega_{k_1}$) or the excited state ($\omega_i - \omega_f < 0$). Conversely, if the initial state is the excited state, the final state will be the ground state ($\omega_i - \omega_f = \omega_k - \omega_{k_1} > 0$), corresponding to an absorptive anti-Stokes effect. Similar considerations apply for the term $P(\tau_1)H(\tau_2)$, which will not be carried

out here. We now wish to handle the term $P(\tau_1)P(\tau_2)$ of Eq. (4.9). This operator is only an operator of the atomic system. From Eq. (4.8) we have

$$P(\tau_1)P(\tau_2)=\sum_{i,j;\,i=1,2;\,j=1,2} Q(\tau_i, k_j)Q(\tau_j, k_i). \tag{4.23}$$

Let us choose the term $Q(\tau_1, k_1)Q(\tau_2, k_2)$ out of the four terms in the summation of Eq. (4.23), since no new physics is to be gained by considering the other terms. We then have

$$\begin{aligned}
&\langle 0,f \mid \frac{a_k{}^{nk}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{nk'}}{(n_{k'}!)^{1/2}} V_{k_1}{}^{-1}V_{k_2}{}^{-1}U_{(2)}(t)\,V_{k_2}V_{k_1}\frac{a_{k'}{}^{\dagger nk'}}{(n_{k'}!)^{1/2}}\frac{a_k{}^{\dagger nk}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2\langle 0,f \mid \frac{a_k{}^{nk}}{(n_k!)^{1/2}}\frac{a_{k'}{}^{nk'}}{(n_{k'}!)^{1/2}} V_{k_1}{}^{-1}V_{k_2}{}^{-1}Q(\tau_1, k_1)Q(\tau_2, k_2)\,V_{k_2}V_{k_1}\frac{a_{k'}{}^{\dagger nk'}}{(n_{k'}!)^{1/2}}\frac{a_k{}^{\dagger nk}}{(n_k!)^{1/2}} \mid i, 0\rangle \\
&=\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2\langle f \mid Q(\tau_1, k_1)Q(\tau_2, k_2) \mid i\rangle \\
&=\sum_j\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2\langle f \mid Q(\tau_1, k_1) \mid j\rangle\langle j \mid Q(\tau_2, k_2) \mid i\rangle,
\end{aligned} \tag{4.24}$$

where we have inserted the unity operator for the complete set of atomic states $\mid j\rangle$ in the last expression. We notice that the initial and final state of the field have to be the same in order for the matrix element to be nonvanishing. This is a direct result of the operator Q being an operator on atomic states only, and of the orthogonality of the stationary states. A more explicit evaluation of Eq. (4.24) yields

$$\begin{aligned}
&\sum_j\left(\frac{1}{i\hbar}\right)^2\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2\langle f \mid Q(\tau_1, k_1) \mid j\rangle\langle j \mid Q(\tau_2, k_2) \mid i\rangle \\
&=\sum_j (1/i\hbar)^2\Big(\langle f \mid L_{k_1}{}^{(+)}(\alpha_{k_1}) \mid j\rangle\langle j \mid L_{k_2}{}^{(+)}(\alpha_{k_2}) \mid i\rangle \\
&\times\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2 \exp[i(\omega_f-\omega_j)-i\omega_{k_1}\tau_1]\exp[i(\omega_j-\omega_i)\tau_2-i\omega_{k_2}\tau_2] \\
&+\langle f \mid L_{k_1}{}^{(+)}(\alpha_{k_1}) \mid j\rangle\langle j \mid L_{k_2}{}^{(+)}(\alpha_{k_2}{}^*) \mid i\rangle \\
&\times\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2 \exp[i(\omega_f-\omega_j)\tau_1-i\omega_{k_1}\tau_1]\exp[i(\omega_j-\omega_i)\tau_2+i\omega_{k_2}\tau_2] \\
&+\langle f \mid L_{k_1}{}^{(-)}(\alpha_{k_1}{}^*) \mid j\rangle\langle j \mid L_{k_2}{}^{(+)}(\alpha_{k_2}) \mid i\rangle \\
&\times\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2 \exp[i(\omega_f-\omega_j)\tau_1+i\omega_{k_1}\tau_1]\exp[i(\omega_j-\omega_i)\tau_2-i\omega_{k_2}\tau_2] \\
&+\langle f \mid L_{k_1}{}^{(-)}(\alpha_{k_1}{}^*) \mid j\rangle\langle j \mid L_{k_2}{}^{(-)}(\alpha_{k_2}{}^*) \mid i\rangle \\
&\times\int_0^t d\tau_1\int_0^{\tau_1} d\tau_2 \exp[i(\omega_f-\omega_j)\tau_1+i\omega_{k_1}\tau_1]\exp[i(\omega_j-\omega_i)\tau_2+i\omega_{k_2}\tau_2]\Big).
\end{aligned} \tag{4.25}$$

In order for the integrals of the four terms on the right of Eq. (4.25) to yield quantities varying linearly with time, the following resonant conditions have to be satisfied:

$$\omega_f-\omega_i=\omega_{k_1}+\omega_{k_2}, \tag{4.26}$$

$$\omega_f-\omega_i=-(\omega_{k_1}+\omega_{k_2}), \tag{4.27}$$

$$\omega_f-\omega_i=\omega_{k_1}-\omega_{k_2}, \tag{4.28}$$

$$\omega_f-\omega_i=-(\omega_{k_1}-\omega_{k_2}). \tag{4.29}$$

Equation (4.26) represents "absorption," i.e., the atom goes to a higher energy level in the final state. The sum of the frequencies of the two coherent waves have to be equal to the energy separation of the two atomic states. Similarly, Eq. (4.27) would represent "emission", i.e., the atom goes from an initial energy level to a final lower energy level. A similar situation arises from Eqs. (4.28) and (4.29). Note, however, that in all these transitions, only the atomic system undergoes a change, whereas the state of the field remains un-

changed. This is the second-order counterpart of Eqs. (3.3) and (3.6).

