

Theory of resonance fluorescence*

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The problem of the interaction between a two-level atom and a quantum electromagnetic field is treated without the use of perturbation theory, without introduction of classical fields or factorization conditions for the states, and without assumptions about loss of memory. The calculation is carried out in the Heisenberg picture, without mode decomposition, and the conclusions all refer to physically measurable quantities, such as the fluorescence detected in the far field of the atom. It is shown that in a coherent field of constant amplitude the system always settles down to a quasistationary state, and that the stationarity is a manifestation of the quantum fluctuations. A solution for the growth of the fluorescent light intensity is presented that holds for any coherent exciting field. The two-time correlation function and the spectral density of the fluorescence are calculated, and are found to agree in the long-time limit with earlier results of Mollow. The two-time intensity correlation function of the field is derived, which corresponds to measurable photoelectric pair correlations, and it is found that this reflects several quantum features of the field. It is shown that quantum fluctuations are manifest more explicitly in two-time correlations in the steady state than in transient effects, like spontaneous emission in the vacuum. The measurement of such correlations therefore presents an opportunity for further experimental tests of quantum electrodynamics.

I. INTRODUCTION

There has recently been greatly renewed interest in the details of the interaction between light and resonant atomic systems. This interest stems partly from the possibility for making precise experimental tests of quantum electrodynamics (QED) in the optical domain through resonant interactions, and partly from possibilities for the development of new practical techniques, such as atomic isotope separation. The theoretical problem of treating the microscopic interaction between an atom and a resonant electromagnetic field has received a great deal of attention since the early treatments of Weisskopf and Wigner¹ and Heitler and Ma,² and calculations based on a variety of different approaches have been published.³⁻¹⁹

However, from the point of view of testing QED by optical experiments through a comparison of its predictions with those of semiclassical theories, such as the neoclassical theory of Jaynes and his co-workers²⁰ or semiclassical source-field theories,²¹ many of these existing treatments turn out to be less than completely satisfactory. For example, calculations based on solving the Heisenberg equations of motion frequently assume that the external or driving field is classical.^{3-5, 9, 10} Most analyses in which the Schrödinger equation of motion is solved deal with a certain limited number of photons^{8, 14} or assume factorization conditions for the density operator of the combined system of atom plus field,⁵ although the recent treatment of Mollow¹⁶ appears to be free from these assumptions. Master-equation calculations

almost invariably introduce Markoffian assumptions about the process,^{6, 15, 18} to ensure that memory fades as the interaction proceeds. Assumptions that are roughly equivalent to the Markoffian assumption or to the factorization condition can also be found in recent Heisenberg-equation calculations.¹¹⁻¹³ Moreover, in the derivation of the correlation functions in resonance fluorescence, further assumptions regarding stationarity of the process are frequently introduced.^{5, 9, 13} If QED is to be tested, and if Markofficity and stationarity are indeed features of QED that are distinct from semiclassical theories, they should emerge as consequences of the equations of motion and not be inserted *a priori*. Finally, we mention that different treatments, even within QED, have led to disagreement about the spectral density of the fluorescent light in the presence of an exciting field.^{5, 6, 8, 11, 13-16, 44}

In the following we tackle the problem of the interaction between a two-level atom and an electromagnetic field near resonance within the framework of QED, without perturbation theory, without assumptions about Markofficity or stationarity or factorization of states, and without the introduction of a classical driving field. The method is based on solution of the Heisenberg equations of motion, and has similarities with some previous treatments,^{4, 10, 12, 13} except that we avoid some commonly made assumptions and do not make use of mode decompositions for the field. Instead, we prefer to express the results in terms of directly measurable quantities at some point in the far field of the atom, rather than in terms of

mode amplitudes, which are not strictly accessible to measurement. Although approximations are introduced in the course of the calculation, we are able to show explicitly that in the presence of a coherent exciting field the fluorescence becomes quasistationary in the wide sense, and that the achievement of a stationary state is a manifestation of the fluctuations of the quantum field. We present solutions for the time development of the fluorescent light intensity in the presence of an arbitrary coherent excitation. In the course of the calculation we observe, as was also noted by Agarwal⁶ and Ackerhalt *et al.*,¹² that certain transient effects—such as spontaneous emission in the vacuum—can be treated without explicit introduction of the quantum properties of the free electromagnetic field,²² because Maxwell's equations hold equally in QED and in semiclassical theory. However, the operator character of the free field enters in a more direct way in the proper treatment of certain two-time correlations of the fluorescent light in the presence of an exciting field. To some extent this is a reflection of the fact that, unlike transient effects, stationary effects necessarily imply the presence of a stochastic process, in this case the fluctuations of the quantum field. The steady-state properties of the fluorescence therefore reflect quantum features of the field more explicitly than do transient effects.

We also study the two-time intensity correlation function of the fluorescence for a monochromatic excitation with arbitrary detuning, and show that it has additional features characteristic of the quantum nature of the field. We are therefore led to the conclusion that measurements of certain two-time correlations—especially in the steady state—provide an opportunity for a more searching test of QED than do measurements of spontaneous emission of an atom undergoing a two-level transition in the vacuum.

We start by deriving the Heisenberg equations of motion and formally integrating them. We then apply them in Sec. III to the problem of spontaneous emission in the vacuum, and in Sec. IV to the calculation of the fluorescent light intensity in the presence of a coherent exciting field. We proceed to evaluate the two-time second-order correlation function and spectral density of the fluorescence in a weak field in Secs. V and VI, and show that they are obtainable directly by integration of the equations of motion and that they involve the free-field commutators. In Secs. VII and VIII we solve the same problem more generally for an exciting field of arbitrary amplitude and detuning; we make some comparison with experimental results and suggest a different kind of

measurement of the spectral density. In Sec. IX we obtain an expression for the two-time intensity correlation function that can be measured directly with a photoelectric detector and appears to be an attractive object for experimental study.

II. DERIVATION OF THE EQUATIONS OF MOTION

As we shall be interested in conditions of near resonance between the electromagnetic field and some atomic transition, in which two energy levels play a dominant role, we make the customary simplification of representing the atom by a spin- $\frac{1}{2}$ system with two energy eigenstates $|1\rangle$ and $|2\rangle$, separated by an energy interval $\hbar\omega_0$. This simplification robs the model of some physical attributes and does not give a proper account of the Lamb shift,²³ but it leaves most of the dynamics in which we shall be interested unchanged. The atom is then described by the three spin variables²⁴ $\hat{R}_1(t)$, $\hat{R}_2(t)$, and $\hat{R}_3(t)$ that obey the commutation rule

$$[\hat{R}_1(t), \hat{R}_m(t)] = i\epsilon_{1mn}\hat{R}_n(t), \quad (1)$$

which may be expressed in terms of lowering and raising operators $\hat{b}(t)$ and $\hat{b}^\dagger(t)$, with

$$\begin{aligned} \hat{R}_1(t) &= \frac{1}{2}[\hat{b}^\dagger(t) + \hat{b}(t)], & \hat{R}_2(t) &= (1/2i)[\hat{b}^\dagger(t) - \hat{b}(t)], \\ \hat{R}_3(t) &= \frac{1}{2}[\hat{b}^\dagger(t), \hat{b}(t)]. \end{aligned} \quad (2)$$

