

Pure-state analysis of resonant light scattering: Radiative damping, saturation, and multiphoton effects

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A fully quantum-mechanical treatment of resonant light scattering is presented. The incident field is assumed to be described by a coherent state, and is allowed to be intense enough to cause saturation. Complete solutions are obtained for the correlated atom-field pure state vector, including multiphoton contributions of arbitrary order. The frequency spectrum of the scattered field is evaluated and is found to agree exactly with the result previously obtained by means of the quantum fluctuation-regression theorem. A derivation of the fluctuation-regression theorem and of the optical Bloch equations is given which is fully quantum mechanical and which relies upon no assumption of statistical factorization of atom and field states. The accuracy of the result found for the scattered-field spectrum is thus shown to be limited only by the assumption of the smallness of the saturated linewidth compared to the (optical) atomic resonance frequency. The one-photon approximation is analyzed in some detail. The method of adding an imaginary term to the upper-atomic-state energy is clarified, and it is shown how the vacuum and one-photon amplitudes thereby obtained may be used, within a simple and plausible iteration scheme, to construct the complete multiphoton spectrum. A variety of commonly used injection schemes and methods of representing atomic relaxation are discussed, and comparisons are made with results found by other authors. The entire analysis is performed with the aid of a canonical transformation which replaces the applied field by a c number. It is thus proved quite rigorously and generally that the use of a c -number applied field is a fully quantum-mechanical procedure, provided only that radiation-reaction terms are retained.

I. INTRODUCTION

The resonant interaction between the electromagnetic field and an atom containing a single pair of participating levels has been discussed extensively for the case in which the field initially consists of a coherent, possibly intense wave incident upon the atom.^{1-10a} Of particular interest are saturation effects, i.e., the effects of the incident field intensity upon the atomic populations, linewidths, and (possibly) radiative frequency shifts, and, more generally, upon the spectral properties of the field radiated (i.e., scattered) by the atom.^{10b,11}

In lowest order in the incident field intensity, the process under discussion is simply the familiar one of elastic one-photon light scattering. Indeed, what one is dealing with is essentially a scattering process even when the incident field becomes quite intense, when multiphoton effects become important. The process is nevertheless rarely discussed in scattering-theory language, but is instead described by a variety of approximation schemes. Notable among these are those in which the incident field is described classically, and Bloch-type equations of motion are obtained for the atomic density operator by methods borrowed from statistical mechanics, i.e., by treating the radiation field as a (zero-temperature) heat bath, coupled to the atom in the Markoff approximation.⁴ The quantum *fluctuation-regression theorem*¹² may

then be used, in the approximation scheme under discussion, to evaluate the two-time atomic correlation function which determines the frequency spectrum of the scattered field.⁵ The method just described, though it in fact leads to accurate results (as is shown in this paper), may well be criticized for its reliance upon the classical description of the incident field and upon the assumption of statistical factorization of atomic and field states inherent in the Markoff approximation. Moreover, the individual multiphoton scattering processes which are responsible for the intensity-dependent effects in question are concealed from view, with only their accumulated effect exhibited. (This drawback is of some practical importance when multiple-atom interference effects occur, since such effects are not easily discussed in the Markoff approximation.)

For these reasons, it is desirable to discuss the scattering process in a direct fashion, i.e., by evaluating the state vector for the joint quantum-mechanical system of atom and electromagnetic field, as a function of time. The difficulty in doing this is due principally to two factors: (a) the difficulty in adequately describing the radiative damping process when many photons are involved, and (b) the need to include the effect of forward-scattering processes when the incident field becomes intense.

Both of these difficulties are overcome by the analysis of the present paper, which begins by

eliminating the photons in the incident field (which is assumed initially to be represented by a *coherent state*¹³) by performing a canonical transformation which allows the incident field to be represented by a *c*-number function. This transformation involves no approximation whatever, and, in particular, in no way modifies the fully quantum-mechanical nature of the analysis. The radiative damping process is then described quite carefully by evaluating in a well-defined set of approximations the term in the interaction Hamiltonian which represents the reabsorption of photons previously emitted by the atom. In the forward-scattering terms, the field is represented by a *c*-number function, as a consequence of the canonical transformation mentioned above. The inclusion of the terms in question, though indeed complicating the analysis, then presents no difficulty of a fundamental kind. Complete and accurate solutions are obtained for the joint time-dependent atom-field state vector, in the form of an infinite-series expansion consisting of terms of successively higher numbers of scattered photons.

In the limit of weak incident fields, where forward-scattering terms can be neglected, the solution which corresponds to a monochromatic incident field has the stationary character normally associated with scattering-theory solutions. The two-photon part is examined in detail, and is shown to represent corrections to the lowest-order (one-photon) expressions for the scattered-field spectrum and pair-correlation function which are identical to those which are found by the Markoff approximation.

When forward-scattering contributions are included, as they must be when the incident-field intensity becomes appreciable, the simple stationary character of the scattered-field state vector (though not of the correlation functions which describe the field at specified spatial locations) is lost. This is due simply to the fact that the inclusion of the terms in question restores the unitarity which is violated in their absence, and thus leads, as the number of scattered photons increases, to the time-dependent decay to zero of the amplitude for finding any fixed number of photons. The general time-dependent solution in this case is found, and the zero-photon and one-photon parts of it are examined in detail, and shown in particular to have the expected decay properties.

The frequency spectrum of the scattered field, including multiphoton contributions of arbitrary order, is evaluated, and is found to agree exactly with results obtained previously by Markoff methods.⁵ This agreement is clarified by a rigorous derivation of the optical Bloch equations and of the quantum fluctuation-regression theorem from the

equations governing the time evolution of the pure state vector for the correlated atom-field system. The derivation in question relies upon no statistical factorization assumption, and is fully quantum mechanical. Its accuracy is limited only by the assumption of the smallness of the saturated linewidth compared to the (optical) resonance frequency.

The rigorously derived multiphoton spectrum is shown to be obtainable from an analysis based on a simple picture, involving the use of vacuum and one-photon amplitudes alone. The picture in question is suitably modified so as to describe the case of collisional relaxation, which is carefully distinguished from the radiative case. A variety of approximation schemes and the analyses of other authors are discussed.

In Sec. II the canonical transformation which allows the incident field to be represented by a *c*-number function is performed. Section III contains the solution for the photon-absorption term in the Hamiltonian, which is then used to formulate an accurate pure-state damping theory. Solutions for the scattered-field state vector are then obtained in Sec. IV, while Sec. V contains an evaluation of the scattered-field frequency spectrum. Rigorous derivations of the optical Bloch equations and of the regression theorem are given in Sec. VI. Specific *n*-photon contributions are evaluated in Sec. VII, and in Secs. VIII and IX the multiphoton spectrum is constructed from one-photon amplitudes in the radiative case and in the collisional case, respectively. In Sec. X, finally, it is shown how the analysis of the preceding sections, which describes the case of a two-level atom, may be generalized so as to describe an atom with many levels.

II. CANONICAL TRANSFORMATION FOR INITIALLY COHERENT FIELD STATE

Let us consider an atom situated at the origin of coordinates and coupled to the electromagnetic field in the electric-dipole approximation. The Hamiltonian for the system of atom and field is then

$$\hat{H} = H_{0a} + H_{0F} - \vec{\mu} \cdot \vec{E}_R(\vec{r}=0), \quad (2.1a)$$

where $\vec{\mu}$ is the atomic electric-dipole operator, and $\vec{E}_R(\vec{r})$ is the quantum-mechanical electric-field operator, with positive- and negative-frequency parts $\vec{\mathcal{E}}_R^+(\vec{r})$ and $\vec{\mathcal{E}}_R^-(\vec{r})$, respectively. It is convenient to adopt the discrete-field-mode expansion

$$\vec{\mathcal{E}}_R^-(\vec{r}) = i \sum_k \vec{u}_k(\vec{r}) b_k \quad (2.1b)$$

$$\vec{u}_k(\vec{r}) = (\hbar \omega_k / 2V)^{1/2} \hat{e}_k e^{i\vec{k} \cdot \vec{r}},$$

where b_k is the photon-annihilation operator for the k th mode, \hat{e}_k is the associated unit polarization vector, and V is the quantization volume. The free Hamiltonian for the field is then

$$H_{\text{of}} = \sum_k \hbar \omega_k b_k^\dagger b_k, \quad (2.1c)$$

while the free atomic Hamiltonian may be left unspecified.

We wish to choose as an initial state for the field that state which resembles as closely as possible a prescribed classical (c -number) function. A freely propagating classical electric field $\vec{E}_c(\vec{r}, t) = \vec{\mathcal{E}}_c(\vec{r}, t) + \vec{\mathcal{E}}_c^*(\vec{r}, t)$ has a positive-frequency part with the modal expansion

$$\vec{\mathcal{E}}_c(\vec{r}, t) = i \sum_k \vec{u}_k(\vec{r}) \beta_k e^{-i\omega_k t}, \quad (2.2)$$

in which the parameters β_k are complex constants. The field will be assumed to vanish at the position of the atom in the infinite past,

$$\lim_{t \rightarrow -\infty} \vec{E}_c(0, t) = 0. \quad (2.3)$$

We choose the initial field state to be the *coherent state*¹³ which is the eigenstate of b_k with eigenvalue $\beta_k e^{-i\omega_k t_0}$ (for all k) at an initial time t_0 . More precisely, we make the limiting statement that the (Schrödinger) state vector $|\hat{f}\rangle$ for the system of atom and field has the limiting form

$$|\hat{f}\rangle \xrightarrow{t \rightarrow -\infty} D(\{\beta_k e^{-i\omega_k t}\}) |0\rangle_F |0\rangle_a, \quad (2.4)$$

where $|0\rangle_a$ is the ground state of the atom (which is taken to have zero energy), $|0\rangle_F$ is the vacuum state of the field, and D is the unitary *displacement operator*¹³

$$D(\{\beta_k\}) = \exp\left(\sum_k (b_k^\dagger \beta_k - \beta_k^* b_k)\right). \quad (2.5)$$

The aforementioned eigenvalue property of the state represented in Eq. (2.4) follows from the displacement property

$$D^{-1}(\{\beta_k\}) b_k D(\{\beta_k\}) = b_k + \beta_k \quad (2.6)$$

and the relation $b_k |0\rangle_F = 0$. That the relation (2.4) makes sense as a limiting statement follows from the assumption in Eq. (2.3), which asserts that the atom and field are decoupled in the infinite past, and from the fact that an initially coherent state of the system of field oscillators will retain its coherent character as long as it oscillates freely.¹⁴

The solving of Schrödinger's equation

$$i\hbar \frac{d}{dt} |\hat{f}\rangle = \hat{H} |\hat{f}\rangle \quad (2.7)$$

corresponding to the initial condition (2.4), is

facilitated by introducing the time-dependent canonical transformation

$$|\hat{f}\rangle \equiv D(\{\beta_k e^{-i\omega_k t}\}) |t\rangle. \quad (2.8)$$

It follows directly from Eqs. (2.7), (2.8), (2.1), (2.6), and the identity

$$\begin{aligned} \frac{d}{dt} D(\{\beta_k(t)\}) = D(\{\beta_k(t)\}) \sum_k \{ [b_k^\dagger + \frac{1}{2}\beta_k^*(t)] \dot{\beta}_k(t) \\ - \dot{\beta}_k^*(t) [b_k + \frac{1}{2}\beta_k(t)] \}, \end{aligned} \quad (2.9)$$

that the state $|t\rangle$ obeys the time-development equation

$$i\hbar \frac{d}{dt} |t\rangle = H(t) |t\rangle, \quad (2.10)$$

where

$$H(t) = H_{\text{oa}} + H_{\text{of}} - \vec{\mu} \cdot [\vec{E}_R(0) + \vec{E}_c(0, t)]. \quad (2.11)$$

The initial condition for the state $|t\rangle$, according to Eqs. (2.4) and (2.8), is simply

$$|t\rangle \xrightarrow{t \rightarrow -\infty} |0\rangle_a |0\rangle_F. \quad (2.12)$$

Thus the state $|\hat{f}\rangle$ of the quantum-mechanical system of atom and field, corresponding to an initial field state which is (a) coherent and (b) decoupled from the atom, can be found by adding a c -number function to the field operator and then solving for the time evolution of the state $|t\rangle$ which initially contains no photons. The untransformed state vector $|\hat{f}\rangle$ can then be found from Eq. (2.8). Alternatively, one may define Heisenberg operators $\vec{E}_R(\vec{r}, t)$ in the usual way in terms of the Hamiltonian defined by Eq. (2.11) (with the initial time in the infinite past), and then evaluate multi-time field-correlation functions as the expectation value of products of the operators

$$\vec{E}(\vec{r}, t) = \vec{E}_R(\vec{r}, t) + \vec{E}_c(\vec{r}, t) \quad (2.13)$$

in the state $|0\rangle_a |0\rangle_F$.

It should be apparent from the foregoing discussion that the use of a c -number applied field, along with the boundary condition (2.12) in scattering problems, involves no approximation whatever. In particular, the validity of the method is not dependent upon any assumption concerning the intensity of the incident field or the number of quanta in it.

III. PURE-STATE DAMPING THEORY IN RESONANT APPROXIMATION

As an example in which solutions can be obtained in closed form and with high accuracy, let us consider an atom consisting of just two levels, a ground state $|0\rangle_a$ with zero energy, and a single excited state $|1\rangle_a$. It will be assumed that the in-

cident field oscillates at a frequency near the resonant-transition frequency $E_1/\hbar \equiv \omega_{10}$, and the Hamiltonian

$$H(t) = H_0 + H_1(t) \quad (3.1)$$

in Eq. (2.11) will be evaluated in the resonant approximation, i.e., with its interaction part approximated as

$$H_1(t) = -\hbar \bar{\lambda}^* \cdot [\bar{\mathcal{E}}_R^\dagger(0) + \bar{\mathcal{E}}_c^*(0, t)] a \\ - a^\dagger \hbar \bar{\lambda} \cdot [\bar{\mathcal{E}}_R(0) + \bar{\mathcal{E}}_c(0, t)], \quad (3.2a)$$

where $\hbar \bar{\lambda} \equiv \langle 1 | \vec{\mu} | 0 \rangle_a$, $a \equiv |0\rangle_a \langle 1|$, and $a^\dagger \equiv |1\rangle_a \langle 0|$. The free Hamiltonian is

$$H_0 = \hbar \omega_{10} a^\dagger a + H_{0F}, \quad (3.2b)$$

with H_{0F} given by Eq. (2.1c).

The interaction-picture state vector

$$|t\rangle \equiv e^{iH_0 t/\hbar} |t\rangle \quad (3.3)$$

obeys the time-development equation

$$i\hbar \frac{d}{dt} |t\rangle = H_1'(t) |t\rangle, \quad (3.4)$$

where

$$H_1'(t) = -\hbar \bar{\lambda}^* \cdot [\bar{\mathcal{E}}_R^\dagger(0, t) + \bar{\mathcal{E}}_c^*(0, t)] a'(t) \\ - a'^\dagger(t) \hbar \bar{\lambda} \cdot [\bar{\mathcal{E}}_R(0, t) + \bar{\mathcal{E}}_c(0, t)], \quad (3.5)$$

$$a'(t) \equiv a e^{-i\omega_{10} t}, \quad (3.6)$$

and

$$\bar{\mathcal{E}}_R'(\vec{r}, t) = i \sum_k \vec{u}_k(\vec{r}) b_k e^{-i\omega_k t}. \quad (3.7)$$

The solution to Eq. (3.4) may be developed as a power series in the coupling constant λ , with the n th term obtained from the preceding term through the recursion relation

$$|t\rangle_n = (i\hbar)^{-1} \int_{-\infty}^t dt' H_1'(t') |t'\rangle_{n-1} \quad (3.8)$$

for $n \geq 1$, while the zeroth term, corresponding to the initial condition (2.12), is just $|0\rangle_a |0\rangle_F$.

The presence of the term involving the photon-absorption operator \mathcal{E}_R in H_1 greatly complicates

the form of the solutions which follow directly from Eqs. (3.3)–(3.7), and in particular makes it difficult to isolate from them the terms representing specified numbers of photons. Inasmuch as the atom can absorb only those photons which it has previously emitted (excluding, of course, the incident photons, which are now accurately represented by the c -number function E_c), it becomes possible to solve for the photon-absorption term, thus eliminating it from the problem.

As a means of accomplishing this purpose, it is convenient to begin by introducing the functions

$$\phi(t-t') \equiv [\bar{\mathcal{E}}_R'(0, t), \bar{\mathcal{E}}_R'^\dagger(0, t')] \\ = \int d\omega_k G(\omega_k) e^{i\omega_k(t-t')} \quad (3.9)$$

and

$$F(\bar{t}-t) \equiv \int_{-\infty}^{\bar{t}} dt' e^{i\omega_{10}(\bar{t}-t')} \phi(\bar{t}-t'), \quad (3.10)$$

where

$$G(\omega_k) = \hbar \omega_k^3 / 6\pi^2 c^3. \quad (3.11)$$

The value of the function $G(\omega_k)$ as given by Eq. (3.11) follows in the limit $V \rightarrow \infty$ directly from Eq. (2.1b).

The function $\phi(\tau)$ is highly singular in the form shown, and must be understood to be regularized, by the introduction of a cutoff, for example, into the definition of $G(\omega_k)$. The functions $\phi(\tau)$ and $F(\tau)$ for $\tau \geq 0$ are both nonzero only within very small time intervals near $\tau = 0$, comparable in magnitude to the period of oscillation ω_{10}^{-1} . At $\tau = 0$, the function F has the value

$$F(0) = (\frac{1}{2}\kappa + i\delta\omega) / |\lambda|^2 - \frac{1}{2}\kappa / |\lambda|^2, \quad (3.12)$$

where κ is the Einstein A -coefficient

$$\kappa = \hbar |\lambda|^2 \omega_{10}^3 / 3\pi c^3. \quad (3.13)$$

The frequency shift $\delta\omega$ will be assumed to have been incorporated into the definition of ω_{10} in the analysis which follows.