In this section we were able to reproduce all the resonant conditions normally associated with second-order effects. In ordinary perturbation theory in the past, this meant having a stationary state as an initial state. By using the displaced stationary state formalism developed in Sec. II, we were able to obtain, in a natural way, the resonant conditions in the case for which the initial state is a coherent state as given by Eq. (2.2), or more generally of the form $V_{k'}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,0\rangle$ (with k' arbitrary).

V. COHERENCE PROPERTIES OF THE DISPLACED STATIONARY STATES

The fact that we were able to reproduce, in the last two sections, all the resonant conditions normally associated with dispersion, Raman scattering, single and double absorption, and emission, provides only scant evidence that the displaced stationary state formalism (states as given by $V_{k'}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,0\rangle$) represents a good description of the physics that is going on in the interaction of a coherent field (such as is provided by the output of a gas laser, say at 50 times above threshold) and an atomic system. The study of the coherence properties of these states will allow us to make predictions upon the outcome of photon-correlation experiments in which these states are involved. We define, in analogy with Eqs. (2.8a) and (2.8b),

$$\mathbf{A}^{(+)}(\mathbf{r},t)=C\sum_{k_1}(\hbar/2\omega_{k_1}V_0)^{1/2}\mathbf{l}_{k_1}a_{k_1}\times\exp[i(\mathbf{k}_1\cdot\mathbf{r}-\omega_{k_1}t)],\quad(5.1a)$$

$$\mathbf{A}^{(-)}(\mathbf{r},t)=C\sum_{k_1}(\hbar/2\omega_{k_1}V_0)^{1/2}\mathbf{l}_{k_1}a_{k_1}{}^{\dagger}\times\exp[-i(\mathbf{k}_1\cdot\mathbf{r}-\omega_{k_1}t)].\quad(5.1b)$$

The first-order coherence relation in the sense of Glauber,[1] for the state

$$V_{k'}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,0\rangle \qquad (5.2)$$

is given by

$$\begin{aligned}&\langle 0\,|\,[a_k{}^{n_k}/(n_k!)^{1/2}]V_{k'}{}^{-1}A^{(-)}A^{(+)}V_{k'}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,0\rangle\\&=\langle 0\,|\,[a_k{}^{n_k}/(n_k!)^{1/2}]V_{k'}{}^{-1}A^{(-)}V_{k'}\{a_k{}^{\dagger n_k-1}/[(n_k-1)!]^{1/2}\}\,|\,0\rangle\\&\times\langle 0\,|\,\{a_k{}^{n_k-1}/[(n_k-1)!]^{1/2}\}V_{k'}{}^{-1}A^{(+)}V_{k'}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,0\rangle\\&+\langle 0\,|\,[a_k{}^{n_k}/(n_k!)^{1/2}]V_{k'}{}^{-1}A^{(-)}V_{k'}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,0\rangle\\&\times\langle 0\,|\,[a_k{}^{n_k}/(n_k!)^{1/2}]V_{k'}{}^{-1}A^{(+)}V_{k'}[a_k{}^{\dagger n_k}/(n_k!)^{1/2}]\,|\,0\rangle.\end{aligned}\qquad(5.3)$$

Equation (5.3) holds for both $k\neq k'$ and $k=k'$. The fact that the terms on the right of Eq. (5.3) are both different from zero for $n_k\neq 0$ indicates that the left side does not factorize for $n_k\neq 0$. Thus the state given by Eq. (5.2) does not possess first-order coherence for $n_k\neq 0$. Similar considerations indicate that this is also true for higher-order coherence. However, if $n_k=0$, the first term on the right vanishes, and the state (5.2) is of course coherent to all orders. This has an interesting physical consequence. Consider the state $V_k a_k{}^{\dagger}\,|\,0\rangle$ for instance, which can be considered spontaneous emission in the forward direction, or Rayleigh scattering in the forward direction for an incident field $V_k\,|\,0\rangle=|\,\alpha_k\rangle$. If such a state $V_k a_k{}^{\dagger}\,|\,0\rangle$ is made to interact with an atomic system, we know from the last two sections that there is a probability for the state $V_k\,|\,0\rangle=|\,\alpha_k\rangle$ being a final state of the field under a proper choice of an initial state of the atomic system. However, this last state is just a coherent state to all orders. It will thus exhibit no photon correlations in an appropriate photon-counting experiment.

The state $V_k a_k{}^{\dagger}\,|\,0\rangle$, on the other hand, will exhibit such correlations. Hence, it would appear, that it ought to be possible to determine experimentally whether the state can indeed make a transition to a coherent state in, say, an absorption process. If this were seen to be so it would indicate that the state $V_k a_k{}^{\dagger}\,|\,0\rangle$ exists and that the displaced stationary state formalism is indeed a good description of the physics prevailing in the interaction of a coherent wave and an atomic system.

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