As is well known,²⁵ the atomic energy relative to a point midway between the two energy levels is then given by $\hbar\omega_0\hat{R}_3(t)$, and the transition dipole moment $\hat{\mu}_i(t)$ can be written $2\mu_i\hat{R}_1(t)$, where $\mu_i \equiv \langle 1|\hat{\mu}_i(0)|2\rangle$ is taken to be real, as for a $\Delta m = 0$ transition. The "electron velocity" of our model atom is then

$$\frac{1}{e} \frac{d\hat{\mu}_i}{dt} - - 2\frac{\omega_0}{e} \mu_i \hat{R}_2(t),$$

and we take the total energy of the quantum system consisting of the atom interacting with the electromagnetic field via an electric dipole interaction to be of the form

$$\begin{aligned} \hat{H} &= \frac{1}{2} \int \left(\epsilon_0 \hat{\mathbf{E}}^2(\vec{r}, t) + \frac{1}{\mu_0} \hat{\mathbf{B}}^2(\vec{r}, t) \right) d^3x \\ &+ \hbar\omega_0 \hat{R}_3(t) + 2\omega_0 \vec{\mu} \cdot \hat{\mathbf{A}}(0, t) \hat{R}_2(t), \end{aligned} \quad (3)$$

where the symbols have their usual meaning. We take the model atom to be located at the origin, and work in the Coulomb gauge, such that all the field vectors $\hat{E}_i(\vec{r}, t)$, $\hat{B}_i(\vec{r}, t)$, and $\hat{A}_i(\vec{r}, t)$ are transverse. The Heisenberg equations of motion for the atomic operator then take the form

$$\dot{\hat{R}}_1(t) = -\omega_0 \hat{R}_2(t) + 2(\omega_0/\hbar) \vec{\mu} \cdot \hat{\mathbf{A}}(0, t) \hat{R}_3(t), \quad (4)$$

$$\dot{\hat{R}}_2(t) = \omega_0 \hat{R}_1(t), \quad (5)$$

$$\dot{\hat{R}}_3(t) = -2(\omega_0/\hbar)\vec{\mu} \cdot \hat{\mathbf{A}}(0, t) \hat{R}_1(t), \quad (6)$$

while the corresponding equations for $\hat{E}_i(\vec{\mathbf{r}}, t)$ and $\hat{B}_i(\vec{\mathbf{r}}, t)$ are the Maxwell equations

$$\frac{d}{dt} \hat{\mathbf{B}}(\vec{\mathbf{r}}, t) = -\vec{\nabla} \times \hat{\mathbf{E}}(\vec{\mathbf{r}}, t), \quad (7)$$

$$\frac{d}{dt} \hat{\mathbf{E}}(\vec{\mathbf{r}}, t) = c^2 \vec{\nabla} \times \hat{\mathbf{B}}(\vec{\mathbf{r}}, t) - \frac{\hat{\mathbf{j}}(\vec{\mathbf{r}}, t)}{\epsilon_0}, \quad (8)$$

which can be formally obtained from the Hamiltonian (3) with the help of the well-known commutation relations for the field operators.²⁶ The transverse current $\hat{\mathbf{j}}_m(\vec{\mathbf{r}}, t)$ is proportional to the transverse δ^T function and is given by

$$\begin{aligned} \hat{\mathbf{j}}_m(\vec{\mathbf{r}}, t) &= -2\omega_0 \mu_i \delta^T_{i_m}(\vec{\mathbf{r}}) \hat{R}_2(t) \\ &= -2\omega_0 [\mu_m \delta^3(\vec{\mathbf{r}}) + (1/4\pi) \partial_m \mu_i \partial_i (1/r)] \hat{R}_2(t). \end{aligned} \quad (9)$$

The differentiation in Eq. (9) is to be interpreted in the sense of generalized function theory, and can be carried out with the help of the differential theorem given by Gel'fand and Shilov.²⁷ We then obtain the result

$$\begin{aligned} \hat{\mathbf{j}}_m(\vec{\mathbf{r}}, t) &= -2\omega_0 \left[\frac{2}{3} \mu_m \delta^3(\vec{\mathbf{r}}) - \mu_m / 4\pi r^3 \right. \\ &\quad \left. + 3(\vec{\mu} \cdot \vec{\mathbf{r}}) r_m / 4\pi r^5 \right] \hat{R}_2(t). \end{aligned} \quad (10)$$

The two Maxwell equations (7) and (8) can, of course, be combined in the inhomogeneous wave equation for the vector potential,

$$\vec{\nabla}^2 \hat{\mathbf{A}}(\vec{\mathbf{r}}, t) - \frac{1}{c^2} \frac{\partial^2 \hat{\mathbf{A}}(\vec{\mathbf{r}}, t)}{\partial t^2} = -\frac{1}{\epsilon_0 c^2} \hat{\mathbf{j}}(\vec{\mathbf{r}}, t), \quad (11)$$

whose solution may be expressed in terms of the retarded Green's function in the well-known form

$$\hat{\mathbf{A}}(\vec{\mathbf{r}}, t) = \frac{1}{4\pi \epsilon_0 c^2} \int \frac{\hat{\mathbf{j}}(\vec{\mathbf{r}}', t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c)}{|\vec{\mathbf{r}} - \vec{\mathbf{r}}'|} u(t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c) d^3 x' + \hat{\mathbf{A}}_{\text{free}}(\vec{\mathbf{r}}, t), \quad (12)$$

where $u(\tau)$ is the unit step function and $\hat{\mathbf{A}}_{\text{free}}(\vec{\mathbf{r}}, t)$ is the solution of the homogeneous or free-field equation that is supposed to characterize the field at time $t=0$. From Eqs. (10) and (12) we have

$$\begin{aligned} \hat{\mathbf{A}}(\vec{\mathbf{r}}, t) &= -\frac{\omega_0}{2\pi \epsilon_0 c^2} \left[\frac{2\vec{\mu}}{3} \int \frac{\delta^3(\vec{\mathbf{r}}') u(t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c)}{|\vec{\mathbf{r}} - \vec{\mathbf{r}}'|} \hat{R}_2(t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c) d^3 x' \right. \\ &\quad \left. + \frac{1}{4\pi} \int \frac{u(t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c)}{r'^3 |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|} \hat{R}_2(t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c) \left(-\vec{\mu} + \frac{3(\vec{\mu} \cdot \vec{\mathbf{r}}') \vec{\mathbf{r}}'}{r'^2} \right) d^3 x' \right] + \hat{\mathbf{A}}_{\text{free}}(\vec{\mathbf{r}}, t). \end{aligned} \quad (13)$$

Equation (13) allows us to calculate the transverse electromagnetic field vectors $\hat{E}_i(\vec{\mathbf{r}}, t)$ and $\hat{B}_i(\vec{\mathbf{r}}, t)$ at any space-time point through the relation

$$\hat{\mathbf{B}}(\vec{\mathbf{r}}, t) = \vec{\nabla} \times \hat{\mathbf{A}}(\vec{\mathbf{r}}, t), \quad (14)$$

together with the integral over time of Eq. (8). For the far field we find, with the help of Eq. (5), to the lowest order in $1/r$, and for $t > r/c$,

$$\begin{aligned} \hat{\mathbf{B}}(\vec{\mathbf{r}}, t) &= -\frac{\omega_0^2}{2\pi \epsilon_0 c^2} \frac{\vec{\mu} \times \vec{\mathbf{r}}}{c r^2} \hat{R}_1\left(t - \frac{r}{c}\right) \\ &\quad + \dots + \hat{\mathbf{B}}_{\text{free}}(\vec{\mathbf{r}}, t), \end{aligned} \quad (15)$$

$$\begin{aligned} \hat{\mathbf{E}}(\vec{\mathbf{r}}, t) &= \frac{\omega_0^2}{2\pi \epsilon_0 c^2} \left(\frac{\vec{\mu}}{r} - \frac{(\vec{\mu} \cdot \vec{\mathbf{r}}) \vec{\mathbf{r}}}{r^3} \right) \hat{R}_1\left(t - \frac{r}{c}\right) \\ &\quad + \dots + \hat{\mathbf{E}}_{\text{free}}(\vec{\mathbf{r}}, t), \end{aligned} \quad (16)$$

which are well-known expressions for the far field of an oscillating dipole,²⁸ if we interpret $-2\omega_0^2 \mu_i \hat{R}_1(t)$ as $d^2 \mu_i(t)/dt^2$.

We point out in passing that while $\hat{E}_{i\text{free}}(\vec{\mathbf{r}}, t)$, $\hat{B}_{i\text{free}}(\vec{\mathbf{r}}, t)$, and $\hat{A}_{i\text{free}}(\vec{\mathbf{r}}, t)$ are taken to be Hilbert-space operators, Eqs. (13), (15), and (16) follow directly from Maxwell's equations, when the expressions for the dipole moment and its derivatives are inserted.