By multiplying both sides of Eq. (3.8) by $\bar{\mathcal{E}}_R'(0, \bar{t})$ (where $\bar{t} \geq t$), one finds, with the aid of Eqs. (3.5) and (3.9), the relation

$$\bar{\mathcal{E}}_R'(0, \bar{t}) |t\rangle_n = i\bar{\lambda}^* \int_{-\infty}^{\bar{t}} dt' \phi(\bar{t}-t') a'(t') |t'\rangle_{n-1} + (i\hbar)^{-1} \int_{-\infty}^{\bar{t}} dt' H_1'(t') \bar{\mathcal{E}}_R'(0, \bar{t}) |t'\rangle_{n-1}, \quad \text{for } \bar{t} \geq t. \quad (3.14)$$

Inasmuch as the function $\phi(\bar{t}-t')$ is appreciable only for $t' \approx \bar{t}$, the first term on the right side of Eq. (3.14) (which is thus appreciable only for $t \approx \bar{t}$) may be evaluated by approximating the slowly varying, interaction-picture state vector $|t'\rangle$ by its value at $t' = t$. When this is done and use is made of Eq. (3.6) and the definition in Eq. (3.10), one finds the relation

$$i\bar{\lambda}^* \int_{-\infty}^{\bar{t}} dt' \phi(\bar{t}-t') a'(t') |t'\rangle_{n-1} = i\bar{\lambda}^* F(\bar{t}-t) a'(\bar{t}) |t\rangle_{n-1}, \quad \text{for } \bar{t} \geq t. \quad (3.15)$$

It is not difficult to show that the second term on the right side of Eq. (3.14) is inappreciable in magnitude,

$$\int_{-\infty}^{\bar{t}} dt' H_1'(t') \bar{\mathcal{E}}_R'(0, \bar{t}) |t'\rangle'_{n-1} \approx 0 \quad \text{for } \bar{t} \geq t, \quad (3.16)$$

and hence that Eq. (3.14) is well approximated as

$$\bar{\mathcal{E}}_R'(0, \bar{t}) |t\rangle'_n = i\bar{\lambda}^* F(\bar{t}-t) a'(\bar{t}) |t\rangle'_{n-1} \quad \text{for } \bar{t} \geq t. \quad (3.17)$$

To justify Eq. (3.16), and thus Eq. (3.17), one may proceed by mathematical induction, first noting that the relations in question are satisfied identically for $n=1$ [note that on the right side of Eq. (3.17), $a|0\rangle_a = 0$]. If one then makes use of Eq. (3.17), as evaluated for $n \rightarrow n-1$, in Eq. (3.16), one finds by making use of the rapid falloff of the function $F(\bar{t}-t')$ and the slow variation both of $|t'\rangle'$ and of the resonant coupling $H_1'(t')$, that the left side of Eq. (3.16) is well approximated as

$$i\bar{\lambda}^* H_1'(t) \left(\int_{-\infty}^{\bar{t}} dt' F(\bar{t}-t') \right) a'(\bar{t}) |t\rangle'_{n-2} \approx 0, \quad \text{for } \bar{t} \geq t. \quad (3.18)$$

That the integral in this expression is indeed inappreciable is guaranteed by the fact that the function $F(\tau)$, for $\tau \geq 0$, (a) is vanishingly small unless $\tau \lesssim \omega_{10}^{-1}$, and (b) remains finite even as $\tau \rightarrow 0$, where it has the value $\frac{1}{2} \kappa / \lambda^2$. The expression evaluated in Eq. (3.18), and hence in (3.16) is thus of order κ / ω_{10} , and must be taken as vanishingly small within the resonant approximation. [As a means of understanding the contrasting treatments accorded to the functions $\phi(\tau)$ and $F(\tau)$ in the integrals in Eqs. (3.15) and (3.18), respectively, it may be useful to think of $\phi(\tau)$ as approximating a δ function, and of $F(\tau)$ as approximating a step function.]

The validity of the approximations in Eqs. (3.15) and (3.18) depends upon the assumed slowness of the time variation of the interaction-picture state vector $|t\rangle'$, compared to that of the rapidly oscillating atomic dipole moment and of the (optical) incident field. Denoting by Ω' the frequency of incident-field-induced variations in $|t\rangle'$ [see Eq. (4.36g)], one may summarize the conditions necessary to justify Eq. (3.17) with the statement

$$\kappa, |\delta\omega|, \Omega' \ll \omega_{10}. \quad (3.19)$$

By summing Eq. (3.17) over n (the left side vanishes for $n=0$), one finds the relation

$$\bar{\mathcal{E}}_R'(0, \bar{t}) |t\rangle' = i\bar{\lambda}^* F(\bar{t}-t) a'(\bar{t}) |t\rangle' \quad \text{for } \bar{t} \geq t. \quad (3.20)$$

At $\bar{t}=t$ one may therefore write, according to Eq. (3.12),

$$\bar{\lambda} \cdot \bar{\mathcal{E}}_R(0) |t\rangle = \frac{1}{2} i \kappa a |t\rangle. \quad (3.21)$$

This relation (which is written in the Schrödinger picture) is the sought-for solution for the term which represents the absorption of photons which have previously been emitted by the atom. It follows from Eqs. (2.10), (3.1), and (3.2) that the state vector $|t\rangle$ obeys to a good approximation the modified time-development equation

$$i\hbar \frac{d}{dt} |t\rangle = \tilde{H}(t) |t\rangle, \quad (3.22)$$

where $\tilde{H}(t)$ is the non-Hermitian operator

$$\begin{aligned} \tilde{H}(t) &= \tilde{H}_0 + \tilde{H}_1(t), \\ \tilde{H}_0 &= \hbar(\omega_{10} - \frac{1}{2} i \kappa) a^\dagger a + H_{0F}, \\ \tilde{H}_1(t) &= -\hbar \bar{\lambda}^* \cdot [\bar{\mathcal{E}}_R^\dagger(0) + \bar{\mathcal{E}}_c^*(0, t)] a - a^\dagger \hbar \bar{\lambda} \cdot \bar{\mathcal{E}}_c(0, t). \end{aligned} \quad (3.23)$$

Thus one may omit the photon-absorption term from the interaction Hamiltonian if one adds at the same time the imaginary term $-\frac{1}{2} i \kappa$ to the energy of the upper atomic state. This procedure is valid only when the initial condition (2.12) is satisfied, and of course would not be applicable if photons other than those eliminated by the canonical transformation of Eq. (2.8) were incident upon the atom.

The relation expressed by Eq. (3.21) is important not only in establishing the validity of Eqs. (3.22) and (3.23), but also in evaluating many of the expressions derived from them. The preservation of the norm of the state vector, for example, follows from the relation

$$\begin{aligned} i \frac{d}{dt} \langle t|t\rangle &= \hbar^{-1} \langle t| [\tilde{H}(t) - \tilde{H}^\dagger(t)] |t\rangle \\ &= \langle t| [-i \kappa a^\dagger a - \bar{\lambda}^* \cdot \bar{\mathcal{E}}_R^\dagger(0) a + a^\dagger \bar{\lambda} \cdot \bar{\mathcal{E}}_R(0)] |t\rangle \\ &= 0, \end{aligned} \quad (3.24)$$

in which the last step depends upon Eq. (3.21) and its conjugate.

As the results in Eqs. (3.21)–(3.23) have been established with some care, it may be worthwhile to discuss more fully their range of validity. Within the context of the two-level model, only the previously mentioned conditions (3.19), reflecting the assumed rapidity of the free atomic oscillations, need be imposed. (More precisely, the results are rigorously valid in the limit in which the frequency ratios κ / ω_{10} , $\delta\omega / \omega_{10}$, and Ω' / ω_{10} , which may be thought of as free parameters, are allowed to approach zero.) The conditions in question are exactly the same as those required to justify the resonant approximation [Eq. (3.2a)], and thus impose no further restriction on the validity of the results. It should be emphasized that the applied field E_c may be allowed to be strong enough to cause appreciable saturation without affecting the accuracy of the method, which thus

remains valid for all times t , and, in particular, even after many quanta have been scattered by the atom. Equations (3.22) and (3.23) apply not only in the limiting case described by lowest-order scattering theory (where they are normally used), but equally well when arbitrarily large numbers of field quanta, interfering coherently with one another, are radiated by the strongly driven atom.

The continued creation of new photons is of course an integral feature of the scattering process, and cannot be ignored, except in the limit of weak incident fields, without doing violence to the theory. In particular, whenever the atom is allowed to be raised to its excited state $|1\rangle_a$, it must also be allowed to emit a new photon, in addition to those emitted previously, simply because the emission and reabsorption of the new photon are required to maintain the correct (and simple) form of the expressions which describe radiative damping and radiative level shifts. No dependence of the level shifts upon the intensity of the incident field, it may be noted, is present¹⁵ in the results obtained above, which include multiphoton effects to all orders: The parameter $\delta\omega$ in Eq. (3.12) is independent of E_c .

The use of the two-level model under discussion is justified when neither induced nor spontaneous transitions to other states of the atom take place. The exclusion of induced transitions requires that the condition

$$\vec{\mu}_{jk} \cdot \vec{E}_c \ll \hbar\omega_{jk} \equiv E_j - E_k \quad (3.25)$$

be imposed, while the exclusion of real spontaneous transitions is certainly valid if $|0\rangle_a$ is the ground state of the atom and if the state $|1\rangle_a$ decays spontaneously only to the state $|0\rangle_a$. (Spontaneous transitions in a multilevel atom are treated in Sec. X.)

Virtual spontaneous transitions to other atomic states, however, can in no case be ruled out; in fact, they play an important role (along with previously dropped counter-resonant terms) in determining the radiative level shifts. The processes in question all occur, however, on a time scale determined by the period of free atomic oscillations, and hence so rapidly that the contributions they make can be evaluated in the usual way (i.e., quite without reference to the presence of the applied field), as long as the conditions (3.19) and (3.25) are obeyed. (Again it is crucial that the basic emission-absorption process be allowed to operate, no matter how many photons have previously been emitted.¹⁶) It will accordingly be understood in what follows that the frequency shift which has been incorporated into the definition of ω_{10} has been calculated in the usual way, i.e., with

the effects of all the atomic states included, but with intensity-dependent effects omitted as insignificant corrections.

IV. SOLUTIONS FOR THE STATE OF THE SCATTERED FIELD

A. Weak incident fields

The state vector $|t\rangle$ for the system of two-level atom and quantum-mechanical radiation field may be expressed at any time as

$$|t\rangle = |0\rangle_a |t\rangle_{0F} + |1\rangle_a |t\rangle_{1F}, \quad (4.1)$$

where $|t\rangle_{0F}$ and $|t\rangle_{1F}$ are vectors in the state space of the radiation field. It follows from Eqs. (3.22) and (3.23) that these quantities obey the coupled equations of motion

$$\begin{aligned} \left(i \frac{d}{dt} - \omega_{10} + \frac{i\kappa}{2} - \frac{H_{0F}}{\hbar} \right) |t\rangle_{1F} &= -\vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) |t\rangle_{0F}, \\ \left(i \frac{d}{dt} - \frac{H_{0F}}{\hbar} \right) |t\rangle_{0F} &= -\vec{\lambda}^* \cdot [\vec{\mathcal{E}}_R^\dagger(0) + \vec{\mathcal{E}}_c^*(0, t)] |t\rangle_{1F}. \end{aligned} \quad (4.2)$$

Equations (4.2) are greatly simplified in the limit of weak incident fields E_c , where it becomes possible to eliminate as a small correction the contribution of forward scattering,

$$\vec{\mathcal{E}}_c^* \rightarrow 0, \quad (4.3)$$

and to use Eqs. (4.2) directly to obtain an expansion of the state of the system in photon-number eigenstates.

For the case of a harmonically oscillating incident field,

$$\vec{\mathcal{E}}_c(0, t) = \vec{\mathcal{E}}_0 e^{-i\omega t} \quad (4.4)$$

(where $\omega \approx \omega_{10}$), the state vector $|t\rangle$ then has a solution of the form

$$|t\rangle = e^{-i(a^\dagger + N)\omega t} | \rangle, \quad (4.5a)$$

where N is the total number of photons in the scattered field,

$$N \equiv \sum_k b_k^\dagger b_k, \quad (4.5b)$$

and $| \rangle$ is a constant vector. [The field frequency ω must be taken to have a small positive imaginary part, in accordance with the initial condition (2.12).]

One readily finds, by making use of Eqs. (4.3), (4.4), and (2.12) in Eqs. (4.2), that in the stationary state described by Eqs. (4.5), the vectors $|t\rangle_{0F}$ and $|t\rangle_{1F}$ have the values

$$|t\rangle_{0F} = e^{-iN\omega t} \frac{1}{1-A} |0\rangle_F, \quad (4.6a)$$

where

$$A = (1/\mathcal{C}_F) \bar{\lambda}^* \cdot \bar{\mathcal{E}}_R^\dagger(0) (\bar{\lambda} \cdot \bar{\mathcal{E}}_0) (\mathcal{C}_F - \delta)^{-1} \quad (4.6b)$$

and

$$|t\rangle_{1F} = e^{-i\omega t} \bar{\lambda} \cdot \bar{\mathcal{E}}_0 (\mathcal{C}_F - \delta)^{-1} |t\rangle_{0F}, \quad (4.6c)$$

where

$$\mathcal{C}_F \equiv \sum_k (\omega_k - \omega) b_k^\dagger b_k, \quad (4.7a)$$

$$\delta \equiv \Delta\omega + \frac{1}{2}i\kappa, \quad (4.7b)$$

$$\Delta\omega \equiv \omega - \omega_{10}, \quad (4.7c)$$

and $|0\rangle_F$ is the vacuum state of the field.

By directly expanding the operator fraction in Eq. (4.6a), one finds a photon-number series of the form

$$|t\rangle_{0F} = |0\rangle_F + e^{-i\omega t} \sum_k \bar{\beta}_k b_k^\dagger |0\rangle_F + \frac{1}{2} e^{-2i\omega t} \sum_{k_1, k_2} \bar{\gamma}_{k_1 k_2} b_{k_2}^\dagger b_{k_1}^\dagger |0\rangle_F + \dots \quad (4.8)$$

for the state which describes the field when the atom is in its ground state. [The field state $|t\rangle_{1F}$, according to Eq. (4.6c), is vanishingly small compared to $|t\rangle_{0F}$ in the limit under discussion ($\mathcal{E}_0 \rightarrow 0$).] The one- and two-photon wave functions in Eq. (4.8) have the values

$$\bar{\beta}_k = \frac{i\bar{\lambda}^* \cdot \bar{\mathbf{u}}_k^*(0) \bar{\lambda} \cdot \bar{\mathcal{E}}_0 / \delta}{\omega_k - \omega - i\epsilon} \quad (4.9)$$

and

$$\bar{\gamma}_{k_1 k_2} = \bar{\beta}_{k_1} \bar{\beta}_{k_2} + \frac{(\bar{\lambda} \cdot \bar{\mathcal{E}}_0)^2}{\delta^2} \frac{\bar{\lambda}^* \cdot \bar{\mathbf{u}}_{k_1}^*(0) \bar{\lambda}^* \cdot \bar{\mathbf{u}}_{k_2}^*(0)}{\omega_{k_1} + \omega_{k_2} - 2\omega - i\epsilon} \times \left(\frac{1}{\omega_{k_1} - \omega - \delta} + \frac{1}{\omega_{k_2} - \omega - \delta} \right). \quad (4.10)$$

The expression found here for the one-photon part of the state vector for the scattered field is identical to the one found by the familiar methods of lowest-order perturbation theory, and describes the elastic scattering of photons at the energy $\hbar\omega$ of an incident-field photon.

The two-photon part of the scattered-field state vector as given by Eq. (4.10) is the sum of two separate terms. In the first term, each of the two photons represented is created during the elementary one-photon scattering process described above, and each consequently has the same energy $\hbar\omega$ as an incident-field photon. The second term in Eq. (4.10), on the other hand, describes a process in which the sum of the energies of the two scattered photons is exactly $2\hbar\omega$, but in which either photon is allowed to have a continuous range of energies.

The contribution of the one- and two-photon terms to the energy density of the scattered field

may be calculated, in lowest order, directly from Eqs. (4.8)–(4.10). The result, which shows maxima at the frequencies $\omega + \Delta\omega$ and at $\omega - \Delta\omega = \omega_{10}$, is in exact agreement with the result which has previously been obtained by means of the fluctuation-regression theorem,¹⁷ and furthermore agrees exactly with the result obtained by Sokolovskii by the familiar methods of formal scattering theory.⁸

It is interesting, in this connection, to evaluate the expectation values

$$\langle t | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}) | t \rangle \approx_F \langle 0 | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}) | t \rangle_{0F} \text{ as } \mathcal{E}_0 \rightarrow 0, \quad (4.11)$$

and

$$\langle t | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_2) \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_1) | t \rangle \approx_F \langle 0 | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_2) \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_1) | t \rangle_{0F} \text{ as } \mathcal{E}_0 \rightarrow 0, \quad (4.12)$$

which are essentially the one- and two-photon configuration-space wave functions, associated with the momentum-space functions given by Eqs. (4.9) and (4.10), respectively. One finds directly from Eqs. (4.8)–(4.10) and (2.1b) that these functions have the values

$$\langle t | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}) | t \rangle = \frac{(\bar{\lambda} \cdot \bar{\mathcal{E}}_0) \omega^2}{\delta 4\pi c^2} \frac{(\hbar \bar{\lambda}^* \times \bar{\mathbf{r}}) \times \bar{\mathbf{r}}}{r^3} e^{i\omega(r/c-t)} \quad (4.13)$$

and

$$\langle t | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_2) \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_1) | t \rangle = \langle t | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_2) | t \rangle \langle t | \bar{\mathcal{E}}_R(\bar{\mathbf{r}}_1) | t \rangle \times \{1 - \exp[(i\Delta\omega - \frac{1}{2}\kappa)(r_2 - r_1)/c]\}. \quad (4.14)$$

Field moments such as the ones evaluated in Eqs. (4.13) and (4.14) are in general expressible in terms of (suitably retarded) atomic current-correlation functions. The function evaluated in Eq. (4.14), for example, is directly obtainable from the non-stationary atomic correlation function¹⁸

$$g_T^{(0,2)}(t_2, t_1) = \hbar \bar{\lambda}^* \hbar \bar{\lambda}^* \langle [a(t_2) a(t_1)]_T \rangle, \quad (4.15)$$

in which the subscript T denotes time ordering. This function is easily evaluated in the Markoff approximation,¹⁹ which leads to a result in exact agreement with the one found in Eq. (4.14).