In order to use Eqs. (4) and (6) we need to calculate $\mu_i \hat{A}_i(\vec{\mathbf{r}}, t)$ at the position of the atom $\vec{\mathbf{r}}=0$, but it is evident from Eq. (13) that the first integral on the right-hand side diverges as $r \rightarrow 0$ for our atomic model. The r dependence of $\hat{R}_2(t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c)$ can be removed with the help of a Taylor expansion about t , together with Eq. (5), and when $r=0$ the $u(t - |\vec{\mathbf{r}} - \vec{\mathbf{r}}'|/c)$ function can be replaced by unity everywhere under the first integral, except when $t=0$. Provided $t > 0$, Eq. (13) therefore leads to

$$\begin{aligned} \vec{\mu} \cdot \hat{\mathbf{A}}(0, t) = & \frac{\omega_0 \mu^2}{2\pi \epsilon_0 c^2} \left(\frac{2\omega_0}{3c} \hat{R}_1(t) - \lim_{r \rightarrow 0} \frac{2}{3} \int \frac{\delta^3(\vec{\mathbf{r}}')}{|\vec{\mathbf{r}} - \vec{\mathbf{r}}'|} d^3x' \hat{R}_2(t) \right. \\ & \left. + \frac{1}{4\pi} \int_0^{ct} dr' \int_0^\pi d\theta \int_0^{2\pi} d\phi \frac{R_2(t-r'/c)}{r'^2} \sin\theta(1-3\cos^2\theta) \right) + \vec{\mu} \cdot \hat{\mathbf{A}}_{\text{free}}(\vec{\mathbf{r}}, t), \end{aligned} \quad (17)$$

in which the integral over θ vanishes. The δ -function integral has the dimensions L^{-1} , but clearly diverges as $r \rightarrow 0$, partly because of the assumed point-dipole nature of the atom. This term leads to a frequency shift of the atomic resonance that we shall identify with the Lamb shift for a real atom.²³ On introducing two frequency parameters defined by

$$\beta \equiv \frac{1}{4\pi \epsilon_0} \frac{2\mu^2 \omega_0^3}{3\hbar c^3}, \quad \gamma \equiv \frac{1}{4\pi \epsilon_0} \frac{2\mu^2 \omega_0^2}{3\hbar c^2 l}, \quad (18)$$

where l is a length whose reciprocal represents the infinite integral, we can rewrite Eq. (17) in the compact form

$$\vec{\mu} \cdot \hat{\mathbf{A}}(0, t) = 2(\hbar/\omega_0)[\beta \hat{R}_1(t) - \gamma \hat{R}_2(t)] + \vec{\mu} \cdot \hat{\mathbf{A}}_{\text{free}}(0, t), \quad (19)$$

with the understanding that $t > 0$.

We now decompose the field and the atomic operators into positive and negative frequency parts, by using Eq. (2) for $\hat{R}_1(t)$ and $\hat{R}_2(t)$ and writing

$$\hat{A}_i(\vec{\mathbf{r}}, t) = \hat{A}_i^{(+)}(\vec{\mathbf{r}}, t) + \hat{A}_i^{(-)}(\vec{\mathbf{r}}, t), \quad (20)$$

where $\hat{A}_i^{(+)}(\vec{\mathbf{r}}, t)$ and $\hat{A}_i^{(-)}(\vec{\mathbf{r}}, t)$ are annihilation and creation operators. The decomposition (20) is most commonly carried out with the help of a mode decomposition, in which case the elements of $\hat{A}_i^{(+)}(\vec{\mathbf{r}}, t)$ and $\hat{A}_i^{(-)}(\vec{\mathbf{r}}, t)$ vary as $e^{-i\omega t}$ and $e^{i\omega t}$, respectively, for a noninteracting field. Without a mode decomposition the separation into positive and negative frequency parts is not unambiguous, especially for sufficiently short times. However, since the dominant contributions to terms involving $\hat{A}_i^{(+)}(\vec{\mathbf{r}}, t)$ and $\hat{A}_i^{(-)}(\vec{\mathbf{r}}, t)$ in a Fourier decomposition would come from certain optical frequencies close to the atomic resonance frequency ω_0 , we may safely make the separation into positive and negative frequency parts for all times t for which $\omega_0 t \gg 1$. This is not a severe restriction on t , as times satisfying the condition $\omega_0 t \gg 1$ can still be very short compared with the atomic lifetime or any other time in which the system changes appreciably. From Eq. (19) we then have, with the help of Eqs. (2) and (20), for times such that $\omega_0 t \gg 1$,

$$\begin{aligned} \vec{\mu} \cdot \hat{\mathbf{A}}^{(+)}(0, t) &= (\hbar/\omega_0)(\beta - i\gamma) \hat{b}(t) + \vec{\mu} \cdot \hat{\mathbf{A}}_{\text{free}}^{(+)}(0, t), \\ \vec{\mu} \cdot \hat{\mathbf{A}}^{(-)}(0, t) &= (\hbar/\omega_0)(\beta + i\gamma) \hat{b}^\dagger(t) + \vec{\mu} \cdot \hat{\mathbf{A}}_{\text{free}}^{(-)}(0, t). \end{aligned} \quad (21)$$

These equations are similar to ones obtained by Ackerhalt *et al.*¹² and express the total field as the sum of contributions from the source and from the vacuum or free field. They were previously obtained by a mode decomposition together with a Markoffian type of approximation, neither of which was used here. The mode decomposition allowed Ackerhalt *et al.*¹² to arrive at a more explicit (but still infinite) expression for the Lamb shift γ . However, by working with the undecomposed fields, we shall not lose sight of the fact that the analysis so far lies within the domain of Maxwell's equations, and we shall more easily be able to recognize the point at which explicitly quantum-mechanical features of the field enter.

We now follow the procedure of Ackerhalt *et al.*¹² of using Eqs. (21) to substitute for $\mu_i \hat{A}_i(0, t)$ in Eqs. (4) and (6), after expressing the operator products $\mu_i \hat{A}_i(0, t) \hat{R}_3(t)$ and $\mu_i \hat{A}_i(0, t) \hat{R}_1(t)$ in normal order. Although $\mu_i \hat{A}_i(0, t)$ commutes with all atomic operators at the same time, so that any operator ordering is allowed, the ease with which terms can be evaluated, and the interpretation of each term, depend on the order, as has recently been discussed.²⁹ On decomposing $\hat{R}_1(t)$ and $\hat{R}_2(t)$, we then obtain the two coupled differential equations

$$\begin{aligned} \dot{\hat{b}}(t) &= (-i\omega_0 - \beta + i\gamma) \hat{b}(t) - (\beta + i\gamma) \hat{b}^\dagger(t) \\ &\quad + 2(\omega_0/\hbar)[\hat{R}_3(t) \vec{\mu} \cdot \hat{\mathbf{A}}_{\text{free}}^{(+)}(0, t) + \text{H.c.}], \\ \dot{\hat{R}}_3(t) &= -2\beta \hat{R}_3(t) + \frac{1}{2} \\ &\quad - (\omega_0/\hbar)\{[\hat{b}(t) + \hat{b}^\dagger(t)] \vec{\mu} \cdot \hat{\mathbf{A}}_{\text{free}}^{(+)}(\vec{\mathbf{r}}, t) + \text{H.c.}\}. \end{aligned} \quad (22)$$

At this point it is convenient to introduce slowly varying dynamical variables that are free from the rapid oscillations at optical frequencies characterizing $\hat{b}(t)$, $\hat{A}_{\text{free}}^{(+)}(\vec{\mathbf{r}}, t)$, etc. They are defined by

$$\begin{aligned} \hat{b}_S(t) &\equiv \hat{b}(t) e^{i\omega_0 t}, \quad \hat{b}_S^\dagger(t) \equiv \hat{b}^\dagger(t) e^{-i\omega_0 t}, \\ \hat{\mathbf{A}}_S^{(+)}(\vec{\mathbf{r}}, t) &\equiv \hat{\mathbf{A}}_{\text{free}}^{(+)}(\vec{\mathbf{r}}, t) e^{i\omega_0 t}, \quad \hat{\mathbf{A}}_S^{(-)}(\vec{\mathbf{r}}, t) \equiv \hat{\mathbf{A}}_{\text{free}}^{(-)}(\vec{\mathbf{r}}, t) e^{-i\omega_0 t}. \end{aligned} \quad (24)$$

(25)

In terms of these new variables Eqs. (22) and (23) become

$$\begin{aligned} \dot{\hat{b}}_S(t) &= (-\beta + i\gamma)\hat{b}_S(t) - (\beta + i\gamma)\hat{b}_S^\dagger(t)e^{2i\omega_0 t} \\ &+ 2(\omega_0/\hbar)[\hat{R}_3(t)\vec{\mu} \cdot \hat{A}_S^{(+)}(0, t) \\ &+ \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t)\hat{R}_3(t)e^{2i\omega_0 t}], \quad (26) \\ \dot{\hat{R}}_3(t) &= -2\beta[\hat{R}_3(t) + \frac{1}{2}] \\ &- (\omega_0/\hbar)[\hat{b}_S^\dagger(t)\vec{\mu} \cdot \hat{A}_S^{(+)}(0, t) \\ &+ \hat{b}_S(t)\vec{\mu} \cdot \hat{A}_S^{(-)}(0, t)e^{-2i\omega_0 t} + \text{H.c.}], \quad (27) \end{aligned}$$

in which we recognize certain terms oscillating at twice the optical frequency. When the equations are integrated over any measurable time interval, these oscillatory terms make a negligible contribution and, after discarding them, we obtain

$$\begin{aligned} \hat{b}_S(t) &= \hat{b}_S(0)e^{(-\beta+i\gamma)t} + \frac{2\omega_0}{\hbar}e^{(-\beta+i\gamma)t} \\ &\times \int_0^t dt' \hat{R}_3(t')\vec{\mu} \cdot \hat{A}_S^{(+)}(0, t')e^{(\beta-i\gamma)t'}, \quad (28) \end{aligned}$$

$$\begin{aligned} \hat{R}_3(t) + \frac{1}{2} &= [\hat{R}_3(0) + \frac{1}{2}]e^{-2\beta t} \\ &- \frac{\omega_0}{\hbar}e^{-2\beta t} \int_0^t dt' [\hat{b}_S^\dagger(t')\vec{\mu} \cdot \hat{A}_S^{(+)}(0, t') \\ &+ \text{H.c.}]e^{2\beta t'}. \quad (29) \end{aligned}$$

This simplification is a form of the rotating-wave approximation. On substituting for $\hat{b}_S(t)$ from Eq. (28) in Eq. (29), and for $\hat{R}_3(t)$ from Eq. (29) in Eq. (28), we finally arrive at the following two integral equations for $\hat{b}_S(t)$ and $\hat{R}_3(t)$:

$$\begin{aligned} \hat{b}_S(t) &= \hat{b}_S(0)e^{(-\beta+i\gamma)t} + \frac{2\omega_0}{\hbar}e^{(-\beta+i\gamma)t}[\hat{R}_3(0) + \frac{1}{2}] \int_0^t dt' \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t')e^{(\beta-i\gamma)t'} \\ &- \frac{\omega_0}{\hbar}e^{(-\beta+i\gamma)t} \int_0^t dt' \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t')e^{(\beta-i\gamma)t'} \\ &- 2\left(\frac{\omega_0}{\hbar}\right)^2 e^{(-\beta+i\gamma)t} \int_0^t dt' e^{(-\beta-i\gamma)t'} \int_0^{t'} dt'' [\hat{b}_S^\dagger(t'')\vec{\mu} \cdot \hat{A}_S^{(+)}(0, t'') + \text{H.c.}]e^{2\beta t''} \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t'), \quad (30) \end{aligned}$$

$$\begin{aligned} \hat{R}_3(t) + \frac{1}{2} &= [\hat{R}_3(0) + \frac{1}{2}]e^{-2\beta t} - \frac{\omega_0}{\hbar} \left(\hat{b}_S^\dagger(0)e^{-2\beta t} \int_0^t dt' \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t')e^{(\beta-i\gamma)t'} + \text{H.c.} \right) \\ &- 2\left(\frac{\omega_0}{\hbar}\right)^2 e^{-2\beta t} \left(\int_0^t dt' e^{(\beta-i\gamma)t'} \int_0^{t'} dt'' \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t'')\hat{R}_3(t'')e^{(\beta+i\gamma)t''} \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t') + \text{H.c.} \right). \quad (31) \end{aligned}$$

III. SPONTANEOUS EMISSION

A somewhat trivial application of these equations is the problem of the spontaneous decay of an excited atomic state in the presence of a vacuum field $|\{0\}\rangle$. If we calculate the expectation values of all terms in Eqs. (30) and (31) for an arbitrary initial atomic state $|\psi\rangle$, we see immediately that since $\hat{A}_{S_i}^{(+)}(\vec{r}, t)|\{0\}\rangle = 0 = \langle\{0\}|\hat{A}_{S_i}^{(-)}(\vec{r}, t)$, all terms except the first term in each equation vanish, and we obtain the well-known results

$$\langle\hat{R}_3(t)\rangle + \frac{1}{2} = [\langle\hat{R}_3(0)\rangle + \frac{1}{2}]e^{-2\beta t}, \quad (32)$$

$$\langle\hat{b}_S(t)\rangle = \langle\hat{b}_S(0)\rangle e^{(-\beta+i\gamma)t}. \quad (33)$$

This shows that the atomic excitation decays exponentially with lifetime $1/2\beta$, and that the dipole moment oscillates at a frequency that is shifted from the "natural" frequency ω_0 by an amount γ that can be identified with the Lamb shift.

These results have, of course, been obtained

many times in different ways,^{1-7, 10, 12} and they are regarded as characteristic consequences of QED. It has even been argued that the process of spontaneous emission is a direct manifestation of the vacuum fluctuations of QED. However, the treatment given by Ackerhalt and Eberly¹² shows that the process may also be considered as a consequence of radiation reaction (see also Jaynes and Cummings²⁰), and the essential equivalence of these two points of view has recently been emphasized.²⁹ We see that when the equations of motion are written in normal order quantum features of the free electromagnetic field do not enter explicitly in the derivation of Eqs. (32) and (33), for the results would remain unchanged if $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ were treated as c numbers and were put equal to zero. We recall that the field was obtained from Maxwell's equations, and the only quantum property that we introduced was the commuting of the total field at the position of the atom with atomic operators at the same time. Actually,

it is possible to give rough arguments based on the transversality of $\hat{A}_i(\vec{r}, t)$ at the position of the atom, together with the equation of motion (5), that also lead to this commuting property, without reference to the operator character of the free field in QED. To be sure, the free field has to be quantized if the theory is to be self-consistent,¹⁰ but if we did not concern ourselves with questions of internal consistency and proceeded to treat $\hat{A}_{Si}^{(+)}(0, t)$ and $\hat{A}_{Si}^{(-)}(0, t)$ as c numbers, we would still arrive at Eqs. (32) and (33). We could then claim that the results were obtained via a semiclassical theory in which the two-level atom is the only quantum system, and its two-dimensional Hilbert space spans all available degrees of freedom. In such a treatment the total field given by Eqs. (12) or (19) would still not be a c number because of the quantum nature of the source.^{21, 30, 31} Still insofar as Eqs. (32) and (33) are derivable without explicit introduction of the operator character of the free field, it does seem that measurement of spontaneous emission in a two-level transition, under different conditions of excitation,³² does not represent the best test of the quantum nature of the field. We shall see that the phenomenon of resonance fluorescence offers an opportunity for a more convincing test.

IV. FLUORESCENCE IN THE PRESENCE OF AN EXCITING FIELD

We now turn to the problem of determining the response of the system when an electromagnetic excitation is present, in some given quantum state. As Eqs. (30) and (31) are written in normally ordered form, they lend themselves to the immediate calculation of the atomic expectation

values $\langle \hat{b}_s(t) \rangle$ and $\langle \hat{R}_3(t) \rangle$ when the exciting field is in a coherent state $|\{v\}\rangle$. This state is the right-hand eigenstate of $\hat{A}_{Si}^{(+)}(0, t)$ and the left-hand eigenstate of $\hat{A}_{Si}^{(-)}(0, t)$,³³ with eigenvalues given by

$$\hat{A}_S^{(+)}(0, t)|\{v\}\rangle = \vec{V}(t)|\{v\}\rangle, \quad \langle\{v\}|\hat{A}_S^{(-)}(0, t) = \vec{V}^*(t)\langle\{v\}|, \quad (34)$$

where $\vec{V}(t)$ is an arbitrary function of time that is determined by the set of complex mode amplitudes $\{v\}$. Then all field operators $\hat{A}_{Si}^{(+)}(0, t)$ and $\hat{A}_{Si}^{(-)}(0, t)$ in Eqs. (30) and (31) may be replaced by their right- and left-hand eigenvalues when expectation values are calculated.