A noteworthy feature of the solution in Eq. (4.14) is that it vanishes for $r_2 = r_1$, a fact which is deducible directly from Eq. (4.15) and the identity $a^2 \equiv 0$. Indeed, it is not difficult to show, to all orders in the field strength, that

$$[\bar{\mathcal{E}}_R(\bar{\mathbf{r}})]^2 | t \rangle = 0, \quad (4.16)$$

and hence that one cannot find two scattered photons at the same point in space (or, more generally, at equal distances from the atom) at the same time. The origin of relation (4.16) may be traced to the same relation which holds at the position $\bar{\mathbf{r}} = 0$ of the two-level atom, as is clear

from Eq. (3.21).

B. Incident fields of arbitrary intensity

When the intensity of the incident field becomes appreciable, it no longer remains permissible to ignore the effect of forward-scattering processes, and the term proportional to \mathcal{E}_0^* must therefore be restored in Eqs. (4.2). It is convenient in the case of an intense harmonically oscillating incident field [Eq. (4.4)] to introduce real parameters Ω and θ and a complex parameter $\bar{\Omega}$ by means of the definitions

$$\bar{\lambda} \cdot \bar{\mathcal{E}}_0 \equiv \frac{1}{2} \Omega e^{-i\theta}, \quad \bar{\Omega} \equiv (\Omega^2 + \delta^2)^{1/2}, \quad (4.17)$$

and then to subject the state vectors $|t\rangle_{0F}$ and $|t\rangle_{\lambda F}$ to the complex transformation

$$\begin{aligned} |t\rangle_{0F} &= e^{-iN\omega t} (\xi_+ |t\rangle_{-F} + \xi_- |t\rangle_{+F}), \\ |t\rangle_{\lambda F} &= e^{-i(N+1)\omega t - i\theta} (-\xi_- |t\rangle_{-F} + \xi_+ |t\rangle_{+F}), \end{aligned} \quad (4.18)$$

in which

$$\xi_{\pm} \equiv \left[\frac{1}{2} (1 \pm \delta/\bar{\Omega}) \right]^{1/2}. \quad (4.19)$$

The quantities $|t\rangle_{+F}$ and $|t\rangle_{-F}$ then obey equations of motion which are decoupled in their dependence upon the incident field, while involving the radiation-field operator \mathcal{E}_R^\dagger in a somewhat more complicated way than do Eqs. (4.2). By substituting Eqs. (4.18) into Eqs. (4.2) and (4.4), one finds the relations

$$\begin{aligned} \left(i \frac{d}{dt} + \frac{1}{2}(\delta + \bar{\Omega}) - \mathcal{C}_F \right) |t\rangle_{+F} \\ = \bar{\lambda}^* \cdot \bar{\mathcal{E}}_R^\dagger(0) (\xi_-^2 |t\rangle_{-F} - \xi_+ \xi_- |t\rangle_{+F}) e^{-i\theta}, \end{aligned} \quad (4.20)$$

$$\begin{aligned} \left(i \frac{d}{dt} + \frac{1}{2}(\delta - \bar{\Omega}) - \mathcal{C}_F \right) |t\rangle_{-F} \\ = \bar{\lambda}^* \cdot \bar{\mathcal{E}}_R^\dagger(0) (-\xi_+^2 |t\rangle_{+F} + \xi_+ \xi_- |t\rangle_{-F}) e^{-i\theta}, \end{aligned}$$

in which \mathcal{C}_F is defined by Eq. (4.7a).

The relations in Eqs. (4.20) and (4.18) suggest a picture in which photons are emitted during transitions from either one of a pair of upper states, with energies $\hbar\omega - \frac{1}{2}\hbar \text{Re}(\delta \pm \bar{\Omega})$, to either one of a pair of lower states, with energies $-\frac{1}{2}\hbar \text{Re}(\delta \pm \bar{\Omega})$. In the limit in which the imaginary

parts of these quantities are negligible compared to the real parts, i.e., for $\delta \approx \Delta\omega$ and $\bar{\Omega} \approx [\Omega^2 + (\Delta\omega)^2]^{1/2} \equiv \Omega'$, the energies of the emitted photons, according to this picture, then lie in small intervals about the three energies $\hbar\omega$, $\hbar(\omega + \Omega')$, and $\hbar(\omega - \Omega')$.⁷

The rough validity of this way of picturing the emission process is confirmed by directly solving the coupled equations (4.20). If the incident field vanishes for $t < 0$ and thereafter has the value given by Eq. (4.4), then, as one finds by using the initial condition

$$|t=0\rangle = |0\rangle_a |0\rangle_F \quad (4.21)$$

in Eqs. (4.1) and (4.18), the solutions to Eqs. (4.20) have Laplace transforms

$$|\bar{v}\rangle_{\pm F} \equiv \int_0^\infty dt e^{i\nu t} |t\rangle_{\pm F} \quad (4.22)$$

which are given by the relations

$$\begin{aligned} |\bar{v}\rangle_{\pm F} &= i \xi_{\mp} (\nu + \frac{1}{2}\delta \mp \frac{1}{2}\bar{\Omega} - \mathcal{C}_F) \\ &\times \frac{1}{(\nu + \frac{1}{2}\delta - \mathcal{C}_F)^2 - \frac{1}{4}\bar{\Omega}^2 - \bar{\lambda} \cdot \bar{\mathcal{E}}_0 \bar{\lambda}^* \cdot \bar{\mathcal{E}}_R^\dagger(0)} |0\rangle_F. \end{aligned} \quad (4.23)$$

(The operator fraction in this relation must be understood to represent a series expansion, with λ as a small parameter.)

The state vector for the atom-field system may be expanded in photon-number eigenstates, either by making use of the Laplace transform solutions in Eqs. (4.23), or by directly solving Eqs. (4.20) [or Eqs. (4.2) and (4.4)] subject to the initial condition in Eq. (4.21). One finds that the amplitudes for finding the atom in the states $|0\rangle_a$ or $|1\rangle_a$ with no photons in the field are

$$\begin{aligned} \psi_0(t) \equiv {}_F\langle 0 | t \rangle_{0F} &= \xi_+^2 e^{i(\delta - \bar{\Omega})t/2} + \xi_-^2 e^{i(\delta + \bar{\Omega})t/2} \\ \text{and} \end{aligned} \quad (4.24)$$

$$\begin{aligned} \psi_1(t) \equiv {}_F\langle 0 | t \rangle_{\lambda F} \\ = -(\bar{\lambda} \cdot \bar{\mathcal{E}}_0 / \bar{\Omega}) e^{-i\omega t} (e^{i(\delta - \bar{\Omega})t/2} - e^{i(\delta + \bar{\Omega})t/2}), \end{aligned}$$

respectively, while the corresponding amplitudes for finding *one* photon with specified momentum in the field are

$$\beta_{k\pm}(t) \equiv {}_F\langle 0 | b_k | t \rangle_{\pm F} = \pm 2 \xi_{\mp} \bar{\lambda}^* \cdot \bar{\mathcal{U}}_k^*(0) (\bar{\lambda} \cdot \bar{\mathcal{E}}_0 / \bar{\Omega}) e^{i(\omega - \omega_k + \delta)t/2} \left(\frac{e^{\pm i\bar{\Omega}t/2} \sin \frac{1}{2}(\omega_k - \omega)t}{\omega_k - \omega} - \frac{\sin \frac{1}{2}(\omega_k - \omega \mp \bar{\Omega})t}{\omega_k - \omega \mp \bar{\Omega}} \right) \quad (4.25a)$$

$$\equiv \xi_{\mp} \beta_{k0}(t) e^{i\omega t} \pm \xi_{\pm} \beta_{k1}(t) e^{2i\omega t + i\theta}. \quad (4.25b)$$

The three photon energies mentioned above are here represented, with the energies $\hbar\omega$ and $\hbar(\omega + \text{Re}\bar{\Omega})$ associated with the field state $|t\rangle_{+F}$, and with the energies $\hbar\omega$ and $\hbar(\omega - \text{Re}\bar{\Omega})$ associated with the state $|t\rangle_{-F}$.

The configuration-space wave functions

$$\bar{\mathcal{G}}_{\pm}^{(1)}(\vec{r}, t) \equiv i \sum_k \vec{u}_k(\vec{r}) \beta_{k\pm}(t) \quad (4.26)$$

are found from Eqs. (4.25) and (2.1b) to have the values

$$\begin{aligned} \bar{\mathcal{G}}_{\pm}^{(1)}(\vec{r}, t) = \mp \xi_{\mp} \frac{\vec{\lambda} \cdot \vec{\mathcal{G}}_{\Omega}}{\Omega} \frac{\omega^2}{4\pi c^2} \frac{(\vec{n} \vec{\lambda}^* \times \vec{r}) \times \vec{r}}{r^3} \\ \times \theta(t - r/c) e^{i\omega r/c} e^{i(\delta \pm \bar{\Omega})t/2} (1 - e^{\mp i\bar{\Omega}(t-r/c)}), \end{aligned} \quad (4.27)$$

where

$$\begin{aligned} \theta(\tau) &\equiv 1 \quad \text{for } \tau \geq 0 \\ &\equiv 0 \quad \text{for } \tau < 0. \end{aligned} \quad (4.28)$$

Before discussing the one-photon amplitudes in greater detail, it is useful to examine the time dependence of the vacuum amplitudes as given by Eqs. (4.24). Inasmuch as the quantities $\delta + \bar{\Omega}$ and $\delta - \bar{\Omega}$ are easily shown both to have positive imaginary parts, it follows that both of the amplitudes in Eqs. (4.24) eventually decay to zero. This result is of course a simple consequence of the fact that the norm-preserving Eqs. (4.2) describe the continuing creation (scattering) of photons, and thus the ever-diminishing probability of finding the radiation field to have no photons in it. In the limit of weak incident fields ($\Omega \rightarrow 0$), one has $\xi_+ \approx 1$ and $\xi_- \approx 0$, while the amplitude-decay rates in Eqs. (4.24) are

$$\frac{1}{2} \text{Im}(\delta - \bar{\Omega}) \approx \frac{1}{8} \kappa \Omega^2 / |\delta|^2, \quad \text{as } \Omega \rightarrow 0 \quad (4.29a)$$

and

$$\frac{1}{2} \text{Im}(\delta + \bar{\Omega}) \approx \frac{1}{2} \kappa \quad \text{as } \Omega \rightarrow 0, \quad (4.29b)$$

with the small first rate characterizing the long-time behavior, and the second rate ($\frac{1}{2}\kappa$) representing a transient effect. The amplitude decay rate given by Eq. (4.29a) is in fact equal to (one half of) the total rate at which photons are scattered, in the limit under discussion. In the limit $t \rightarrow \infty$, the asymptotic forms of the solutions in Eqs. (4.24) are then simply the constant solutions obtained with neglect of forward scattering, multiplied by the expected unitarity-preserving time-dependent factor $\exp(-\frac{1}{8} t \kappa \Omega^2 / |\delta|^2)$.

In the limit of intense fields, one has $\bar{\Omega} \rightarrow \Omega$, and the amplitude decay rates under discussion are both equal to $\frac{1}{4}\kappa$. There is no meaningful distinction between transient and long-time behavior in this limit, and the functions $\psi_0(t)$ and $\psi_1(t)$ both de-

cay rapidly to zero, within the natural lifetime of the atom. The probability of finding no photons in the field is simply

$$P^{(0)}(t) = e^{-\kappa t/2}, \quad \Omega \gg \kappa, \quad |\Delta\omega|. \quad (4.30)$$

The one-photon part of the scattered-field state vector contains time-dependent decay factors similar to those which appear in the vacuum part. In the limit of weak incident fields, for example, the solutions for the configuration-space wave functions obtained by making use of Eqs. (4.27) in Eqs. (4.18) are equal to the product of the unitarity-preserving factor $\exp(-\frac{1}{8} t \kappa \Omega^2 / |\delta|^2)$ mentioned above, times the solution for the same quantities obtained with neglect of forward scattering. [The latter solutions differ in form from the asymptotic solution in Eq. (4.13) through the presence of a step-function factor and a transient part, both due to the "switching on" of the field at $t=0$.]

In the limit of intense incident fields, the one-photon wave functions, like the vacuum amplitudes, all contain the exponential factor $e^{-\kappa t/4}$, and again no distinction can be drawn between the transient and long-time parts of the solutions. It is not difficult to show that the total probability of finding one photon in the field in this limit is

$$P^{(1)}(t) = \frac{1}{2} \kappa t e^{-\kappa t/2} [1 - (\sin\Omega t)/\Omega t] \quad (\Omega \gg \kappa, |\Delta\omega|), \quad (4.31)$$

and thus vanishes both at $t=0$ and in the limit $t \rightarrow \infty$.

The time-dependent exponential decrease of the one-photon amplitudes and probabilities is due to the fact that the quantities in question refer to the possibility of finding one *and only one* photon in the field. Their decay to zero is thus caused by the same process that causes the decay of the vacuum amplitudes, namely, by the continued creation of new photons brought about by the scattering process.

C. The one-photon approximation

Before proceeding further, it will be useful to compare the one-photon amplitudes found in Eqs. (4.25) with those found by Stroud,⁷ in a careful analysis which is limited, however, by its exclusion at the outset of terms representing more than one (real) emitted photon from the basic system of equations. One may construct an analogous approximation within the framework of our formalism by expanding the state vectors $|t\rangle_{0F}$ and $|t\rangle_{1F}$ in Eq. (4.1) as

$$\begin{aligned} |t\rangle_{0F} &= |0\rangle_F \psi_0(t) + \sum_k b_k^\dagger |0\rangle_F \beta_{k0}(t), \\ |t\rangle_{1F} &= |0\rangle_F \psi_1(t) + \sum_k b_k^\dagger |0\rangle_F \beta_{k1}(t), \end{aligned} \quad (4.32)$$

similarly excluding terms representing two or more photons. The argument leading to Eqs. (4.2) remains unchanged insofar as it applies to the vacuum parts of the state vectors in Eqs. (4.32), which consequently obey the equations²⁰

$$\begin{aligned} \left(i \frac{d}{dt} - \omega_1 + \frac{1}{2} i \kappa\right) \psi_1(t) + \bar{\lambda} \cdot \bar{\mathcal{E}}_c(0, t) \psi_0(t) &= 0, \\ \left(i \frac{d}{dt} - \omega_0\right) \psi_0(t) + \bar{\lambda}^* \cdot \bar{\mathcal{E}}_c^*(0, t) \psi_1(t) &= 0, \end{aligned} \quad (4.33)$$

in which the damping term represents absorption from the one-photon state. Equations (4.33) are thus unaffected by the approximation in Eqs. (4.32), and in fact have solutions already given in Eqs. (4.24).²⁰

The equations which govern the *one*-photon amplitudes in Eqs. (4.32), on the other hand, are importantly affected by the omission of terms representing more than one photon, since the emission-reabsorption process responsible for radiative damping is not allowed to take place after one photon has been emitted. The one-photon amplitudes consequently obey the undamped equations

$$\left(i \frac{d}{dt} - \omega_1 - \omega_k\right) \beta_{k1}(t) + \bar{\lambda} \cdot \bar{\mathcal{E}}_c(0, t) \beta_{k0}(t) = 0, \quad (4.34)$$

$$\begin{aligned} \left(i \frac{d}{dt} - \omega_0 - \omega_k\right) \beta_{k0}(t) + \bar{\lambda}^* \cdot \bar{\mathcal{E}}_c^*(0, t) \beta_{k1}(t) \\ = i \bar{\lambda}^* \cdot \bar{\mathbf{u}}_k^*(0) \psi_1(t), \end{aligned} \quad (4.35a)$$

the former equation taking the place of the more accurate equation

$$\left(i \frac{d}{dt} - \omega_1 + \frac{1}{2} i \kappa - \omega_k\right) \beta_{k1}(t) + \bar{\lambda} \cdot \bar{\mathcal{E}}_c(0, t) \beta_{k0}(t) = 0, \quad (4.35b)$$

which follows from Eq. (4.2a),²⁰ and in which the damping term represents absorption from the two-photon state.

The solutions to Eqs. (4.34), (4.35a), and (4.33) for the one-photon amplitudes $\beta_{k0}(t)$ and $\beta_{k1}(t)$, corresponding to the initial condition (4.21) and with the incident field given by Eq. (4.4), are

$$\beta_{k0}(t) = e^{-i(\omega_k + \omega_0)t} \{[(\mu_- - \mu_+) \exp[\frac{1}{2} i (\Omega' + \Delta \omega)t] + (\Omega' - -\Omega') + [(\eta_- - \eta_+) \exp[i(\omega_k - \omega + \frac{1}{2} \bar{\Omega} + \frac{1}{2} \delta)t] + (\bar{\Omega} - -\bar{\Omega})]\} \quad (4.36a)$$

and

$$\beta_{k1}(t) = e^{-i(\omega_k + \omega_0)t - i\theta} \{[(\mu_- + \mu_+) \exp[\frac{1}{2} i (\Omega' + \Delta \omega)t] + (\Omega' - -\Omega') + [(\eta_- + \eta_+) \exp[i(\omega_k - \omega + \frac{1}{2} \bar{\Omega} + \frac{1}{2} \delta)t] + (\bar{\Omega} - -\bar{\Omega})]\}, \quad (4.36b)$$

in which

$$\mu_{\pm} = \frac{\pm i \bar{\lambda}^* \cdot \bar{\mathbf{u}}_k^*(0) \frac{1}{4} \bar{\lambda} \cdot \bar{\mathcal{E}}_c(\Omega' - \Delta \omega \mp \Omega)}{\Omega' A_+ B}, \quad (4.36c)$$

$$\eta_{\pm} = \frac{\mp i \bar{\lambda}^* \cdot \bar{\mathbf{u}}_k^*(0) \frac{1}{2} \bar{\lambda} \cdot \bar{\mathcal{E}}_c(\omega - \omega_k \pm \frac{1}{2} \Omega - \frac{1}{2} \bar{\Omega} + \Delta \omega - \frac{1}{2} \delta)}{\bar{\Omega} A_+ A_-}, \quad (4.36d)$$

$$A_{\pm} = \omega - \omega_k + \frac{1}{2} (\pm \Omega' - \bar{\Omega} + \Delta \omega - \delta), \quad (4.36e)$$

$$B = \omega - \omega_k + \frac{1}{2} (\Omega' + \bar{\Omega} + \Delta \omega - \delta), \quad (4.36f)$$

$$\Omega' = [\Omega^2 + (\Delta \omega)^2]^{1/2}, \quad (4.36g)$$

and the remaining parameters are defined by Eqs. (4.7) and (4.17).