The choice of the coherent state for the electromagnetic field has other advantages, besides simplifying the calculations. In the first place, it is a good approximation to the state characterizing a laser beam, and therefore corresponds closely to the experimental situation in which atoms are excited by a laser beam. With the recent development of tunable dye lasers this is likely to be the preferred method for making experimental tests, and several such experiments have already been reported.³⁴ Secondly, the coherent state is the quantum state that comes closest to the traditional classical characterization of the electromagnetic field,³³ and is therefore well suited for making comparison with semiclassical calculations. Thirdly, there is a sense in which any quantum state of the field may be regarded as a mixture of coherent states with some generalized weighting function,^{33, 35} so that the calculation of expectation values for a coherent state is a step in the treatment of the more general situation.

With the help of Eqs. (34) we then obtain from Eq. (31)

$$\begin{aligned} \langle \hat{R}_3(t) \rangle + \frac{1}{2} = & [\langle \hat{R}_3(0) \rangle + \frac{1}{2}] e^{-2\beta t} - \frac{\omega_0}{\hbar} e^{-2\beta t} \left(\langle \hat{b}_S^\dagger(0) \rangle \int_0^t dt' \vec{\mu} \cdot \vec{V}(t') e^{(\beta-i\gamma)t'} + \text{c.c.} \right) \\ & - 2 \left(\frac{\omega_0}{\hbar} \right)^2 e^{-2\beta t} \left(\int_0^t dt' e^{(\beta-i\gamma)t'} \vec{\mu} \cdot \vec{V}(t') \int_0^{t'} dt'' \vec{\mu} \cdot \vec{V}^*(t'') e^{(\beta+i\gamma)t''} \langle \hat{R}_3(t'') \rangle + \text{c.c.} \right), \end{aligned} \quad (35)$$

which is an integral equation for $\langle \hat{R}_3(t) \rangle$ of the Volterra type. If we integrate by parts we may express it in the form

$$\langle \hat{R}_3(t) \rangle = y(t) + \int_0^t K(t, t') \langle \hat{R}_3(t') \rangle dt', \quad (36)$$

in which the function $y(t)$ and the kernel $K(t, t')$ are given by

$$\begin{aligned} y(t) = & -\frac{1}{2} + [\langle \hat{R}_3(0) \rangle + \frac{1}{2}] e^{-2\beta t} \\ & - \frac{\omega_0}{\hbar} e^{-2\beta t} \left(\langle \hat{b}_S^\dagger(0) \rangle \int_0^t dt' \vec{\mu} \cdot \vec{V}(t') e^{(\beta-i\gamma)t'} + \text{c.c.} \right), \end{aligned} \quad (37)$$

$$\begin{aligned} K(t, t') = & -2 \left(\frac{\omega_0}{\hbar} \right)^2 e^{-2\beta t} \vec{\mu} \cdot \vec{V}^*(t') e^{(\beta+i\gamma)t'} \\ & \times \int_0^{t'} dt'' e^{(\beta-i\gamma)t''} \vec{\mu} \cdot \vec{V}(t'') + \text{c.c.} \end{aligned} \quad (38)$$

If $\vec{V}(t)$ is bounded, as it must always be in practice, the equation can always be solved by successive substitution in the form of an absolutely and uniformly convergent series,³⁶

$$\begin{aligned} \langle \hat{R}_3(t) \rangle = & y(t) + \int_0^t dt' K(t, t') y(t') \\ & + \int_0^t dt' \int_0^{t'} dt'' K(t, t') K(t', t'') y(t'') + \dots \end{aligned} \quad (39)$$

This solution holds for an arbitrary initial state of the atom and for an arbitrary coherent state of the field, whether in the form of a pulse or continuous, and it was obtained within the framework of QED. However, once again we point out that no explicitly quantum-mechanical features of the free field have entered in the derivation, which would remain unchanged if the operators $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ were replaced by the complex c numbers $\vec{V}(t)$ and $\vec{V}^*(t)$ from the beginning, provided the total field at $\vec{r}=0$ commutes with atomic operators at the same time.

The quantity $\langle \hat{R}_3(t) \rangle + \frac{1}{2}$ has an important interpretation in terms of directly measurable quantities, besides being the energy expectation value of the atom in units of $\hbar\omega_0$. The fluorescent light intensity $\langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t) \rangle$ radiated by the atom and detected by a photoelectric detector at \vec{r}, t in the far field may be expressed with the help of Eq. (16), after decomposition into positive and negative frequency parts, in the form

$$\begin{aligned} \langle \hat{E}^{(-)}(\vec{r}, t) \cdot \hat{E}^{(+)}(\vec{r}, t) \rangle = & \left(\frac{\omega_0^2}{4\pi\epsilon_0 c^2} \right)^2 \left(\frac{\mu^2}{r^2} - \frac{(\vec{\mu} \cdot \vec{r})^2}{r^4} \right) \\ & \times \left\langle \hat{b}_s^+ \left(t - \frac{r}{c} \right) \hat{b}_s \left(t - \frac{r}{c} \right) \right\rangle \\ = & \left(\frac{\omega_0^2 \mu \sin\psi}{4\pi\epsilon_0 c^2 r} \right)^2 \left[\langle \hat{R}_3 \left(t - \frac{r}{c} \right) \rangle + \frac{1}{2} \right], \\ & t \geq \frac{r}{c}, \end{aligned} \quad (40)$$

provided the space-time point \vec{r}, t is chosen so that $\hat{E}_{i\text{free}}^{(+)}(\vec{r}, t) | \{v\} \rangle = 0$, and the exciting field vanishes there. ψ is the angle between $\vec{\mu}$ and \vec{r} . It follows that $\langle \hat{R}_3(t) \rangle + \frac{1}{2}$ is a measure of the light intensity in the far field, after due allowance is made for retardation.

Certain general features of the solution follow from Eq. (35). If the excitation eventually dies out, so that $\vec{V}(t) \rightarrow 0$ as $t \rightarrow \infty$, then, since $\langle \hat{R}_3(t) \rangle$ is bounded, the exponential factors $e^{-2\beta t}$ cause all terms on the right-hand side of Eq. (35) to vanish as $t \rightarrow \infty$, and therefore $\langle \hat{R}_3(t) \rangle \rightarrow -\frac{1}{2}$. The atom must therefore end up in the ground state. On the

other hand, if the excitation $\vec{V}(t)$ reaches a steady amplitude eventually, which we take to be of the form (expressed in terms of slowly varying variables)

$$\vec{V}(t) = \vec{\epsilon} \alpha e^{i(\omega_0 - \omega_1)t + i\phi}, \quad (41)$$

where α, ϕ are real, $\vec{\epsilon}$ is a unit vector, and the frequency ω_1 of the exciting light does not necessarily coincide with the atomic frequency ω_0 , then the first two terms on the right-hand side of Eq. (35) still die out as $t \rightarrow \infty$, but the third does not. $\langle \hat{R}_3(t) \rangle + \frac{1}{2}$ then tends to a nonzero steady-state value. This value is easily found from Eq. (35), as the factors $e^{\beta t'}$ and $e^{\beta t''}$ in the integrand cause the steady-state value of $\langle \hat{R}_3(t) \rangle$ to dominate the integral as $t \rightarrow \infty$. We then obtain from Eqs. (35) and (41)

$$\langle \hat{R}_3(t) \rangle + \frac{1}{2} \rightarrow -2 \left(\frac{\omega_0 \vec{\mu} \cdot \vec{\epsilon} \alpha}{\hbar} \right)^2 \frac{\langle \hat{R}_3(t) \rangle}{2\beta[\beta + i(\gamma + \omega_1 - \omega_0)]} + \text{c.c.}$$

or

$$\langle \hat{R}_3(t) \rangle + \frac{1}{2} \rightarrow \frac{\frac{1}{4}\Omega^2}{\frac{1}{2}\Omega^2 + \beta^2 + (\gamma + \omega_1 - \omega_0)^2}, \quad (42)$$

where Ω is the Rabi frequency $2\omega_0 \vec{\mu} \cdot \vec{\epsilon} \alpha / \hbar$.^{37, 25} This equation expresses the steady-state fluorescent light intensity in terms of the strength of the exciting field Ω and of the detuning $\omega_1 - \omega_0$. The expression is known as the absorption spectrum, and was long ago derived by semiclassical methods, including solution of the semiclassical Bloch equations.^{38, 25} It has recently been measured for a number of atoms with the help of tunable dye lasers.³⁹