When the applied-field frequency coincides exactly with the unshifted resonance frequency, one must put $\Delta \omega = \Delta_0 - \Delta_1$, where $\hbar \Delta_0$ and $\hbar \Delta_1$ are the radiative energy shifts of the states $|0\rangle_a$ and $|1\rangle_a$, calculated in the usual way, in accordance with the discussion at the end of Sec. III. The solutions in

Eqs. (4.36) are then found to be in exact agreement with the ones found by Stroud.^{7b} This agreement should not be surprising, since the methods of solution are in both cases fully quantum mechanical (see Sec. II), share equally the same limiting assumptions [notably conditions (3.19) and (3.25), and the dipole approximation], and are both based on the same one-photon truncation scheme.¹⁶

The solutions in Eqs. (4.36) have been presented here in order to underline the fact that the method developed in this paper, in which the photon-absorption terms are solved for at the outset and thereafter represented by damping terms in the basic system of equations, is fully as accurate as the direct method, in which photon-absorption terms are retained throughout the calculation. Our method, unlike the direct method, can be applied with equal ease when the effects of two and more photons are included, and leads then to the amplitudes given by Eqs. (4.25). There would seem to be every reason to prefer the latter solutions to those in Eqs. (4.36), which are limited by

the one-photon truncation approximation of Eqs. (4.32).

[The solutions in Eqs. (4.25), it may be noted, can be obtained from the approximate solutions in Eqs. (4.36) simply by restoring the damping parameter in the latter solutions, by means of the substitution

$$\Delta\omega \rightarrow \Delta\omega + \frac{1}{2}i\kappa \equiv \delta \quad (4.37)$$

(and hence $\Omega' \rightarrow \bar{\Omega}$) throughout Eqs. (4.36).]

Within a short time interval about the initial time, however, one might expect the elimination of terms representing more than one photon to be unimportant, and indeed a comparison of Eqs. (4.36) with Eqs. (4.25) shows that the two solutions agree quite well near $t=0$. Thus one can say that precisely within the limited time interval within which the field is accurately represented by one photon, the solution for the one-photon amplitude has been accurately obtained by Stroud.^{7b} Outside this interval, it is correspondingly necessary not merely to use the improved solutions in Eqs. (4.25) for the one-photon amplitudes, but to include the contributions to the spectrum made by arbitrary numbers of photons, as represented formally in Eqs. (4.23) and (4.22). (The complete spectrum is evaluated in Sec. V, and is constructed from the vacuum and one-photon amplitudes in Sec. VIII.)

V. FREQUENCY SPECTRUM OF SCATTERED FIELD

The solution for the frequency spectrum of the scattered field can be obtained in a direct fashion from the equations governing the pure state vector for the joint atom-field system by singling out a particular field mode k of frequency ω_k , and then describing the state-space of the radiation field as the direct product of two vector spaces, (a) the state-space for the field mode k , and (b) the joint state-space for all the other field modes. The radiation-field state vectors $|t\rangle_{0F}$ and $|t\rangle_{1F}$ in Eqs. (4.1) then have expansions of the form

$$\begin{aligned} |t\rangle_{0F} &= |0\rangle_k |A(t)\rangle_0 + |1\rangle_k |B_k(t)\rangle_0, \\ |t\rangle_{1F} &= |0\rangle_k |A(t)\rangle_1 + |1\rangle_k |B_k(t)\rangle_1, \end{aligned} \quad (5.1)$$

where $|0\rangle_k$ is the ground state of mode k , and $|1\rangle_k \equiv b_k^\dagger |0\rangle_k$ is the state of mode k containing exactly one photon. The quantities $|A(t)\rangle_{0,1}$ and $|B_k(t)\rangle_{0,1}$ are vectors in the joint state-space for the remaining modes.

The omission in Eq. (5.1) of terms representing more than one photon in the single discrete mode k is easily justified in the limit of infinite quantization volume V , where the frequency separation between adjacent modes approaches zero. The probability $p_n^{(k)}(t)$ of finding n photons in a particular discrete mode k at a given time t is easily shown

from Eq. (2.1b) to be of order V^{-n} , while the field-frequency spectrum involves a sum over field modes and hence is proportional to $VN_k(t)$, where $N_k(t) = \sum_n n p_n^{(k)}(t)$ is the mean number of photons in mode k . It is therefore apparent that in the limit $V \rightarrow \infty$, contributions from terms representing more than one photon in any particular mode approach zero. The expansion (5.1) is then accurate without additional terms, and the mean number of photons in the mode k is just

$$N_k(t) = {}_0\langle B_k(t) | B_k(t) \rangle_0 + {}_1\langle B_k(t) | B_k(t) \rangle_1, \quad (5.2)$$

and is small compared to unity. [The validity of the above remarks requires that an initial condition such as the one in Eq. (4.21) be imposed, in order to ensure that the mean total number of photons be finite at any time t . The limit $V \rightarrow \infty$ must then be taken at any given time in such a way as to allow the quantization volume greatly to exceed the volume occupied by the radiation field. Only then can one ignore the possibility of finding more than one photon in any particular mode.]

It is important to stress that it is the probability of finding more than one photon *in a single mode* which contributes inappreciably in the limit $V \rightarrow \infty$, not the probability of finding more than one photon in the *entire field*. The state vectors $|A(t)\rangle_{0,1}$ and $|B_k(t)\rangle_{0,1}$ each have expansions of the form

$$\begin{aligned} \varphi^{(0)}(t)|0\rangle_F &+ \sum_{(j \neq k)} \varphi_j(t) b_j^\dagger |0\rangle_F \\ &+ \sum_{(j_1, j_2 \neq k)} \varphi_{j_1 j_2}^{(2)}(t) b_{j_1}^\dagger b_{j_2}^\dagger |0\rangle_F + \dots, \end{aligned} \quad (5.3)$$

and describe (as they must) states in which the *total* number of photons in *all* of the radiation field modes is quite unrestricted. [The one-photon approximation in Eqs. (4.32), it may be noted, can be directly represented as in Eqs. (5.1) by eliminating terms representing more than one photon in the expansion (5.3) for $|A(t)\rangle$, and by eliminating all terms except the vacuum amplitude in the expansion for $|B_k(t)\rangle$.]

By substituting Eqs. (5.1) into Eqs. (4.2) and making use of Eqs. (2.1b) and (2.1c), one finds that the state vectors $|A(t)\rangle_{0,1}$ and $|B_k(t)\rangle_{0,1}$ obey the equations

$$\left(i \frac{d}{dt} - \omega_{10} + \frac{1}{2}i\kappa - \frac{H'_{0F}}{\hbar} \right) |A(t)\rangle_1 + \vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) |A(t)\rangle_0 = 0, \quad (5.4a)$$

$$\left(i \frac{d}{dt} - \frac{H'_{0F}}{\hbar} \right) |A(t)\rangle_0 + \vec{\lambda}^* \cdot [\vec{\mathcal{E}}_c^*(0, t) + \vec{\mathcal{E}}_c'^\dagger(0)] |A(t)\rangle_1 = 0, \quad (5.4b)$$

$$\left(i \frac{d}{dt} - \omega_{10} + \frac{1}{2} i \kappa - \omega_k - \frac{H'_{0F}}{\hbar} \right) |B_k(t)\rangle_1 + \vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) |B_k(t)\rangle_0 = 0, \quad (5.4c)$$

and

$$\left(i \frac{d}{dt} - \omega_k - \frac{H'_{0F}}{\hbar} \right) |B_k(t)\rangle_0 + \vec{\lambda}^* \cdot [\vec{\mathcal{E}}_c^*(0, t) + \vec{\mathcal{E}}_R'^{\dagger}(0)] |B_k(t)\rangle_1 = i \vec{\lambda}^* \cdot \vec{u}_k^*(0) |A(t)\rangle_1, \quad (5.4d)$$

in which

$$H'_{0F} \equiv \hbar \sum_{k' (k' \neq k)} \omega_{k'} b_{k'}^{\dagger} b_{k'}, \quad (5.4e)$$

and

$$\vec{\mathcal{E}}_R'(0) \equiv i \sum_{k' (k' \neq k)} \vec{u}_{k'}(0) b_{k'}. \quad (5.4f)$$

The state vectors governed by Eqs. (5.4) represent states with ever-increasing numbers of photons, and hence do not themselves assume steady-state values. [They are in this respect quite different from the state vectors of infinite norm which were obtained with neglect of forward scattering in Sec. IV A.] The rate of emission of photons of specified frequency, however, does

assume a steady-state value after an initial transient regime. One may evaluate the expectation value in Eq. (5.2) by first noting that Eqs. (3.21) and (4.1) imply the relations

$$\vec{\lambda} \cdot \vec{\mathcal{E}}_R(0) |t\rangle_{0F} = \frac{1}{2} i \kappa |t\rangle_{1F}, \quad (5.5)$$

$$\vec{\lambda} \cdot \vec{\mathcal{E}}_R(0) |t\rangle_{1F} = 0,$$

which in turn imply that the state vectors on the right-hand side of Eqs. (5.1) obey the relations

$$\vec{\lambda} \cdot \vec{\mathcal{E}}_R'(0) |A(t)\rangle_0 = \frac{1}{2} i \kappa |A(t)\rangle_1, \quad (5.6a)$$

$$\vec{\lambda} \cdot \vec{\mathcal{E}}_R'(0) |A(t)\rangle_1 = 0, \quad (5.6b)$$

$$\vec{\lambda} \cdot \vec{\mathcal{E}}_R'(0) |B_k(t)\rangle_0 = \frac{1}{2} i \kappa |B_k(t)\rangle_1, \quad (5.6c)$$

$$\vec{\lambda} \cdot \vec{\mathcal{E}}_R'(0) |B_k(t)\rangle_1 = 0. \quad (5.6d)$$

[Terms which make a vanishing contribution in the limit of infinite quantization volume have been omitted in Eqs. (5.6).]

By making use of Eqs. (5.4) and (5.6) in Eq. (5.2), one readily finds that the rate of emission of photons into mode k is

$$W_k \equiv \frac{dN_k(t)}{dt} = \vec{\lambda} \cdot \vec{u}_k(0) {}_1\langle A(t) | B_k(t) \rangle_0 + c.c. \quad (5.7)$$

The right side of this relation may be obtained from the solution to the coupled equations

$$\left(i \frac{d}{dt} - \omega_k + Q(t) \right) \begin{bmatrix} {}_0\langle A(t) | B_k(t) \rangle_1 \\ {}_1\langle A(t) | B_k(t) \rangle_0 \\ {}_1\langle A(t) | B_k(t) \rangle_1 \\ {}_0\langle A(t) | B_k(t) \rangle_0 \end{bmatrix} = i \vec{\lambda}^* \cdot \vec{u}_k^*(0) \begin{bmatrix} 0 \\ {}_1\langle A(t) | A(t) \rangle_1 \\ 0 \\ {}_0\langle A(t) | A(t) \rangle_1 \end{bmatrix}, \quad (5.8)$$

which follow from Eqs. (5.4) and (5.6), and in which $Q(t)$ represents the matrix

$$Q(t) \equiv \begin{bmatrix} -\omega_{10} + \frac{1}{2} i \kappa & 0 & -\vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) & \vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) \\ 0 & \omega_{10} + \frac{1}{2} i \kappa & \vec{\lambda}^* \cdot \vec{\mathcal{E}}_c^*(0, t) & -\vec{\lambda}^* \cdot \vec{\mathcal{E}}_c^*(0, t) \\ -\vec{\lambda}^* \cdot \vec{\mathcal{E}}_c^*(0, t) & \vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) & i \kappa & 0 \\ \vec{\lambda}^* \cdot \vec{\mathcal{E}}_c^*(0, t) & -\vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) & -i \kappa & 0 \end{bmatrix}. \quad (5.9)$$

The quantities on the right side of Eq. (5.8) may be similarly found from the solutions to the equations

$$\left(i \frac{d}{dt} + Q(t) \right) \begin{bmatrix} {}_0\langle A(t) | A(t) \rangle_1 \\ {}_1\langle A(t) | A(t) \rangle_0 \\ {}_1\langle A(t) | A(t) \rangle_1 \\ {}_0\langle A(t) | A(t) \rangle_0 \end{bmatrix} = 0, \quad (5.10)$$

which again follow from Eqs. (5.4) and (5.6).

Subject to the initial condition (4.21), the solu-

tion to Eqs. (5.10), (5.9), and (4.4) for each of the quantities ${}_{\mu}\langle A(t) | A(t) \rangle_{\nu}$ (where $\mu, \nu = 0, 1$) consists of the superposition of a transient and a steady-state part. The steady-state parts of the solutions for the functions ${}_1\langle A(t) | A(t) \rangle_1$ and ${}_0\langle A(t) | A(t) \rangle_1$ are

$$\begin{aligned} {}_1\langle A | A \rangle_1 &= \frac{1}{4} \Omega^2 / (\frac{1}{2} \Omega^2 + |\delta|^2) \equiv \bar{\rho}_{11}, \\ {}_0\langle A(t) | A(t) \rangle_1 &= -e^{-i\omega t} \vec{\lambda} \cdot \vec{\mathcal{E}}_0 \delta^* / (\frac{1}{2} \Omega^2 + |\delta|^2) \\ &\equiv \bar{\rho}_{10} e^{-i\omega t}, \end{aligned} \quad (5.11)$$

with δ and Ω defined by Eqs. (4.7) and (4.17).

The functions ${}_{\mu}\langle A(t)|B_k(t)\rangle_{\nu}$ as determined by Eqs. (5.8), like the solutions to Eqs. (5.10), consist of transient plus steady-state parts. By making use of Eqs. (5.11) in Eq. (5.8), one finds that the steady-state part of the solution for the function ${}_{1}\langle A(t)|B_k(t)\rangle_0$ is independent of time and has the value

$${}_{1}\langle A|B_k\rangle_0 = \frac{-\vec{\lambda}^* \cdot \vec{u}_k^*(0)}{\nu_k f(-i\nu_k)} \left\{ \bar{\rho}_{11} \nu_k [(\nu_k + i\kappa)(\nu_k + \delta) - \frac{1}{2}\Omega^2] + \frac{1}{2}\bar{\rho}_{10} \Omega (\nu_k + i\kappa)(\nu_k + \delta) \right\}, \quad (5.12a)$$

where

$$\nu_k \equiv \omega - \omega_k \quad (5.12b)$$

and

$$f(s) \equiv (s + \kappa)(s + i\delta^*)(s - i\delta) + \Omega^2(s + \frac{1}{2}\kappa). \quad (5.12c)$$

The steady-state rate of emission of photons into the mode k is thus determined by Eqs. (5.7), (5.12), and (5.11). The result so specified is in exact agreement with the value of the emission spectrum which has been obtained previously by means of the Markoff approximation and the quantum fluctuation-regression theorem.⁵ The present derivation is superior to the previous one in that it is based upon an ongoing solution to the equations of motion for the joint correlated atom-field state vector, rather than upon a perturbation-theory calculation involving an assumption of statistical factorization of the states of the atom and the field. The analysis presented here is of course fully quantum mechanical (see Sec. II), and manifestly represents the effects of multiphoton contributions of arbitrary order. [Equations (5.4b), (5.4d), and (5.6) would be violated if the expansion (5.3) of the state vectors were arbitrarily truncated at a finite photon number. Also, in such a truncation scheme, the damping parameters in Eqs. (5.4a) and (5.4c) would be absent in the equations governing the amplitudes representing the greatest allowed number of photons.] The only restrictions upon the validity of the present derivation are in fact the ones already noted at the end of Sec. III.

VI. PROOF OF VALIDITY OF OPTICAL BLOCH EQUATIONS AND QUANTUM FLUCTUATION-REGRESSION THEOREM

The Markoff approximation and the quantum fluctuation-regression theorem upon which the derivation of the emission spectrum in Ref. 5 was based can be directly and rigorously derived from the equations of motion for the joint atom-field state vector $|t\rangle$, without use of perturbation theory and, in particular, without any assumption of sta-

tistical factorization of atom and field states. The elements of the reduced atomic density matrix may be expressed as

$$\rho_{10}(t) = \langle t|a|t\rangle, \quad (6.1a)$$

$$\rho_{01}(t) = \langle t|a^\dagger|t\rangle, \quad (6.1b)$$

$$\rho_{11}(t) = \langle t|a^\dagger a|t\rangle, \quad (6.1c)$$

$$\rho_{00}(t) = \langle t|aa^\dagger|t\rangle. \quad (6.1d)$$

By directly differentiating Eq. (6.1a) and then making use of the time-development equation (3.22) obeyed by $|t\rangle$, one finds the relation

$$\frac{d}{dt} \rho_{10}(t) = (i\hbar)^{-1} \langle t|[a\tilde{H}(t) - \tilde{H}^\dagger(t)a]|t\rangle. \quad (6.2)$$

By making use of Eqs. (3.23) in this relation, one then finds the relation

$$\begin{aligned} \frac{d}{dt} \rho_{10}(t) = & -i(\omega_{10} - \frac{1}{2}i\kappa)\rho_{10}(t) \\ & + i\vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t)[\rho_{00}(t) - \rho_{11}(t)]. \end{aligned} \quad (6.3)$$

Eliminated from the right-hand side is the term involving the radiation field operator in $\tilde{H}^\dagger(t)$,

$$\langle t|a^\dagger \vec{\lambda} \cdot \vec{\mathcal{E}}_R(0)a|t\rangle = \langle t|a^\dagger a \vec{\lambda} \cdot \vec{\mathcal{E}}_R(0)|t\rangle = 0, \quad (6.4)$$

where the last step follows from Eq. (3.21) and the identity $a^2 = 0$. [The identity $a^2 = 0$ also implies directly the vanishing of the term involving \mathcal{E}_R^\dagger in Eqs. (6.2) and (3.23).]

The time-development equation for $\rho_{01}(t)$,

$$\begin{aligned} \frac{d}{dt} \rho_{01}(t) = & i(\omega_{10} + \frac{1}{2}i\kappa)\rho_{01}(t) \\ & - i\vec{\lambda}^* \cdot \vec{\mathcal{E}}_c^*(0, t)[\rho_{00}(t) - \rho_{11}(t)], \end{aligned} \quad (6.5)$$

may be obtained by methods parallel to those used to reach Eq. (6.3) [and using the conjugate of Eq. (3.21)] or simply by taking the complex conjugate of Eq. (6.3).

By differentiating Eq. (6.1c) one finds, with the aid of Eqs. (3.22) and (3.23), that

$$\begin{aligned} \frac{d}{dt} \rho_{11}(t) = & (i\hbar)^{-1} \langle t|[a^\dagger a \tilde{H}(t) - \tilde{H}^\dagger(t)a^\dagger a]|t\rangle \\ = & -\kappa\rho_{11}(t) + i\rho_{01}(t)\vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) \\ & - i\rho_{10}(t)\vec{\lambda}^* \cdot \vec{\mathcal{E}}_c^*(0, t). \end{aligned} \quad (6.6)$$

Here the terms involving the radiation operators \mathcal{E}_R^\dagger and \mathcal{E}_R both vanish, simply due to the identities $a^2 = 0$ and $a^{\dagger 2} = 0$.

By differentiating Eq. (6.1d), one finds, finally, the relation

$$\begin{aligned} \frac{d}{dt} \rho_{00}(t) = & (i\hbar)^{-1} \langle t|[aa^\dagger \tilde{H}(t) - \tilde{H}^\dagger(t)aa^\dagger]|t\rangle \\ = & -i\rho_{01}(t)\vec{\lambda} \cdot \vec{\mathcal{E}}_c(0, t) \\ & + i\rho_{10}(t)\vec{\lambda}^* \cdot \vec{\mathcal{E}}_c^*(0, t) + \kappa\rho_{11}(t). \end{aligned} \quad (6.7)$$

Here the terms involving the radiation field operators do not vanish, but instead have the value

$$\begin{aligned} i \langle t | a a^\dagger \tilde{\lambda}^* \cdot \tilde{\mathcal{E}}_R^\dagger(0) a | t \rangle - i \langle t | a^\dagger \tilde{\lambda} \cdot \tilde{\mathcal{E}}_R(0) a a^\dagger | t \rangle \\ = i \langle t | \tilde{\lambda}^* \cdot \tilde{\mathcal{E}}_R^\dagger(0) a a^\dagger a | t \rangle - i \langle t | a^\dagger a a^\dagger \tilde{\lambda} \cdot \tilde{\mathcal{E}}_R(0) | t \rangle \\ = \kappa \rho_{11}(t), \end{aligned} \quad (6.8)$$

the last step following from Eq. (3.21) and its conjugate.

The coupled equations (6.3) and (6.5)–(6.7) are identical to those obtained by means of the Markoff approximation. [See, for example, Ref. 4 or Eqs. (3.13) of Ref. 5.] It follows immediately from this fact that the mean values of the scattered-field amplitude and of the scattered-field intensity, which are proportional to the values of $\rho_{10}(t)$ and $\rho_{11}(t)$, respectively, at suitably retarded times, are also given correctly by the Markoff approximation.²¹

The frequency spectrum of the scattered radiation may be found by evaluating the mean occupation number $N_k(t) \equiv \langle t | b_k^\dagger b_k | t \rangle$, as a function of the photon frequency ω_k . This is most easily done by working in the Heisenberg picture, where it can be shown that, subject to the initial condition in Eq. (4.21), N_k may be expressed as

$$\begin{aligned} N_k(T) = |\tilde{\lambda} \cdot \tilde{\mathbf{u}}_k(0)|^2 \int_0^T dt \int_0^t dt' e^{i\omega_k(t-t')} \\ \times \langle a^\dagger(t') a(t) \rangle + \text{c.c.} \end{aligned} \quad (6.9)$$

In this relation, the atomic correlation function $\langle a^\dagger(t') a(t) \rangle$ (which also directly determines the cross-spectral correlation function $\langle \tilde{\mathcal{E}}_R^\dagger(\bar{\mathbf{r}}', t') \tilde{\mathcal{E}}_R(\bar{\mathbf{r}}, t) \rangle$ for the scattered field^{5,18} at any pair of space-time points) may be expressed as

$$\langle a^\dagger(t') a(t) \rangle = \langle t' | a^\dagger U(t', t) a | t \rangle, \quad (6.10)$$

where $|t\rangle$ is the Schrödinger state vector for the atom-field system, and $U(t, t')$ is the unitary time-development operator for the system, defined by the relations

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} U(t, t') = H(t) U(t, t'), \\ U(t', t') = 1. \end{aligned} \quad (6.11)$$

It should be emphasized that $H(t)$ here represents the full, Hermitian Hamiltonian given by Eqs. (3.1) and (3.2), not the modified, non-Hermitian Hamiltonian $\tilde{H}(t)$ in Eq. (3.23). It will now be shown, however, that the time-dependent vector

$$|\psi(t; t')\rangle \equiv U(t, t') a | t' \rangle, \quad (6.12)$$

in terms of which the quantity evaluated in Eq. (6.10) may be expressed as

$$\langle a^\dagger(t') a(t) \rangle = \langle \psi(t; t') | a | t \rangle, \quad (6.13)$$

obeys the time-development equation

$$i\hbar \frac{\partial}{\partial t} |\psi(t; t')\rangle = \tilde{H}(t) |\psi(t; t')\rangle, \quad t \geq t', \quad (6.14)$$

where $\tilde{H}(t)$ is defined by Eqs. (3.23).

To prove this, it is useful to think of t' as a fixed initial time, and to define interaction-picture operators and an interaction-picture vector $|\psi(t; t')\rangle'$ in terms of the time difference $t - t'$ rather than in terms of t alone. One may then seek to establish, in analogy with Eq. (3.20), the relation

$$\tilde{\mathcal{E}}_R'(0, \bar{t}) |\psi(t; t')\rangle' = i\tilde{\lambda}^* F(\bar{t} - t) a'(\bar{t}) |\psi(t; t')\rangle', \quad \bar{t} \geq t. \quad (6.15)$$

This relation is in fact satisfied at $t = t'$, where $|\psi(t; t')\rangle' = a | t' \rangle$: Both sides of it then vanish, the right-hand side simply by virtue of the identity $a^2 = 0$, the left-hand side as a consequence of the relation

$$\tilde{\mathcal{E}}_R'(0, \bar{t}) a | t' \rangle = a \tilde{\mathcal{E}}_R'(0, \bar{t}) | t' \rangle = 0, \quad (6.16)$$

in which the last step follows from Eq. (3.20) and the identity $a^2 = 0$.

It is clear from the nature of the iteration procedure used to establish Eq. (3.20) that the relation in question does not require for its validity the particular initial condition in Eq. (2.12), but, more generally, must remain valid for all times t following some initial time t' , provided only that it is satisfied at $t = t'$ (though for all $\bar{t} > t'$). By the same reasoning, the validity of Eq. (6.15) follows from the validity (established above) of the same relation at $t = t'$, and one then arrives at Eq. (6.14) through steps analogous to those leading to Eq. (3.22).

(The strict satisfaction of Eq. (6.15) at $t = t'$ is in fact not quite necessary in this derivation. If the operator a^\dagger rather than a appeared in the definition (6.12) [as it would, for example, in an evaluation of the correlation function in Eq. (4.15)], then the relation (6.15), though not satisfied exactly at $t = t'$, would become satisfied soon thereafter, due to the rapid falloff of the function $F(\tau)$. The small time interval within which the relation was not satisfied initially would introduce a negligible error into the final result at later times.)

By differentiating Eq. (6.13) and making use of Eqs. (3.22) and (6.14), one finds the relation

$$\begin{aligned} \frac{\partial}{\partial t} \langle a^\dagger(t') a(t) \rangle = (i\hbar)^{-1} \langle \psi(t; t') | [a \tilde{H}(t) - \tilde{H}^\dagger(t) a] | t \rangle, \\ t \geq t', \end{aligned} \quad (6.17)$$

in analogy to Eq. (6.2) for $(d/dt)\langle a(t) \rangle$. It is clear

therefore that by substituting Eq. (3.23) for $\tilde{H}(t)$ into Eq. (6.17) and evaluating the time derivatives of the expressions which then appear, one is led to a set of four coupled equations, identical in form to Eqs. (6.3) and (6.5)–(6.7), but involving the four functions

$$\begin{aligned}\langle \psi(t; t') | a | t \rangle &= \langle a^\dagger(t') a(t) \rangle \equiv R_{10}(t; t'), \\ \langle \psi(t; t') | a^\dagger | t \rangle &= \langle a^\dagger(t') a^\dagger(t) \rangle \equiv R_{01}(t; t'), \\ \langle \psi(t; t') | a^\dagger a | t \rangle &= \langle a^\dagger(t') a^\dagger(t) a(t) \rangle \equiv R_{11}(t; t'), \\ \langle \psi(t; t') | a a^\dagger | t \rangle &= \langle a^\dagger(t') a(t) a^\dagger(t) \rangle \equiv R_{00}(t; t'),\end{aligned}\quad (6.18)$$

in place of the functions given by Eqs. (6.1).

This demonstration of the formal correspondence between the equations of motion for the atomic single-time expectation values and those for the two-time correlation functions completes the proof of the validity of the quantum *fluctuation-regression theorem* as it applies in this case, and explains the accuracy of the expressions which have been found through its use for the frequency spectrum of the scattered field.⁵

VII. PHOTON-NUMBER DISTRIBUTIONS: n -PHOTON CONTRIBUTION TO COMPLETE SOLUTIONS

Our discussion of the state of the atom and of the emission-field spectrum need not be confined to an evaluation of quantities in which multiphoton contributions of all orders are summed. It is a simple matter to divide the state-space of the atom-field system into subspaces representing fixed numbers of radiated photons, and to evaluate separately the contributions from each subspace. The state vectors $|t\rangle_{\mu F}$ (where $\mu = 0, 1$) in Eq. (4.1) have expansions of the form

$$|t\rangle_{\mu F} = \sum_{n=0}^{\infty} |t\rangle_{\mu F}^{(n)}, \quad (7.1a)$$

in which each term is an eigenstate of the photon-number operator N defined by Eq. (4.5b),

$$(N - n) |t\rangle_{\mu F}^{(n)} \equiv 0. \quad (7.1b)$$

It is useful to introduce a restricted species of atomic-density matrix $\rho_{\mu\nu}^{(n)}$ by projection onto the subspace with exactly n photons, i.e., by means of the definition

$$\rho_{\mu\nu}^{(n)}(t) \equiv \frac{\langle \nu | \rho(t) | \mu \rangle_{\mu F}^{(n)}}{\nu_F \mu_F}. \quad (7.2)$$

The equations of motion for the quantities $\rho_{\mu\nu}^{(n)}(t)$ follow directly from Eqs. (4.2) and the relations

$$\begin{aligned}\tilde{\lambda} \cdot \tilde{\mathcal{E}}_R^\dagger(0) |t\rangle_{0F}^{(n)} &= \frac{1}{2} i \kappa |t\rangle_{1F}^{(n-1)}, \\ \tilde{\lambda} \cdot \tilde{\mathcal{E}}_R^\dagger(0) |t\rangle_{1F}^{(n)} &= 0,\end{aligned}\quad (7.3)$$

which follow directly from Eqs. (5.5) and (7.1). One finds

$$\left(\frac{d}{dt} + i \omega_{10} + \frac{1}{2} \kappa \right) \rho_{10}^{(n)}(t) + i \tilde{\lambda} \cdot \tilde{\mathcal{E}}_c(0, t) [\rho_{11}^{(n)}(t) - \rho_{00}^{(n)}(t)] = 0, \quad (7.4a)$$

$$\begin{aligned}\left(\frac{d}{dt} - i \omega_{10} + \frac{1}{2} \kappa \right) \rho_{01}^{(n)}(t) - i \tilde{\lambda}^* \cdot \tilde{\mathcal{E}}_c^*(0, t) [\rho_{11}^{(n)}(t) - \rho_{00}^{(n)}(t)] \\ = 0,\end{aligned}\quad (7.4b)$$

$$\begin{aligned}\left(\frac{d}{dt} + \kappa \right) \rho_{11}^{(n)}(t) + i \tilde{\lambda}^* \cdot \tilde{\mathcal{E}}_c^*(0, t) \rho_{10}^{(n)}(t) - i \tilde{\lambda} \cdot \tilde{\mathcal{E}}_c(0, t) \rho_{01}^{(n)}(t) \\ = 0,\end{aligned}\quad (7.4c)$$

$$\begin{aligned}\frac{d}{dt} \rho_{00}^{(n)}(t) - i \tilde{\lambda}^* \cdot \tilde{\mathcal{E}}_c^*(0, t) \rho_{10}^{(n)}(t) + i \tilde{\lambda} \cdot \tilde{\mathcal{E}}_c(0, t) \rho_{01}^{(n)}(t) \\ = \kappa \rho_{11}^{(n-1)}(t),\end{aligned}\quad (7.4d)$$

with the right side of Eq. (7.4d) understood as zero for $n=0$.

In contrast to the homogeneous equations (6.3) and (6.5)–(6.7) which govern the full atomic-density matrix $\rho(t)$, the Eqs. (7.4) which govern the projected atomic-density matrix $\rho^{(n)}(t)$ are inhomogeneous in form, the inhomogeneous term [i.e., the right side of Eq. (7.4d)] representing the atomic transition $|1\rangle_a \rightarrow |0\rangle_a$ which accompanies the emission of the n th photon.

The infinite hierarchy of Eqs. (7.4), for $n=0, 1, \dots$, is easily solved by iteration, subject to the initial condition (4.21) and with the applied field given by Eq. (4.4). The trace

$$P^{(n)}(t) = \rho_{00}^{(n)}(t) + \rho_{11}^{(n)}(t), \quad (7.5)$$

which represents the probability of finding exactly n photons in the radiation field, is found in this way to have Laplace transform

$$\tilde{P}^{(n)}(\nu) \equiv \int_0^\infty P^{(n)}(t) e^{i\nu t} dt = \frac{i \kappa^n \left(-\frac{1}{2} i \Omega^2 \bar{\nu} \right)^n \left\{ \left(\bar{\nu} + \frac{1}{2} i \kappa \right) \left[\bar{\nu}^2 - (\Delta \omega)^2 \right] - \Omega^2 \bar{\nu} \right\}}{\left[(\bar{\nu}^2 - \Omega_R^2) (\bar{\nu}^2 + \Omega_I^2) \right]^{n+1}}, \quad (7.6)$$

where $\bar{\nu} \equiv \nu + \frac{1}{2}i\kappa$, and $\Delta\omega$, Ω , and $\bar{\Omega}_R + i\bar{\Omega}_I \equiv \bar{\Omega}$ are defined by Eqs. (4.7c) and (4.17).

When the applied field is exactly on resonance ($\Delta\omega = 0$), $P^{(n)}(t)$ has the general form

$$P^{(n)}(t) = e^{-\kappa t/2} [C_n(t) + D_n(t)e^{-i\bar{\Omega}t} + D_n^*(t)e^{i\bar{\Omega}t}], \quad (7.7)$$

where $C_n(t)$ and $D_n(t)$ are n th degree polynomials in t , obtainable from Eq. (7.6). In the limit of intense applied fields, $P^{(n)}(t)$ is accurately represented after a very short time interval following the switching on of the field by the Poisson distribution

$$P^{(n)}(t) = e^{-\kappa t/2} (\frac{1}{2}\kappa t)^n / n! \quad \text{for } \Omega \gg \kappa, |\Delta\omega|; \quad \Omega t \gg 1. \quad (7.8)$$

For more general applied-field intensities, a good approximation to the solution to Eqs. (7.4) can be found for large values of n and t , simply by recognizing that the solution is then weakly dependent on n , and consequently admits the substitution

$$\rho_{11}^{(n-1)}(t) - \rho_{11}^{(n)}(t) \quad (7.9)$$

on the right side of Eq. (7.4d). Equations (7.4) under this substitution have the same form as the homogeneous equations (6.3) and (6.5)–(6.7) which govern the full atomic-density matrix. By recognizing then that the quantities $\rho_{11}^{(n)}(t)$, $\rho_{00}^{(n)}(t)$, $e^{i\omega t}\rho_{10}^{(n)}(t)$, and $e^{-i\omega t}\rho_{01}^{(n)}(t)$ are slowly varying functions of time in the limit under consideration, one directly finds the solution

$$\rho_{\mu\nu}^{(n)}(t) = \bar{\rho}_{\mu\nu}(t)P^{(n)}(t), \quad n \gg 1, \quad (7.10)$$

in which $\bar{\rho}_{\mu\nu}(t)$ is the steady-state solution to the optical Bloch equations (6.3) and (6.5)–(6.7), and the trace $P^{(n)}(t)$ is a slowly varying function of time which, according to Eqs. (7.5), (7.4c), and (7.4d), obeys the equation

$$\begin{aligned} \frac{d}{dt} P^{(n)}(t) &= \kappa [\rho_{11}^{(n-1)}(t) - \rho_{11}^{(n)}(t)] \\ &= \kappa \bar{\rho}_{11} [P^{(n-1)}(t) - P^{(n)}(t)], \quad n \gg 1, \end{aligned} \quad (7.11)$$

the latter relation following from Eq. (7.10).