We may use Eqs. (41) and (42) in Eq. (28) to evaluate the solution for $\langle \hat{b}_s(t) \rangle$ as $t \rightarrow \infty$. We then obtain

$$\begin{aligned} \langle \hat{b}_s(t) \rangle = & \frac{2\omega_0}{\hbar} e^{(-\beta+i\gamma)t} \langle \hat{R}_3(\infty) \rangle \\ & \times \int_0^t dt' \vec{\mu} \cdot \vec{V}(t') e^{(\beta-i\gamma)t'} \\ = & \frac{-\frac{1}{2}\Omega[\beta + i(\gamma + \omega_1 - \omega_0)]}{\frac{1}{2}\Omega^2 + \beta^2 + (\gamma + \omega_1 - \omega_0)^2} e^{i(\omega_0 - \omega_1)t + i\phi}, \end{aligned} \quad (43)$$

which shows that the induced dipole moment continues to oscillate in time at the driving frequency, with an amplitude that increases with the exciting field strength (measured by the Rabi frequency Ω) for small Ω , but decreases inversely with Ω for large Ω . The oscillation amplitude is greatest when

$$\Omega^2 = 2\beta^2 + 2(\gamma + \omega_1 - \omega_0)^2. \quad (44)$$

The solution given by Eq. (39) simplifies considerably in the important special case in which the exciting field $\vec{V}(t)$ has a constant amplitude,

so that Eq. (41) holds for all times $t > 0$. In that case we find from Eqs. (37) and (38)

$$y(t) = -\frac{1}{2} + [\langle \hat{R}_3(0) \rangle + \frac{1}{2}] e^{-2\beta t} - \frac{1}{2} \frac{\Omega}{\beta} \left[\langle \hat{b}_S^\dagger(0) \rangle e^{i\phi} \frac{e^{-\beta(1+i\theta)t} - e^{-2\beta t}}{1-i\theta} + \text{c.c.} \right], \quad (45)$$

$$K(t, t') = \frac{-\Omega^2}{\beta(1+\theta^2)} \times \{ e^{-\beta(t-t')} [\cos\beta\theta(t-t') + \theta \sin\beta\theta(t-t')] - e^{-2\beta(t-t')} \}, \quad (46)$$

$$\theta \equiv (\gamma + \omega_1 - \omega_0)/\beta.$$

We see that the kernel $K(t, t')$ is a function only of the difference $t - t'$, and may be written in the form

$$K(t, t') \equiv (\Omega^2/\beta^2) H(t-t') \equiv \lambda H(t-t'), \quad (47)$$

where $\lambda \equiv \Omega^2/\beta^2$ depends on the strength of the excitation and $H(t)$ does not. The integral equation (36) can now be solved in closed form by Laplace transformation. Thus if

$$\int_0^\infty H(t) e^{-pt} dt \equiv \bar{H}(p),$$

$$\int_0^\infty y(t) e^{-pt} dt \equiv \bar{y}(p), \quad (48)$$

$$\int_0^\infty \langle \hat{R}_3(t) \rangle e^{-pt} dt \equiv \bar{R}_3(p),$$

we have from Eq. (36)

$$\bar{R}_3(p) = \bar{y}(p) + \lambda \bar{H}(p) \bar{R}_3(p)$$

or

$$\bar{R}_3(p) = \bar{y}(p) / [1 - \lambda \bar{H}(p)]. \quad (49)$$

By inverse Laplace transformation we then have

$$\langle \hat{R}_3(t) \rangle = \frac{1}{2\pi i} \int_{-i\infty+\kappa}^{i\infty+\kappa} \frac{\bar{y}(p)}{1 - \lambda \bar{H}(p)} e^{pt} dp, \quad t > 0, \quad (50)$$

where κ is a constant chosen so that all singularities of the integrand lie to the left-hand side of the line $\text{Re } p = \kappa$ in the complex p plane. From Eqs. (45)–(48) we readily find

$$\bar{y}(p) = -\frac{1}{2p} + \frac{\langle \hat{R}_3(0) \rangle + \frac{1}{2}}{p+2\beta} - \frac{1}{2} \frac{\Omega}{\beta} \left[\frac{\langle \hat{b}_S^\dagger(0) \rangle e^{i\phi}}{1-i\theta} \left(\frac{1}{p+\beta(1+i\theta)} - \frac{1}{p+2\beta} \right) + \frac{\langle \hat{b}_S(0) \rangle}{1+i\theta} \left(\frac{1}{p+\beta(1-i\theta)} - \frac{1}{p+2\beta} \right) e^{-i\phi} \right], \quad (51)$$

$$\bar{H}(p) = -\beta^2(\beta+p)/(2\beta+p)[(\beta+p)^2 + \beta^2\theta^2], \quad (52)$$

and application of the Cauchy residue theorem to the integral in (50) then leads to the solution

$$\langle \hat{R}_3(t) \rangle + \frac{1}{2} = \frac{\frac{1}{2}\lambda}{\frac{1}{2}\lambda + 1 + \theta^2} - \sum_{i=1}^3 \frac{(2\beta+p_i)[(\beta+p_i)^2 + \beta^2\theta^2] e^{p_i t}}{2(p_i-p_j)(p_i-p_k)p_i} + [\langle \hat{R}_3(0) \rangle + \frac{1}{2}] \sum_{i=1}^3 \frac{[(p_i+\beta)^2 + \beta^2\theta^2] e^{p_i t}}{(p_i-p_j)(p_i-p_k)} - \frac{1}{2} \frac{\Omega}{\beta} \sum_{i=1}^3 \frac{e^{p_i t}}{(p_i-p_j)(p_i-p_k)} \left(\frac{\langle \hat{b}_S^\dagger(0) \rangle e^{i\phi}}{(1-i\theta)} \{ [p_i+\beta(1-i\theta)](p_i+2\beta) - (p_i+\beta)^2 - \beta^2\theta^2 \} + \frac{\langle \hat{b}_S(0) \rangle e^{-i\phi}}{(1+i\theta)} \{ [p_i+\beta+i\beta\theta](p_i+2\beta) - (p_i+\beta)^2 - \beta^2\theta^2 \} \right), \quad (53)$$

where p_1, p_2, p_3 are the three roots (assumed to be unequal) of the cubic equation

$$p^3 + 4\beta p^2 + (5 + \theta^2 + \lambda)\beta^2 p + (2 + 2\theta^2 + \lambda)\beta^3 = 0. \quad (54)$$

The roots can be expressed in terms of the parameters θ and λ and are given by⁴⁰

$$p_1/\beta = -\frac{4}{3} + \eta_+ + \eta_-,$$

$$p_2/\beta = -\frac{4}{3} - \frac{1}{2}(\eta_+ + \eta_-) + i(\sqrt{3}/2)(\eta_+ - \eta_-), \quad (55a)$$

$$p_3/\beta = -\frac{4}{3} - \frac{1}{2}(\eta_+ + \eta_-) - i(\sqrt{3}/2)(\eta_+ - \eta_-),$$

with η_\pm given by the principal values of

$$\eta_\pm = \frac{1}{3} \left\{ \left(\frac{9}{2}\lambda - 9\theta^2 - 1 \right) \pm \left[\left(\frac{9}{2}\lambda - 9\theta^2 - 1 \right)^2 + (3\lambda + 3\theta^2 - 1)^3 \right]^{1/2} \right\}^{1/3}. \quad (55b)$$

Cases for which the roots are degenerate, such as $\theta = 0, \lambda = \frac{1}{4}$, have to be treated separately, but the solution (53) can be used as the critical values are approached. In the special case in which the initial atomic state is the ground state, $\langle \hat{R}_3(0) \rangle + \frac{1}{2}$

$= 0 = \langle \hat{\delta}_s(0) \rangle = \langle \hat{\delta}_s^\dagger(0) \rangle$, and the right-hand side of Eq. (53) reduces to the first two terms. Solutions of the still simpler problem in which the effective detuning θ also vanishes were already published some time ago.^{4, 10}

Some solutions given by Eqs. (53)–(55) for certain combinations of the parameters $\lambda \equiv \Omega^2/\beta^2$ and θ , and for certain initial atomic states, are represented in Figs. 1–4. In Figs. 1–3 the atom starts in the ground state, whereas in Fig. 4 the initial atomic state is a superposition of excited and ground states. After a long time the light intensity becomes independent of the initial state, which is evident from a comparison of Figs. 2 and 4. It will be seen that the evolution of the fluorescent light intensity tends to become increasingly oscillatory with time as the detuning increases. But even in the absence of detuning, the response has oscillatory features at sufficiently high field strengths, as has long been known within the context of semiclassical radiation theory, from solution of the Bloch equations.^{25, 37, 38} Indeed, the solution (53) is essentially equivalent to one obtained by Torrey³⁸ from the Bloch equations, and this emphasizes the small role played by the quantum field in the derivation.