It follows immediately from Eq. (7.11) that the photon-number probability $P^{(n)}(t)$ for large n is well approximated at the (large) values of the time t at which it is appreciable by the Poisson formula with mean photon number $\kappa \bar{\rho}_{11} t$,

$$P^{(n)}(t) = e^{-\kappa \bar{\rho}_{11} t} (\kappa \bar{\rho}_{11} t)^n / n!, \quad n \gg 1, \quad (7.12)$$

where $\bar{\rho}_{11}$ represents the steady-state probability

of finding the atom in its excited state. Equation (7.12) is of course just the solution which would be implied by a picture of the emission process as consisting of random events occurring at the mean rate $\kappa \bar{\rho}_{11}$.

The solution given by Eqs. (7.10) and (7.12) describes an approximate degree of statistical independence of atom and field, which should not be surprising, since most of the field has been radiated at times far previous to the time at which the quantities in question are evaluated. It need hardly be emphasized that no semiclassical reasoning of any kind whatever has been employed in reaching the relations in question, nor has any Markoffian assumption been made. The results of this section all follow directly from the rigorously derived equations of motion (4.2) and subsidiary condition (3.21) obeyed by the joint atom-field state vector $|t\rangle$, and hence in effect represent straightforward reductions of the complete solutions for $|t\rangle$ found formally in Sec. IV B.

An evaluation of the contribution to the spectrum made by a particular (n -photon) term in the expansion of the radiated-field state vector can be carried out in terms closely resembling those of the preceding discussion. One may begin by expanding the vectors $|A(t)\rangle$ and $|B(t)\rangle$ in Eqs. (5.1) in photon-number eigenstates, with n representing the total number of photons in all the field modes other than the particular mode k under consideration. [These effectively comprise the entirety of the field in the limit of infinite quantization volume.] It is then a simple matter to derive equations governing the n -photon projections of the quantities appearing in Eqs. (5.7), (5.8), and (5.10). By reasoning similar to that used to reach Eq. (7.10), one then finds that the probability of finding one photon in mode k and n photons in the remaining modes of the field is well approximated for large n by the expression

$$W_k^{(n)}(t) = W_k t P^{(n)}(t), \quad n \gg 1, \quad (7.13)$$

in which W_k is the steady-state emission rate given by Eqs. (5.7), (5.12), and (5.11), and $P^{(n)}(t)$ is given by Eq. (7.12).

It should be understood that while the asymptotic expressions found here for large n represent good approximations in a *quantitative* sense, the analytic forms of the exact n -photon solutions differ considerably from those of the full solutions. The wave function $\beta^{(n)}(t)$ in the expansion

$$|t\rangle_{\mu F}^{(n)} = \frac{1}{n!} \sum_{k_1 \dots k_n} \beta_{\mu k_1 \dots k_n}^{(n)}(t) b_{k_1}^\dagger \dots b_{k_n}^\dagger |0\rangle_F \quad (7.14)$$

can be evaluated directly by inverting the Laplace transform of the n th term in the expansion of Eq. (4.23), with the aid of Eqs. (2.1b), (4.7), and (4.17)–(4.19). The solution for $\beta^{(n)}(t)$ found in this

way consists of a superposition of $2n+1$ decaying exponential functions of time, containing singular resonant denominators similar to those in Eqs. (4.25). Like the one-photon frequency spectrum implied by Eqs. (4.25), the n -photon spectrum thus contains terms with time-dependent widths. Indeed, for intense applied fields, the widths of the lines in the n -photon spectrum are all inversely proportional to time, and are entirely independent of κ . The approximate equality (7.13) between the n -photon spectrum and the full spectrum owes its

validity to the narrowness of the time interval during which the n -photon probability $P^{(n)}(t)$, as given by Eq. (7.12), is appreciable.

The configuration-space wave function

$$\mathcal{G}_{\pm}^{(n)}(\vec{r}_1, \dots, \vec{r}_n, t) \equiv \sum_{k_1 \dots k_n} \beta_{\pm k_1 \dots k_n}^{(n)}(t) \prod_j \vec{u}_{k_j}(\vec{r}_j) \quad (7.15)$$

is found from the solutions described above to be given by the relation

$$\mathcal{G}_{\pm}^{(n)}(\vec{r}_1, \dots, \vec{r}_n, t) = \xi_{\mp} \left(\prod_{j=1}^n \vec{\mathcal{E}}_s(\vec{r}_j) \right) (\pm \vec{\lambda} \cdot \vec{\mathcal{E}}_0 / \Omega)^n \theta(t - r_1/c) e^{i(\delta \pm \bar{\Omega})t/2} (1 - e^{\mp i \bar{\Omega}(t - r_1/c)}) \prod_{j=1}^{n-1} (1 - e^{\mp i \bar{\Omega}(r_j - r_{j+1})/c})$$

for $r_1 > r_2 \dots > r_n$ (7.16a)

(and with other orderings determined by symmetry), where $\vec{\mathcal{E}}_s(\vec{r})$ is the elementary one-photon function

$$\vec{\mathcal{E}}_s(\vec{r}) \equiv \frac{-\omega^2}{4\pi c^2} \frac{(\hbar \vec{\lambda}^* \times \vec{r}) \times \vec{r}}{r^3} e^{i\omega r/c}, \quad (7.16b)$$

and $\theta(t - r_1/c)$ is a unit step function which vanishes unless $r_1 < ct$.

Smithers and Freedhoff²² have recently evaluated momentum-space wave functions in the strong-field limit, in an approximation which omits all but one of the $2n+1$ oscillating terms in the full solution, and which moreover neglects the

damping of the term which is retained. All $2n+1$ terms in the full solution, however, are damped by the same exponential factor $e^{-\kappa t/4}$ in the strong-field limit under consideration, and hence the justification for retaining one term while dropping the others is not clear.

That the approximation of Smithers and Freedhoff does not yield a fully accurate description of the steady-state regime becomes evident upon an evaluation of the configuration-space wave functions. The full solution for $\mathcal{G}_{\pm}^{(n)}(\vec{r}_1, \dots, \vec{r}_n, t)$ is found from Eq. (7.16a) to have the strong-field limit

$$\mathcal{G}_{\pm}^{(n)}(\vec{r}_1, \dots, \vec{r}_n, t) = e^{-\kappa t/4} (i^n / \sqrt{2}) \left(\prod_{j=1}^n \vec{\mathcal{E}}_s(\vec{r}_j) \right) e^{\pm i \Omega r_n / 2c} \theta(t - r_1/c) \sin \frac{1}{2} \Omega (t - r_1/c) \prod_{j=1}^{n-1} \sin \left[\frac{1}{2} \Omega (r_j - r_{j+1}) / c \right]$$

for $r_1 > r_2 \dots > r_n$, $\Omega \gg \kappa, |\Delta\omega|$, (7.17)

while for t large, the expression for the same quantity as evaluated from the solution given by Smithers and Freedhoff is just $e^{\kappa r_n / 4c}$ times the expression given by Eq. (7.17).

Thus, in place of the factor $e^{-\kappa t/4}$ which damps the full n -photon solution (and which accurately represents the further creation of photons), there appears in Smithers' and Freedhoff's solution the factor $e^{-(\kappa/4)(t - r_n/c)}$, which leaves their solution undamped, while implying that all of the radiated photons in it are located far from the atom (at a distance ct from it), within a thin spherical shell, of thickness comparable to the transient atomic-

pulse length c/κ . In the full solution, by contrast, the photons are uniformly distributed between $r=0$ and $r=ct$, as one would expect them to be in the steady-state regime.

An exact integral expression for the n -photon contribution to the frequency spectrum can be obtained in the strong-field limit by using the solution in Eq. (7.17) to evaluate the quantity ${}^{(n)}\langle t | \vec{\mathcal{E}}_R^{\dagger}(\vec{r}') \vec{\mathcal{E}}_R(\vec{r}) | t \rangle^{(n)}$, and then performing an appropriate wave-vector decomposition of the function so obtained. In this way one finds, after some work, the relation

$$N_k^{(n)}(t) = P^{(n)}(t) |\vec{\lambda} \cdot \vec{u}_k(0)|^2 \int_0^{ct} dr \int_0^r dr' c^{-2} e^{ik(r'-r)} g^{(n)}(r-r'; t) + \text{c.c.}, \quad \Omega \gg \kappa, |\Delta\omega|, t^{-1} \quad (7.18a)$$

for $n=0, 1, \dots$, where

$$g^{(n)}(r; t) = e^{i\omega r/c} \left[\frac{1}{4}(1 - r/ct)^n + \frac{1}{8}(1 - \frac{3}{2}r/ct)^n (e^{i\Omega r/c} + e^{-i\Omega r/c}) \right], \quad r > 0, \quad (7.18b)$$

$k = \omega_k/c$, and $P^{(n)}(t)$ is given by Eq. (7.8).

By summing the solution in Eqs. (7.18) over n , one finds the full solution $N_k(t) = W_k t$, where W_k is the constant spectral emission rate found in Ref. 5 for the strong-field limit, consisting of a central peak of width $\frac{1}{2}\kappa$ and two sidebands, each of width

$\frac{3}{4}\kappa$. That the n -photon contribution specified by Eqs. (7.18) is proportional, for n large, to the full solution, follows from the fact that $g^{(n)}(r; t)$ is then well approximated, where it is appreciable, as a sum of three exponential functions, thus implying through Eq. (7.18a) the approximate relation

$$N_k^{(n)}(t) \cong t P^{(n)}(t) |\vec{\lambda} \cdot \vec{u}_k(0)|^2 \left(\frac{\frac{1}{2}n/t}{(\omega_k - \omega)^2 + (n/t)^2} + \frac{\frac{3}{8}n/t}{(\omega_k - \omega - \Omega)^2 + (\frac{3}{2}n/t)^2} + \frac{\frac{3}{8}n/t}{(\omega_k - \omega + \Omega)^2 + (\frac{3}{2}n/t)^2} \right), \quad n \gg 1, \Omega \gg \kappa, |\Delta\omega|, t^{-1}. \quad (7.19)$$

The time-dependent widths in this relation, as stated previously, differ little from the widths associated with the (constant) steady-state solution, during the small time interval about the time $t = 2n/\kappa$ within which the function $P^{(n)}(t)$ is appreciable.

VIII. CONSTRUCTION OF MULTIPHOTON SPECTRUM FROM VACUUM- AND ONE-PHOTON AMPLITUDES

The projected atomic density matrix $\rho_{\mu\nu}^{(n)}(t)$, which describes the state of the atom when exactly n photons are present in the field, obeys Eqs. (7.4) in which the photon-emission process is represented by decay parameters in the homogeneous parts (the left sides), as well as by a production rate in the inhomogeneous part [the right side of Eq. (7.4d)]. Inasmuch as the decay rates are all obtainable simply by adding the imaginary term $-\frac{1}{2}i\kappa$ to the energy of the upper atomic state, the homogeneous parts of Eqs. (7.4) have product solutions of the form $\psi_\mu(t)\psi_\nu^*(t)$, with the pure-state amplitude functions $\psi_\mu(t)$ obeying the vacuum-amplitude equations (4.33).

Indeed, the full solution to Eqs. (7.4) for $\rho^{(n)}(t)$ can be directly expressed in terms of $\rho^{(n-1)}(t)$ as

$$\rho_{\mu\nu}^{(n)}(t) = \int_0^t \psi_\mu(t; t_0) \psi_\nu^*(t; t_0) \rho_{11}^{(n-1)}(t_0) \kappa dt_0, \quad (8.1)$$

where the pure-state amplitudes $\psi_\mu(t; t_0)$ are the solutions (4.24) to Eqs. (4.33), with the initial time (at which the atom is in its ground state) taken at $t = t_0$.

The full (unprojected) atomic density matrix, it may be noted, can be found in the steady-state regime from the similar equation

$$\bar{\rho}_{\mu\nu}(t) = \bar{\rho}_{11} \int_{-\infty}^t \psi_\mu(t; t_0) \psi_\nu^*(t; t_0) \kappa dt_0, \quad (8.2)$$

with the constant parameter $\bar{\rho}_{11}$ determined by the relation $\text{tr}\bar{\rho} = 1$.

It is interesting that the statistical properties of the driven radiating atom can be described entirely in terms of pure-state atomic wave functions, if a hierarchy of such functions is introduced, in which each member corresponds to the presence of a given number of photons in the field. One may introduce functions $\psi_\mu^{(n)}(t|t_n \dots t_1)$ (where $0 \leq t_1 \leq \dots \leq t_n \leq t < \infty$, and $n=0, 1, \dots$) which obey Eqs. (4.33) with t as the time variable, and in which the times t_1, \dots, t_n (which should be thought of as production instants for the radiated photons) are to be averaged over in the evaluation of atomic expectation values. One finds indeed that Eqs. (7.4) are obeyed exactly by the quantities

$$\rho_{\mu\nu}^{(n)}(t) = \int_0^t \kappa dt_n \int_0^{t_n} \kappa dt_{n-1} \dots \int_0^{t_2} \kappa dt_1 \times \psi_\mu^{(n)}(t|t_n, \dots, t_1) \psi_\nu^{(n)*}(t|t_n, \dots, t_1), \quad (8.3)$$

provided that the functions $\psi_\mu^{(n)}(t|t_n, \dots, t_1)$ obey Eqs. (4.33) and satisfy the initial conditions

$$\psi_0^{(n)}(t_n|t_n, \dots, t_1) \equiv \psi_1^{(n-1)}(t_n|t_{n-1}, \dots, t_1), \quad (8.4a)$$

$$\psi_1^{(n)}(t_n|t_n, \dots, t_1) \equiv 0. \quad (8.4b)$$

The picture suggested by this formal construction is one in which a large number of atomic "systems" coexist at any time, each one of which is described by a pure-state wave function which obeys Eqs. (4.33), and which consequently has norm decaying to zero. New systems in the atomic ground state are constantly being created from the existing systems, at rate κ but according to Eq. (8.4a) with quantum-mechanical amplitude ψ_1 , where ψ_1 is the upper-state amplitude for the parent system at the time the new system is created from it. (The actual probability of creat-

ing a new system in time dt is then $\kappa|\psi_1|^2 dt$, the expected spontaneous-emission probability.)

The description just given of the radiative-damping process deserves to be contrasted with the one which applies when the atom is abruptly returned to its ground state by a collisional process, or, more generally, by any abrupt external random process (see Sec. IX). In that case, though pure-state amplitudes may be used to describe the system between collisions, they cannot by themselves be used to describe the damping caused by the collisional process. A particular system in the collisional case must be described by *undamped* equations of motion up until the time a collision takes place, at which time it must be postulated to *cease to exist abruptly*, its place taken by a new system in its ground state, with quantum-mechanical norm equal to unity. The wave function for the old system is thus quenched abruptly to zero by a classical random process, and it is this quenching alone which introduces damping terms into the equations governing the atomic density matrix. In the radiative case, by contrast, the damping occurs entirely within the equations governing the pure-state wave functions, and the

parent system correspondingly continues to exist, though with uniformly decaying amplitude, even as new systems are created from it.

The frequency spectrum of the radiation field can be evaluated by a simple generalization of the method just shown to describe the atomic density matrix, involving the use of one-photon amplitudes $\beta_{k\mu}(t)$ along with the vacuum amplitudes $\psi_\mu(t)$ used so far. One may begin by isolating a particular field mode k as in Sec. V, and consider this mode along with the atom to constitute a single quantum system, described by the four-component wave function $(\psi_0(t), \psi_1(t), \beta_{k0}(t), \beta_{k1}(t))$, with amplitudes which obey Eqs. (4.33) and (4.35). (The exclusion of terms representing more than one photon in a particular mode was shown to be justified in the limit of infinite quantization volume in Sec. V.) The emission of photons into the remainder of the field modes is represented, as before, by introducing a hierarchy of pure-state wave functions, which now include one-photon wave functions $\beta_{k\mu}^{(n)}(t|t_n, \dots, t_1)$. The validity of this construction is shown by the fact that Eqs. (5.7), (5.8), and (5.10) are obeyed exactly by the quantities

$$N_k(t) = \sum_{n=0}^{\infty} \int_0^t \kappa dt_n \int_0^{t_n} \kappa dt_{n-1} \cdots \int_0^{t_2} \kappa dt_1 \sum_{\mu=0,1} |\beta_{k\mu}^{(n)}(t|t_n, \dots, t_1)|^2, \quad (8.5a)$$

$$\mu \langle A(t) | B_k(t) \rangle_\nu = \sum_{n=0}^{\infty} \int_0^t \kappa dt_n \int_0^{t_n} \kappa dt_{n-1} \cdots \int_0^{t_2} \kappa dt_1 \psi_\mu^{(n)*}(t|t_n, \dots, t_1) \beta_{k\nu}^{(n)}(t|t_n, \dots, t_1), \quad (8.5b)$$

$$\mu \langle A(t) | A(t) \rangle_\nu = \sum_{n=0}^{\infty} \int_0^t \kappa dt_n \int_0^{t_n} \kappa dt_{n-1} \cdots \int_0^{t_2} \kappa dt_1 \psi_\mu^{(n)*}(t|t_n, \dots, t_1) \psi_\nu^{(n)}(t|t_n, \dots, t_1) = \rho_{\nu\mu}(t), \quad (8.5c)$$

provided that the functions $\psi_\mu^{(n)}(t|t_n, \dots, t_1)$ and $\beta_{k\mu}^{(n)}(t|t_n, \dots, t_1)$ obey Eqs. (4.35) and (4.33), and satisfy the initial condition (8.4) and the similar condition

$$\beta_{k0}^{(n)}(t_n|t_n, \dots, t_1) = \beta_{k1}^{(n-1)}(t_n|t_{n-1}, \dots, t_1), \quad (8.6a)$$

$$\beta_{k1}^{(n)}(t_n|t_n, \dots, t_1) = 0, \quad (8.6b)$$

with the right side of Eq. (8.6a) understood as zero for $n=0$.