Despite the fact that the normal ordering procedure we have followed avoids the need to explicitly bring in the operator character of the free field $\hat{A}_{St}^{(+)}(0, t)$ and $\hat{A}_{St}^{(-)}(0, t)$, as has already been discussed in the context of spontaneous emission,¹² we wish to emphasize that the fluorescent light has quantum features, which are clearly reflected in its fluctuations. This conclusion is especially evident if we examine the steady-state or long-time limit, for which we have in the far field,

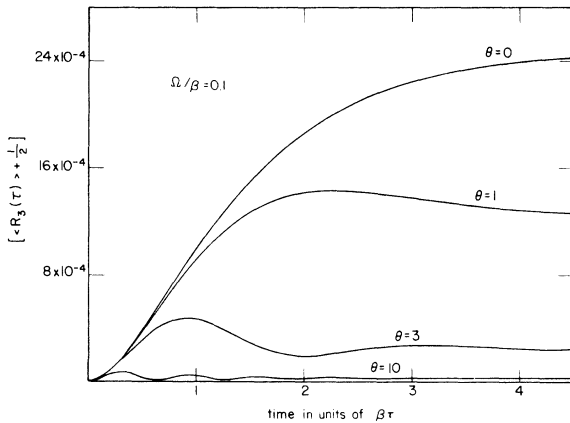


FIG. 1. Time development of the fluorescent light intensity, which is proportional to $\langle \hat{R}_3(\tau) \rangle + \frac{1}{2}$, for various detunings $\theta = (\omega_1 - \omega_0 + \gamma)/\beta$ of the exciting field. The atom is initially in the ground state, and the Rabi frequency $\Omega = 0.1\beta$.

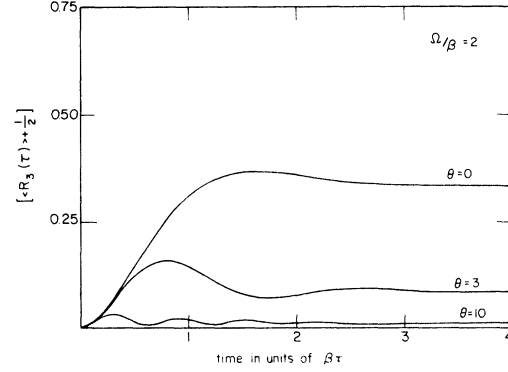


FIG. 2. Time development of the fluorescent light intensity, which is proportional to $\langle \hat{R}_3(\tau) \rangle + \frac{1}{2}$, for various detunings $\theta = (\omega_1 - \omega_0 + \gamma)/\beta$ of the exciting field. The atom is initially in the ground state, and the Rabi frequency $\Omega = 2\beta$.

from Eqs. (16) and (43),

$$\langle \hat{\mathbf{E}}^{(+)}(\vec{r}, t) \rangle = \frac{-\omega_0^2}{4\pi\epsilon_0 c^2} \left(\frac{\vec{\mu}}{r} - \frac{(\vec{\mu} \cdot \vec{r})\vec{r}}{r^3} \right) \times \frac{\frac{1}{2}(\Omega/\beta)(1+i\theta)}{\frac{1}{2}\Omega^2/\beta^2 + 1 + \theta^2} e^{i(\omega_0 - \omega_1)(t - r/c)} e^{i\phi},$$

while from Eqs. (40) and (42)

$$\langle \hat{\mathbf{E}}^{(-)}(\vec{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\vec{r}, t) \rangle = \left(\frac{\omega_0^2 \mu \sin\psi}{4\pi\epsilon_0 c^2 r} \right)^2 \frac{\frac{1}{4}\Omega^2/\beta^2}{\frac{1}{2}\Omega^2/\beta^2 + 1 + \theta^2}.$$

If we take the dimensionless ratio

$$\frac{\langle \hat{\mathbf{E}}^{(-)}(\vec{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\vec{r}, t) \rangle - \langle \hat{\mathbf{E}}^{(-)}(\vec{r}, t) \rangle \cdot \langle \hat{\mathbf{E}}^{(+)}(\vec{r}, t) \rangle}{\langle \hat{\mathbf{E}}^{(-)}(\vec{r}, t) \rangle \cdot \langle \hat{\mathbf{E}}^{(+)}(\vec{r}, t) \rangle} = \frac{\frac{1}{2}\Omega^2/\beta^2}{1 + \theta^2 + \frac{1}{2}\Omega^2/\beta^2} \quad (56)$$

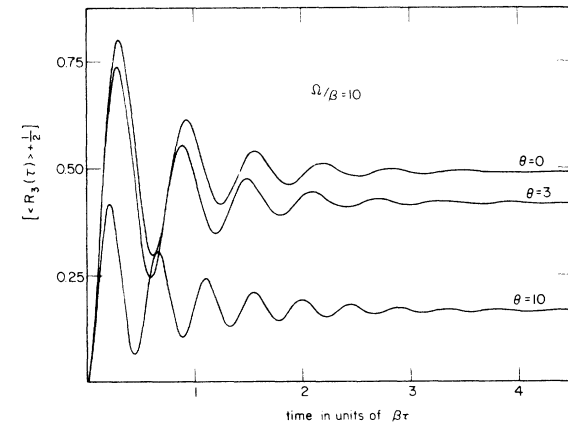


FIG. 3. Time development of the fluorescent light intensity, which is proportional to $\langle \hat{R}_3(\tau) \rangle + \frac{1}{2}$, for various detunings $\theta = (\omega_1 - \omega_0 + \gamma)/\beta$ of the exciting field. The atom is initially in the ground state, and the Rabi frequency $\Omega = 10\beta$.

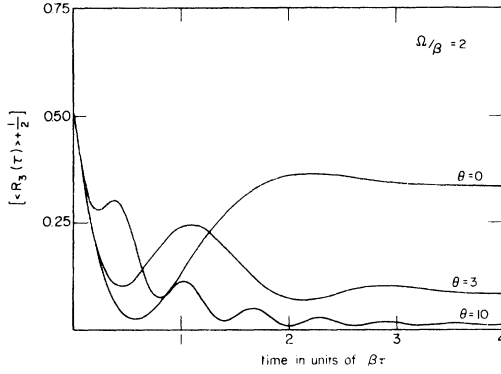


FIG. 4. Time development of the fluorescent light intensity, which is proportional to $\langle R_3(\tau) \rangle + \frac{1}{2}$, for various detunings $\theta = (\omega_1 - \omega_0 + \gamma)/\beta$ of the exciting field. The atom is initially in the partly excited state $(1/\sqrt{2})(|1\rangle + |2\rangle)$, the Rabi frequency $\Omega = 2\beta$ and the phase $\phi = 0$. Note that in the steady state the light intensity has the same values as in Fig. 2.

as a measure of the relative magnitude of the light fluctuations, we see that despite the coherent nature of the exciting field the fluorescent light is not coherent in the steady state, since the relative variance is nonzero. Indeed, the stronger and more "classical" the exciting field is, the greater are the fluctuations of the fluorescence, which must be attributed to quantum effects in the interaction between the atom and the field. Even with the introduction of so-called phenomenological damping terms, the semiclassical Bloch equations do not predict these fluctuations.^{25,41} The fact that a quasistationary state is reached can itself be regarded as a manifestation of these quantum fluctuations, as we show more explicitly in Sec. V.