The mean occupation number $N_k(t)$ of mode k as represented by Eq. (8.5a) is simply a statistical average of one-photon probabilities, where the averaging is taken over the times at which photons have been emitted previously into the other modes of the field. The decay of the one-photon amplitudes as governed by Eqs. (4.35), like that of the vacuum amplitudes, is fully and accurately represented by the addition of the imaginary term $-\frac{1}{2}i\kappa$ to the energy of the upper atomic state.

For times t near the initial time $t=0$, only the first term (i.e., the one for which $n=0$) in Eq. (8.5a) is appreciable, and the field spectrum is accurately represented by the one-photon amplitudes given by Eqs. (4.25). For large values of t , on the other hand, the quantity $N_k(t)$ approaches the asymptotic value $W_k t$, with W_k given by Eqs. (5.7), (5.12), and (5.11). [This follows from the fact that the quantities in Eqs. (8.5) obey Eqs. (5.7), (5.8), and (5.10).] Indeed, each term in Eq. (8.5a), for n large, is proportional to $W_k t$, and is in fact given by Eq. (7.13).

The difference between the forms assumed by the spectrum at small and at large values of the time t is due not merely to the averaging over initial times in Eq. (8.5a), but also to the initial condition in Eqs. (8.6). This condition, which is crucial to the accuracy of the construction in Eqs. (8.5), is a natural generalization of the initial condition (8.4): Both conditions state that the quan-

tum system of atom plus single field mode k , immediately after a photon has been emitted into one of the other field modes, is described by a state vector which is simply the product of the atomic lowering operator a times the pre-emission state vector.

Of particular importance is the fact that the amplitude for finding a photon in mode k is not completely destroyed by the emission process, but instead, a part of it (the part associated with the upper atomic state before the emission) survives and becomes an initial amplitude for the next stage of the process. The initial quantum state described by Eqs. (8.4) and (8.6) then is not simply a complex constant times the atom-field ground state $|0\rangle_a |0\rangle_k$ which corresponds to the solutions in Eqs. (4.24) and (4.25).

It is nevertheless possible, however, to express the spectral emission rate W_k in a form in which only the vacuum amplitudes $\psi_\mu(t)$ and one-photon amplitudes $\beta_{k\mu}(t)$ given by Eqs. (4.24) and (4.25) appear. To accomplish this, one may begin by evaluating the functions $\psi_\mu^{(n)}(t|t_n, \dots, t_1)$ and $\beta_{k\mu}^{(n)}(t|t_n, \dots, t_1)$ as the solutions to Eqs. (4.33) and (4.35), subject to the initial conditions (8.4) and (8.6). One finds the relations

$$\psi_\mu^{(n)}(t|t_n, \dots, t_1) = \psi_\mu(t; t_n) \psi_1^{(n-1)}(t_n|t_{n-1}, \dots, t_1), \quad (8.7a)$$

$$\begin{aligned} \beta_{k\mu}^{(n)}(t|t_n, \dots, t_1) &= \beta_{k\mu}(t; t_n) \psi_1^{(n-1)}(t_n|t_{n-1}, \dots, t_1) \\ &\quad + [e^{i\omega_k(t_n-t)} \psi_\mu(t; t_n) \\ &\quad \times \beta_{k1}^{(n-1)}(t_n|t_{n-1}, \dots, t_1)], \end{aligned} \quad (8.7b)$$

in which $\psi_\mu(t; t_n)$ and $\beta_{k\mu}(t; t_n)$ are just the solutions given by Eqs. (4.24) and (4.25), but with initial time taken as t_n rather than as zero. [For $n=0$, the solutions in Eqs. (8.7) are identical with those in Eqs. (4.24) and (4.25).]

By substituting Eqs. (8.7) into Eq. (8.5b), one finds an integral equation obeyed by the function ${}_\mu\langle A(t)|B_k(t)\rangle_\nu$ which may be used directly to evaluate the spectral emission rate in Eq. (5.7). With the aid of the identity

$$\begin{aligned} \frac{d}{dt} [|\beta_{k0}(t)|^2 + |\beta_{k1}(t)|^2] \\ = -\kappa |\beta_{k1}(t)|^2 + 2 \operatorname{Re} [\vec{\lambda} \cdot \vec{u}_k(0) \psi_1^*(t) \beta_{k0}(t)] \end{aligned} \quad (8.8)$$

obeyed by the functions appearing in Eqs. (4.35), one finds in this way the steady-state expression

$$W_k = W_k^{(1)} + \Delta W_k, \quad (8.9a)$$

where

$$W_k^{(1)} = \kappa \bar{\rho}_{11} \int_0^\infty |\beta_{k1}(t)|^2 \kappa dt, \quad (8.9b)$$

and

$$\Delta W_k = 2 \operatorname{Re} \left(\vec{\lambda} \cdot \vec{u}_k(0) \eta_k \int_0^\infty \psi_1^*(t) \psi_0(t) e^{-i\omega_k t} \kappa dt \right). \quad (8.9c)$$

The constant parameter η_k in Eq. (8.9c) is the steady-state value of ${}_\mu\langle A(t)|B_k(t)\rangle_\nu e^{i\omega t}$, and is found by an application of the integral equation mentioned above to be the solution to the equation

$$\eta_k = \int_0^\infty [\bar{\rho}_{11} \psi_1^*(t) \beta_{k1}(t) e^{i\omega t} + \eta_k |\psi_1(t)|^2 e^{i(\omega - \omega_k)t}] \kappa dt. \quad (8.9d)$$

The functions $\psi_\mu(t)$ and $\beta_{k\mu}(t)$ in these relations are just the elementary vacuum- and one-photon amplitudes given by Eqs. (4.24) and (4.25), respectively.

The value of W_k given by the solution in Eqs. (8.9) is of course identical to the one found in Eqs. (5.7) and (5.12) (and hence is identical to the value found in Ref. 5). The representation of W_k in Eqs. (8.9) is offered here as a means of clarifying the relationship between the full multiphoton spectrum and the one-photon amplitudes $\beta_{k\mu}(t)$, and in particular of showing how the former can be expressed in terms of the latter.

The quantity $W_k^{(1)}$ as given by Eq. (8.9b) is in a sense an improved one-photon approximation to the steady-state emission spectrum, representing, in effect, an appropriate averaging of the elementary one-photon solution over initial preparation times. The amplitude $\beta_{k1}(t)$, it may be recalled, is the joint amplitude for finding the atom in its excited state and for finding a photon in mode k at time t , given that at $t=0$ the system was prepared with the atom in its ground state and with no photons present. One may therefore think of $|\beta_{k1}(t)|^2 \kappa dt$ as the joint probability that in a system so prepared a photon will be present in mode k at time t , and that the system will then decay between times t and $t+dt$. If the decay process (which actually represents the effect of a spontaneous photon emission into other modes of the field) is thought of as destroying the system, the approximation in Eq. (8.9b) then represents the overall probability that a photon is emitted into mode k before the system is destroyed, multiplied by the constant rate $\kappa \bar{\rho}_{11}$ at which new systems are created.

The approximate nature of the picture just described consists in the fact that the new systems in it are created independently of already existing systems, and, in particular, with no photons present. [The nonzero initial amplitude in Eq. (8.6a) is represented wholly by the term ΔW_k in Eq. (8.9a).] The approximate value $W_k^{(1)}$ for the spectral emission rate may be shown to correspond

to the correct *total* emission rate, i.e., to obey the relation

$$\sum_k W_k^{(1)} = \sum_k W_k = \kappa \bar{\rho}_{11}, \quad (8.10)$$

and to correspond through Eq. (6.9) to the approximate atomic correlation function

$$\begin{aligned} \langle a^\dagger a(t) \rangle^{(1)} &= \text{Tr}[\bar{\rho} a^\dagger V^\dagger(t) a V(t)] \\ &= \bar{\rho}_{01} \psi_1(t) \psi_0^*(t) + \bar{\rho}_{11} \chi_1(t) \psi_0^*(t), \end{aligned} \quad (8.11)$$

which has been discussed by Notkin, Rautian, and Feoktistov.¹ Here, $V(t)$ is the simple time-development operator for the damped atom in the presence of a prescribed incident field, i.e., the solution to the equations

$$\begin{aligned} i \frac{dV(t)}{dt} &= [(\omega_{10} - \frac{1}{2} i \kappa) a^\dagger a - a^\dagger \bar{\lambda} \cdot \bar{\mathcal{E}}_c(0, t) \\ &\quad - a \bar{\lambda}^* \cdot \bar{\mathcal{E}}_c^*(0, t)] V(t) \end{aligned} \quad (8.12)$$

and $V(0) = 1$, while $\psi_\mu(t) = {}_a \langle \mu | V(t) | 0 \rangle_a$ as given by Eqs. (4.24) and $\chi_\mu(t) = {}_a \langle \mu | V(t) | 1 \rangle_a$ are the solutions to Eqs. (4.33) corresponding to the initial conditions $\psi_\mu(0) = \delta_{\mu 0}$ and $\chi_\mu(0) = \delta_{\mu 1}$, respectively.

[The approximate correlation function in Eq. (8.11) is in fact the leading term in a simple expansion for the exact correlation function as determined by the quantum fluctuation-regression theorem. The Eqs. (6.3) and (6.5)–(6.7) for the quantities $R_{\mu\nu}(t)$ defined by Eqs. (6.18) (for $t' = 0$), subject to the initial conditions obtained from Eqs. (6.18), lead to the integral equation

$$\begin{aligned} R_{\mu\nu}(t) &= \bar{\rho}_{01} \psi_\mu(t) \psi_\nu^*(t) + \bar{\rho}_{11} \chi_\mu(t) \psi_\nu^*(t) \\ &\quad + \int_0^t \psi_\mu(t; t_0) \psi_\nu^*(t; t_0) R_{11}(t_0) \kappa dt_0, \end{aligned} \quad (8.13)$$

which yields the full solution by iteration.^{23]}

For values of κ small compared to the other characteristic frequency parameters (Ω and $\Delta\omega$) of the problem, the spectrum consists of a superposition of sharply peaked functions, which the approximation $W_k^{(1)}$ may be shown to represent accurately in intensity but inaccurately in width. In particular, the coherent term proportional to $\delta(\omega - \omega_k)$ is not present in the one-photon approximation $W_k^{(1)}$, and is contained entirely within the term ΔW_k in Eq. (8.9a).

The δ -function component in the emission spectrum of course requires an infinite amount of time to become established, and therefore it should not be surprising to learn that it originates from the term [the second on the right side of Eq. (8.7b)] which represents the survival of the coherent-photon amplitude given by Eq. (8.6a) during the emission of photons into other modes of the field. The decaying one-photon amplitudes in Eqs. (4.25)

of course have no δ -function components, nor do the amplitudes for any fixed number n of photons, except, as in Eq. (7.13), in an asymptotic sense in the limit $n \rightarrow \infty$.

IX. COLLISIONAL RELAXATION IN A PURE-STATE FORMALISM

The model of atomic relaxation in which the atom is returned to its ground state at random times by collisions or by any abrupt, classically describable process, is important to understand both because of its practical importance and because it is equivalent, in a limiting sense to be described, to some of the familiar models employing injection schemes. The collisional case can be described solely in terms of pure-state amplitudes by a suitable modification of the method used in Sec. VIII to describe the radiative case.

In the limit in which the radiative decay rate κ is small compared to the collision rate κ_c , the effect of spontaneous photon emission on the atomic relaxation can be ignored, and a one-photon approximation becomes accurate. A hierarchy of pure-state functions may be introduced in this case to describe the system subject to the restriction that any fixed number n of collisions have occurred. The probability that these occur between the times t_j and $t_j + dt_j$ (where $0 \leq t_1 \cdots \leq t_n \leq t$) and at no other times between time $t_0 = 0$ and time t is just

$$e^{-\kappa_c(t-t_n)} \prod_{j=1}^n e^{-\kappa_c(t_j - t_{j-1})} \kappa_c dt_j = e^{-\kappa_c t} \prod_{j=1}^n \kappa_c dt_j. \quad (9.1)$$

A system which is described immediately before a collision by the pure-state wave function $(\psi_0, \psi_1, \beta_{k0}, \beta_{k1})$ must be described immediately after the collision in a way which takes into account the fact that, while the atom is put into its ground state by the collision (so that $\psi'_1 = \beta'_{k1} = 0$), the collision changes neither the state norm nor the probability $N_k = |\beta_{k0}|^2 + |\beta_{k1}|^2$ of finding a photon in mode k , nor indeed the expectation value of any radiation-field-mode operator. This means that one must describe the system immediately after the collision by means of *two* pure-state functions, one of which is obtained from the pre-collision *ground*-state amplitudes and has the unchanged value $(\psi_0, 0, \beta_{k0}, 0)$, while the other is obtained from the pre-collision *excited*-state amplitudes ψ_1 and β_{k1} and has the value $(\psi_1, 0, \beta_{k1}, 0)$ after the collision has put the atom into its ground state. These two post-collision state vectors, which originate from orthogonal parts of the pre-collision state vector, must be treated as distinct classical alternatives, and of course must not be

allowed to interfere coherently with one another. (The above discussion is proposed as a definition of an instantaneous classical transition mechanism.)

In accordance with the above remarks, the pure-state amplitude functions at any time t depend not only upon the (prior) times t_j at which collisions have occurred, but also upon a set of state indices μ_j which specify which branch of the pre-collision state vector is taken at each collision time to form the basis for the further time development of the system. One is thus led to introduce functions $\psi_\mu^{(n)}(t|t_n\mu_n, \dots, t_1\mu_1)$ and $\beta_{k\mu}^{(n)}(t|t_n\mu_n, \dots, t_1\mu_1)$ which obey the initial conditions

$$\psi_\mu^{(n)}(t_n|t_n\mu_n, \dots, t_1\mu_1) = \delta_{\mu_0}\psi_\mu^{(n-1)}(t_n|t_{n-1}\mu_{n-1}, \dots, t_1\mu_1), \quad (9.2a)$$

$$\beta_{k\mu}^{(n)}(t_n|t_n\mu_n, \dots, t_1\mu_1) = \delta_{\mu_0}\beta_{k\mu}^{(n-1)}(t_n|t_{n-1}\mu_{n-1}, \dots, t_1\mu_1), \quad (9.2b)$$

$$\langle \varphi_\mu^*(t)\varphi_\nu(t) \rangle \equiv \sum_{n=0}^{\infty} \sum_{\mu_n^* \dots \mu_1} \int_0^t \kappa_c dt_n \int_0^{t_n} \kappa_c dt_{n-1} \dots \int_0^{t_2} \kappa_c dt_1 e^{-\kappa_c t} \varphi_\mu^{(n)*}(t|t_n\mu_n, \dots, t_1\mu_1) \varphi_\nu^{(n)}(t|t_n\mu_n, \dots, t_1\mu_1), \quad (9.4)$$

where $\varphi = \psi$ or $\varphi = \beta_k$. In particular, the probability of finding a photon in mode k is given by the relation

$$N_k(t) = \sum_{\mu} \langle \beta_{k\mu}^*(t)\beta_{k\mu}(t) \rangle, \quad (9.5)$$

and has the time derivative

$$W_k = \frac{dN_k(t)}{dt} = \vec{\lambda} \cdot \vec{u}_k(0) \langle \psi_1^*(t)\beta_{k0}(t) \rangle + \text{c.c.} \quad (9.6)$$

The right side of Eq. (9.6) can be evaluated by making use of Eqs. (9.3) in Eq. (9.4), to obtain an integral equation obeyed by the function $\langle \psi_\mu^*(t)\beta_{k\nu}(t) \rangle$. With the aid of Eq. (8.8) (with $\kappa = 0$), one finds in this way the steady-state spectral-emission rate

$$W_k = W_k^{(1)} + \Delta W_k, \quad (9.7a)$$

where

$$W_k^{(1)} = \kappa_c \int_0^{\infty} \sum_{\mu} |\beta_{k\mu}^{ZD}(t)|^2 e^{-\kappa_c t} \kappa_c dt \quad (9.7b)$$

and

$$\Delta W_k = 2 \operatorname{Re} \left(\vec{\lambda} \cdot \vec{u}_k(0) \gamma_k \int_0^{\infty} \psi_1^{ZD*}(t)\psi_0^{ZD}(t) \times e^{-i\omega_k t - \kappa_c t} \kappa_c dt \right). \quad (9.7c)$$

The constant parameter γ_k in Eq. (9.7c) is the steady-state value of $\sum_{\mu} \langle \psi_\mu^*(t)\beta_{k\mu}(t) \rangle e^{i\omega t}$, and is found from the integral equation mentioned above to have the value

and undamped time-development equations, obtainable by putting $\kappa = 0$ in Eqs. (4.33) and (4.35). The solution to these equations, subject to the initial conditions (9.2), is

$$\psi_\mu^{(n)}(t|t_n\mu_n, \dots, t_1\mu_1) = \psi_\mu^{ZD}(t; t_n)\psi_{\mu_n}^{(n-1)}(t_n|t_{n-1}\mu_{n-1}, \dots, t_1\mu_1), \quad (9.3a)$$

$$\begin{aligned} \beta_{k\mu}^{(n)}(t|t_n\mu_n, \dots, t_1\mu_1) &= \beta_{k\mu}^{ZD}(t; t_n)\psi_{\mu_n}^{(n-1)}(t_n|t_{n-1}\mu_{n-1}, \dots, t_1\mu_1) \\ &+ e^{i\omega_k(t_n-t)}\psi_\mu^{ZD}(t; t_n)\beta_{k\mu_n}^{(n-1)}(t_n|t_{n-1}\mu_{n-1}, \dots, t_1\mu_1), \end{aligned} \quad (9.3b)$$

where the zero-damped functions $\psi_\mu^{ZD}(t; t_n)$ and $\beta_{k\mu}^{ZD}(t; t_n)$ are obtainable from the solutions (4.24) and (4.25) simply by setting $\kappa = 0$ and taking the initial time at $t = t_n$.

Statistical averages can now be constructed with the aid of the quantities [see Eq. (9.1)]

$$\gamma_k = \frac{\kappa_c + i(\omega_k - \omega)}{i(\omega_k - \omega)} \int_0^{\infty} \sum_{\mu} \psi_\mu^{ZD*}(t)\beta_{k\mu}^{ZD}(t) e^{i\omega t - \kappa_c t} \kappa_c dt. \quad (9.7d)$$

The functions $\psi_\mu^{ZD}(t)$ and $\beta_{k\mu}^{ZD}(t)$ in the above relations are just the elementary zero-damped relations for the vacuum- and one-photon amplitudes, respectively, corresponding to the initial condition (4.21), and are thus obtainable simply by putting $\kappa = 0$ in the solutions (4.24) and (4.25).

The solution for the spectral-emission rate given by Eqs. (9.7) agrees exactly with the (zero-temperature) result which has been obtained previously²⁴ with the aid of the quantum fluctuation-regression theorem. (The case of nonzero temperature can be treated by a straightforward generalization of the methods described above.)

The expression $W_k^{(1)}$ given by Eq. (9.7b) in effect represents a *one-collision* approximation to the spectral-emission rate. One may think of a system created in the atomic ground state at $t = 0$ as having probability $e^{-\kappa_c t} \kappa_c dt$ of being destroyed by a collision between the times t and $t + dt$, and one may thus describe $W_k^{(1)}$ as the probability that such a system will emit a photon into mode k before it is destroyed, multiplied by the creation rate κ_c of new systems. As in the analogous radiative approximation of Eq. (8.9b), the new systems are created here independently of the old systems, and without any photons present. [One may also interpret Eq. (9.7b) as representing κ_c

times the probability that a photon has been emitted into mode k since the last collision, calculated under the assumption that no photons were present at the time of the last collision.]

By making use of Eqs. (4.25) in Eq. (9.7b), one finds that the one-collision approximation to the spectral-emission rate has the value

$$W_k^{(1)} = |\vec{\lambda} \cdot \vec{u}_k(0)|^2 \bar{\rho}_{11} \times 2 \operatorname{Re} \left(\frac{p^2 + \frac{1}{2} p (\kappa_c - i \Delta \omega) + \frac{1}{2} (\Omega'^2 - i \kappa_c \Delta \omega)}{p(p + i \Omega')(p - i \Omega')} \right), \quad (9.8a)$$

where

$$p \equiv i(\omega_k - \omega) + \kappa_c, \quad (9.8b)$$

$$\bar{\rho}_{11} = \frac{1}{2} \Omega^2 / (\Omega'^2 + \kappa_c^2), \quad (9.8c)$$

and the remaining parameters are defined by Eqs. (4.7c), (4.17a), and (4.36g).

The approximation given by Eqs. (9.8) is identical to the (zero-temperature) result found by Newstein.² [Equation (39) of Ref. 2 contains a minor error on the right-hand side, which should be proportional to

$$\operatorname{Re} \left\{ \frac{1}{2} a_{++}(p_1) [1 + \tau^{-1} \langle \sigma \rangle a_{zz}(p_2)] - \tau^{-1} \langle \sigma \rangle a_{+z}(p_1) a_{-z}(p_2) \right\},$$

while $\langle \sigma \rangle$ should be the negative of the value given by Eq. (30).]

The approximation $W_k^{(1)}$ to the spectral emission rate, like the analogous approximation in the radiative case, leads to the correct total line strength [Eq. (8.10) is satisfied with $\bar{\rho}_{11}$ given by Eq. (9.8c).] Unlike the analogous radiative approximation, the one-collisional approximation is fully accurate in the limit of intense applied fields, yielding the correct widths as well as the correct intensities of the spectral lines in that limit.

In the general case, however, the one-collision approximation under discussion here, like the analogous radiative approximation, is limited in its accuracy by its failure to correlate the creation of new systems with the destruction of old systems, and in particular by its failure to include the effect of the nonzero photon amplitude [in Eq. (9.2b)] which survives the collisional process. The coherent-emission rate proportional to $\delta(\omega_k - \omega)$ is of course entirely absent in $W_k^{(1)}$, and is wholly contained within ΔW_k .

In the limit of vanishing collision rate κ_c , where one might expect the one-collision approximation to have a certain asymptotic validity, one finds that it leads to the spectral-emission rate

$$W_k^{(1)} = \frac{2\kappa_c D_0}{(\omega_k - \omega)^2 + \kappa_c^2} + \frac{2\kappa_c D_{\pm}}{(\omega_k - \omega + \Omega')^2 + \kappa_c^2} + \frac{2\kappa_c D_{-}}{(\omega_k - \omega - \Omega')^2 + \kappa_c^2}, \quad \kappa_c \rightarrow 0, \quad (9.9)$$

where

$$D_0 = |\vec{\lambda} \cdot \vec{u}_k(0)|^2 \frac{1}{4} \Omega^2 / \Omega'^2 \quad (9.10a)$$

and

$$D_{\pm} = |\vec{\lambda} \cdot \vec{u}_k(0)|^2 \frac{1}{8} \Omega^2 (\Omega' \pm \Delta \omega) / \Omega'^3 \quad (9.10b)$$

In the same limit, the full expression for W_k (obtainable from Eqs. (9.7) or from Ref. 24) has the value

$$W_k = B' 2\pi \delta(\omega_k - \omega) + \frac{2\kappa_c (D_0 - B')}{(\omega_k - \omega)^2 + \kappa_c^2} + \frac{2\kappa_c D_{+}}{(\omega_k - \omega + \Omega')^2 + \kappa_c^2} + \frac{2\kappa_c D_{-}}{(\omega_k - \omega - \Omega')^2 + \kappa_c^2}, \quad (\kappa_c \rightarrow 0), \quad (9.11a)$$

where

$$B' = |\vec{\lambda} \cdot \vec{u}_k(0)|^2 \frac{1}{4} \Omega^2 (\Delta \omega)^2 / \Omega'^4, \quad (9.11b)$$

and D_0 and D_{\pm} are given by Eqs. (9.10).

If one approximates the sharply peaked Lorentzian functions in the above expressions as δ functions, one finds that the one-collision spectrum and the full spectrum are indeed equal, each having the value

$$W_k^{(1)} \underset{\kappa_c \rightarrow 0}{\sim} W_k \underset{\kappa_c \rightarrow 0}{\sim} 2\pi [D_0 \delta(\omega_k - \omega) + D_{+} \delta(\omega_k - \omega + \Omega') + D_{-} \delta(\omega_k - \omega - \Omega')], \quad (9.12)$$

with the coefficients D_0 and D_{\pm} still given by Eqs. (9.10).

It should be emphasized that in the approximation of Eq. (9.12), the expression $D_0 \delta(\omega_k - \omega)$ for the central line in the spectrum actually represents the sum of two terms, one the genuine δ function in Eq. (9.11a) representing the *coherent* spectral-emission rate, the other a sharply peaked Lorentzian function centered at $\omega_k = \omega$, representing part of the *incoherent* spectrum.

Gush and Gush⁹ have evaluated scattering spectra for two-level systems subject to very intense fields, in particular intense enough to violate the conditions (3.19) on which the resonant approximation of this paper is based. They are able to evaluate the contributions made by harmonics of arbitrary order to the emission spectrum. No relaxation mechanism is introduced explicitly in their analysis, however. Instead, they assume the system to be prepared at $t=0$ in its ground state, and then obtain the spectral-emission rate by means of the familiar approximation

$$\sum_{\mu} |\beta_{k\mu}^{\text{ZD}}(t)|^2 \approx \sum_{\alpha} D_{\alpha} \frac{4\sin^2 \frac{1}{2}(\omega_k - \omega_{\alpha})t}{(\omega_k - \omega_{\alpha})^2} \quad (9.13a)$$

$$\rightarrow \left(\sum_{\alpha} D_{\alpha} 2\pi\delta(\omega_k - \omega_{\alpha}) \right) t, \quad (9.13b)$$

where the one-photon amplitudes $\beta_{k\mu}^{\text{ZD}}(t)$ are the solutions to undamped equations of motion.

By substituting the general form (9.13a) into Eq. (9.7b) and taking the limit $\kappa_c \rightarrow 0$, one finds exactly the form of the emission rate implied by Eq. (9.13b). Thus, it should come as no surprise to learn that for the limiting case in which the resonant approximation is valid [i.e., for incident fields not so intense as to violate the condition (3.19), though possibly intense enough to cause saturation], the spectrum found by Gush and Gush²⁵ is identical to the spectrum given by Eqs. (9.12) and (9.10).

The relaxation mechanism implicit in the transition-rate scheme of Eqs. (9.13) is thus identical in its implications to those of the collisional relaxation mechanism considered in an approximation in which the collision rate vanishes, and furthermore in which a coherent and an incoherent emission rate are incorporated within a single term. That the analysis of Gush and Gush does not describe the *radiative* case is perhaps most apparent in the limit of vanishing incident-field intensity, where (as they note) the spectrum they find does not reduce to the expected single coherent term at the incident-field frequency.

X. RADIATIVE RELAXATION IN THE MULTILEVEL ATOM

The case of a many-level atom in resonant interaction with the electromagnetic field can be analyzed in terms closely paralleling those for the two-level case. One may introduce the atomic-transition operators $a_{jk} = |k\rangle_a \langle j|$ which in the interaction picture have the time dependence $a'_{jk}(t) = a_{jk} e^{-i\omega_{jk}t}$, where $\omega_{jk} \equiv (E_j - E_k)/\hbar$. One then finds, by steps analogous to those used to reach Eq. (3.20), the relation

$$\vec{\mathcal{E}}'_R(0, \bar{t}) |t\rangle' = i \sum_{j,k} \vec{\lambda}_{jk}^* F_{jk}(\bar{t}-t) a'_{jk}(\bar{t}) |t\rangle', \quad \bar{t} \geq t, \quad (10.1)$$

in which $\vec{\lambda}_{jk} \equiv \langle j | \vec{\mu} | k \rangle_a$, and

$$F_{jk}(\bar{t}-t) \equiv \int_{-\infty}^{\bar{t}} dt' e^{i\omega_{jk}(\bar{t}-t')} \phi(\bar{t}-t'), \quad (10.2)$$

with ϕ still defined by Eq. (3.9). The value of $F_{jk}(\tau)$ at $\tau=0$ is found to be

$$F_{jk}(0) = \frac{1}{2} \kappa_{jk} / |\lambda_{jk}|^2, \quad (10.3)$$

where the frequency shift has again been ignored, and

$$\begin{aligned} \kappa_{jk} &= \hbar |\lambda_{jk}|^2 \omega_{jk}^3 / 3\pi c^3 \quad \text{for } E_j > E_k \\ &= 0 \quad \text{for } E_j < E_k. \end{aligned} \quad (10.4)$$

At $\bar{t}=t$, Eq. (10.1) reduces to the relation

$$\vec{\mathcal{E}}'_R(0) |t\rangle = \frac{1}{2} i \sum_{j,k} (\vec{\lambda}_{jk}^* \kappa_{jk} / |\lambda_{jk}|^2) a_{jk} |t\rangle. \quad (10.5)$$

By making use of Eq. (10.5) to evaluate the photon-absorption term in the Hamiltonian and retaining only secular terms, one finds that, in the absence of degeneracy, the effect of the term in question is to add the imaginary terms $-\frac{1}{2}i\kappa_j$ to the state energies, where

$$\kappa_j \equiv \sum_k \kappa_{jk}. \quad (10.6)$$

The effective (Schrödinger) Hamiltonian is then

$$\begin{aligned} \vec{H}(t) &= \vec{H}_0 + \vec{H}_1(t), \\ \vec{H}_0 &= \sum_j (E_j - \frac{1}{2}i\kappa_j) |j\rangle_a \langle j| + H_{0F}, \end{aligned} \quad (10.7)$$

$$\vec{H}_1(t) = -\hbar \sum_{\substack{j,k \\ (E_j > E_k)}} \vec{\lambda}_{jk}^* \cdot \vec{\mathcal{E}}'_R(t) a_{jk} - \vec{\mu} \cdot \vec{E}_c(0, t),$$

and the state vector $|t\rangle$ for the atom-field system obeys Eq. (3.22).

One may directly evaluate the time derivative of the reduced atomic density matrix element $\rho_{jk}(t) = \langle t | a_{jk} | t \rangle$ with the aid of Eqs. (3.22), (10.7), and (10.5). One finds that

$$\begin{aligned} \frac{d}{dt} \rho_{jk}(t) &= -(i\omega_{jk} + \frac{1}{2}\kappa_j + \frac{1}{2}\kappa_k) \rho_{jk}(t) \\ &+ i\hbar^{-1} [\vec{\mu} \cdot \vec{E}_c(0, t), \rho(t)]_{jk}, \quad j \neq k. \end{aligned} \quad (10.8)$$

The terms involving the radiation-field operators \mathcal{E}_R and \mathcal{E}_R^\dagger , which have been omitted from the right-hand side, are

$$\frac{1}{2} \sum_{j', j''} (\vec{\lambda}_{j''k} \cdot \vec{\lambda}_{j'j}^* \kappa_{j'j} / |\lambda_{j'j}|^2) \rho_{j'j''}(t) + (j \leftrightarrow k)^*. \quad (10.9)$$

Under the condition of well-separated resonance frequencies ω_{jk} , it is clear that for $j \neq k$, only the term for which $j' = j$ and $j'' = k$ can oscillate at the same frequency as ρ_{jk} . But this term vanishes, since $\lambda_{jj} = 0$.

For $j = k$, on the other hand, all of the terms for which $j' = j''$ have the same slow time dependence as the diagonal density matrix element $\rho_{jj}(t)$. The terms in (10.9) now make a nonvanishing contribution, and in place of Eq. (10.8) one finds the relation

$$\frac{d}{dt} \rho_{jj}(t) = -\kappa_j \rho_{jj}(t) + \sum_{j'} \rho_{j'j'}(t) \kappa_{j'} + i\hbar^{-1} [\vec{\mu} \cdot \vec{E}_c(0, t), \rho(t)]_{jj}. \quad (10.10)$$

The radiation-field-dependent terms in this case

then represent the spontaneous emission *to* the state in question.

The proof of the validity of the fluctuation-regression theorem for the many-level atom parallels quite closely the corresponding proof for the two-level atom, and will not be presented here.

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⁷(a) C. R. Stroud, Jr., Phys. Rev. A 3, 1044 (1971); (b) C. R. Stroud, Jr., in *Coherence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973), p. 537.

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^{10b}F. Schuda, C. R. Stroud, Jr., and M. Hercher, J. Phys. B 7, L198 (1974), have recently observed quite clearly the sideband structure produced in the emission spectrum by an intense applied field.

¹¹It should be noted that some processes, e.g., resonant pulse propagation, are well described by approximation schemes which include only that part of the scattered field which coherently modifies the form of the incident field. See, for example, L. Matulic and J. H. Eberly, Phys. Rev. A 6, 822 (1972), and references cited therein.

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¹⁴R. J. Glauber, Phys. Lett. 21, 650 (1966); C. L. Mehta and E. C. G. Sudarshan, Phys. Rev. Lett. 22, 574 (1966); B. R. Mollow, Phys. Rev. 162, 1256 (1967).

¹⁵Intensity-dependent effects on the radiative level shifts may arise, however, when the intensity of the incident field becomes great enough to violate the inequality $\Omega \ll \omega_{10}$, which is implicit in Eq. (3.19). See, for example, R. H. Lehberg, Phys. Lett. 33A, 501 (1970). For such intense incident fields, there will also be direct field-dependent contributions to the level shifts. See, for example, B. R. Mollow, Phys. Rev. A 5, 1827 (1972), Eq. (27).

¹⁶In Stroud's improved one-photon treatment in Ref. 7(b), the atom is allowed to have many levels and to emit (and reabsorb) a second *virtual* photon during transi-

tions to the other levels. The level shifts are found to be independent of the intensity of the incident field. Strictly speaking, a two-photon state originating entirely from transitions between the pair of resonantly coupled states themselves must also be allowed to exist, in a virtual sense, in Ref. 7(b). If it is not, then a spurious intensity-dependent frequency shift, similar to the one found in Ref. 7(a), will appear. The inclusion of the contributions made by other states of the atom (which have no postulated relationship with the original pair of states) does not by itself eliminate the intensity dependence of the frequency shift which arises in the strict one-photon approximation.

¹⁷Reference 5, Eq. (4.30).

¹⁸B. R. Mollow, Phys. Rev. A 8, 2684 (1973).

¹⁹B. R. Mollow, Phys. Rev. A 8, 1949 (1973).

²⁰The lower state $|0\rangle_a$ in Eqs. (4.33)–(4.36) is allowed to have a nonzero energy $\hbar\omega_0$, while all energy *differences* are assumed unchanged. Direct comparison with the other equations in this paper requires that the state amplitudes in the latter equations all be multiplied by $e^{-i\omega_0 t}$.

²¹Derivations of the optical Bloch equations similarly free from any atom-field statistical factorization assumption have recently been obtained in the Heisenberg picture by J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D 10, 3350 (1974), and by H. J. Kimble and L. Mandel, Phys. Rev. Lett. 34, 1485 (1975). The atomic cross-spectral correlation function implied by Kimble and Mandel's Eqs. (9) and (10) can be shown to be identical with the one obtained in Ref. 5 by means of the fluctuation-regression theorem.

²²M. E. Smithers and H. S. Freedhoff, J. Phys. B 7, L432 (1974). The conditional probabilities evaluated in Smithers' and Freedhoff's Eqs. (10) and (11) coincide exactly with the quantities $\rho_{11}^{(n)}(t)/P^{(n)}(t)$ and $\rho_{00}^{(n)}(t)/P^{(n)}(t)$ as determined by Eqs. (7.4) of this paper, in the limit in which $P^{(n)}(t)$ is given by Eq. (7.8).

²³The solution of Eq. (8.13) by iteration is easily carried out in the limit of intense applied fields, where the full solution for the field spectrum has sidebands with width $\frac{3}{2}\kappa$, broadened from the value $\frac{1}{2}\kappa$ which describes the first-order solution. A similar comparison between the full solution and the essentially one-photon solution has been noted by H. J. Carmichael and D. F. Walls, J. Phys. B 8, L77 (1975).

²⁴B. R. Mollow, Phys. Rev. A 2, 76 (1970).

²⁵Reference 9, Eq. (4.10).