V. TWO-TIME CORRELATION FUNCTIONS OF THE FLUORESCENCE IN THE WEAK FIELD

We now turn to the calculation of the spectral density of the fluorescent light emission, which might be measured via some combination of an interferometer with a photodetector in the far field. As is well known, this spectral density can be obtained by a Fourier transformation of the two-time correlation function of the electromagnetic field in normal order, and from Eq. (16), after decomposition of the field into positive and negative frequency parts, we obtain

$$\begin{aligned} & \langle \hat{\mathbf{E}}^{(-)}(\vec{r}, t) \cdot \hat{\mathbf{E}}^{(+)}(\vec{r}, t + \tau) \rangle \\ &= \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi \epsilon_0 c^2 r} \right)^2 e^{-i\omega_0 \tau} \langle \hat{b}_S^\dagger(t - r/c) \hat{b}_S(t + \tau - r/c) \rangle. \end{aligned} \quad (57a)$$

Here ψ is the angle between \vec{r} and $\vec{\mu}$ vectors, and we choose the point \vec{r} so that it lies outside the exciting beam and

$$\hat{\mathbf{E}}_{\text{free}}^{(+)}(\vec{r}, t) | \{v\} \rangle = 0. \quad (57b)$$

The required spectrum therefore follows directly from the atomic two-time correlation function of the second order, which we now examine.

To evaluate $\langle \hat{b}_S^\dagger(t) \hat{b}_S(t + \tau) \rangle$ we once again take the exciting field to be in a coherent state $|\{v\}\rangle$, with eigenvalues of $\hat{A}_{S1}^{(+)}(0, t)$ and $\hat{A}_{S1}^{(-)}(0, t)$ given by Eqs. (34) and (41). For simplicity we shall take the initial atomic state to be the ground state $|1\rangle$, which causes certain terms to vanish. Although the choice of initial state affects the initial transient response of the system, it has no effect on the asymptotic behavior in a constant field, and we shall therefore focus our attention on the long-term form of the correlation $\langle \hat{b}_S^\dagger(t) \hat{b}_S(t + \tau) \rangle$. From Eq. (30) we obtain by direct multiplication, after taking expectation values,

$$\begin{aligned} \langle \hat{b}_S^\dagger(t) \hat{b}_S(t + \tau) \rangle &= \frac{1}{4} \Omega^2 e^{(-\beta + i\gamma)\tau} e^{-2\beta t} \int_0^t dt' \int_0^{t+\tau} dt'' e^{\beta(1+i\theta)t''} e^{\beta(1-i\theta)t''} \\ &+ \frac{1}{4} \Omega^3 e^{(-\beta + i\gamma)\tau} e^{-2\beta t} \int_0^t dt' e^{\beta(1+i\theta)t'} \int_0^{t+\tau} dt'' e^{-\beta(1+i\theta)t''} \int_0^{t''} dt''' [\langle \hat{b}_S^\dagger(t''') \rangle e^{i(\omega_0 - \omega_1)t'''' + i\theta} + \text{c.c.}] e^{2\beta t''''} \\ &+ \frac{1}{4} \Omega^3 e^{(-\beta + i\gamma)\tau} e^{-2\beta t} \int_0^{t+\tau} dt' e^{\beta(1-i\theta)t'} \int_0^t dt'' e^{-\beta(1-i\theta)t''} \int_0^{t''} dt''' [\langle \hat{b}_S^\dagger(t''') \rangle e^{i(\omega_0 - \omega_1)t'''' + i\theta} + \text{c.c.}] e^{2\beta t''''} \\ &+ \Omega^2 \left(\frac{\omega_0}{\hbar} \right)^2 e^{(-\beta + i\gamma)\tau} e^{-2\beta t} \int_0^t dt_1 e^{-\beta(1-i\theta)t_1} \\ &\quad \times \int_0^{t_1} dt_2 \left\langle [\vec{\mu} \cdot \vec{V}^*(t_2) \hat{b}_S(t_2) + \hat{b}_S^\dagger(t_2) \vec{\mu} \cdot \vec{A}_S^{(+)}(0, t_2)] e^{2\beta t_2} \right. \\ &\quad \times \int_0^{t_2+\tau} dt_3 e^{-\beta(1+i\theta)t_3} \int_0^{t_3} dt_4 [\hat{b}_S^\dagger(t_4) \vec{\mu} \cdot \vec{V}(t_4) \\ &\quad \left. + \vec{\mu} \cdot \vec{A}_S^{(-)}(0, t_4) \hat{b}_S(t_4)] \right\rangle e^{2\beta t_4}. \end{aligned} \quad (58)$$

Similarly, we may use Eq. (30) to construct correlation functions in some other order, such as $\langle \hat{b}_s(t) \hat{b}_s^\dagger(t+\tau) \rangle$. The resulting system of coupled integral equations can then be solved for each correlation function in turn. We shall return to this general problem in Sec. VII. However, when the external field is weak, so that $\Omega/\beta \ll 1$ and the higher powers of Ω/β can be discarded, Eq. (58) lends itself directly to a term-by-term evaluation of $\langle \hat{b}_s^\dagger(t) \hat{b}_s(t+\tau) \rangle$, as we now show. The condition $\Omega/\beta \ll 1$ can be interpreted to mean that the Rabi

oscillation frequency in the external field is small compared with the spontaneous decay rate.

We first observe that, because of the factors $e^{2\beta t}$ in the integrands of the second and third terms, the steady-state values of $\langle \hat{b}_s(t) \rangle$ and $\langle \hat{b}_s^\dagger(t) \rangle$ make the dominant contribution to these terms in the long-time limit. If we insert the long-time solutions given by Eq. (43) into these terms in Eq. (58), and leave the last term containing the fourfold integral for the moment, we find that, as $t \rightarrow \infty$,

$$\begin{aligned} \left[\text{Contributions of first three terms to } \langle \hat{b}_s^\dagger(t) \hat{b}_s(t+\tau) \rangle \right] &\rightarrow \frac{\Omega^2 e^{i(\omega_0 - \omega_1)\tau}}{4\beta^2(1+\theta^2)} - \frac{\Omega^4 e^{i(\omega_0 - \omega_1)\tau}}{8\beta^4(1+\theta^2)(1+\theta^2 + \Omega^2/2\beta^2)} \\ &\quad - \frac{\Omega^4 e^{i(\omega_0 - \omega_1)\tau}}{8\beta^4(1+\theta^2)(1+\theta^2 + \Omega^2/2\beta^2)} \\ &= \frac{1}{4} \frac{\Omega^2 e^{i(\omega_0 - \omega_1)\tau}}{\beta^2(1+\theta^2)} \left(1 - \frac{\Omega^2}{\frac{1}{2}\Omega^2 + \beta^2(1+\theta^2)} \right). \end{aligned} \quad (59)$$

This contribution to the correlation function $\langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t+\tau) \rangle$ therefore becomes independent of t after a long time, but continues to oscillate indefinitely with increasing τ at the driving frequency ω_1 . It obviously corresponds to monochromatic light that is elastically scattered by the atom and contributes no spectral width to the fluorescence. Any decay of the correlation function with τ , i.e., any finite spectral width, must evidently come from the last term in Eq. (58), which can itself be divided into four contributions involving the following four correlation functions:

$$\begin{aligned} &\vec{\mu} \cdot \vec{V}^*(t_2) \vec{\mu} \cdot \vec{V}(t_4) \langle \hat{b}_s(t_2) \hat{b}_s^\dagger(t_4) \rangle, \\ &\langle \hat{b}_s^\dagger(t_2) \vec{\mu} \cdot \hat{A}_s^{(+)}(0, t_2) \vec{\mu} \cdot \hat{A}_s^{(-)}(0, t_4) \hat{b}_s(t_4) \rangle, \\ &\vec{\mu} \cdot \vec{V}^*(t_2) \langle \hat{b}_s(t_2) \vec{\mu} \cdot \hat{A}_s^{(-)}(0, t_4) \hat{b}_s(t_4) \rangle, \\ &\vec{\mu} \cdot \vec{V}(t_4) \langle \hat{b}_s^\dagger(t_2) \vec{\mu} \cdot \hat{A}_s^{(+)}(0, t_2) \hat{b}_s^\dagger(t_4) \rangle. \end{aligned} \quad (60)$$

We now proceed to evaluate these correlations up to terms of the second order in Ω/β with the help of Eq. (30), so as to obtain the value of the last term in Eq. (58) to $O((\Omega/\beta)^4)$.

We start with the first term in (60), which calls for the determination of $\langle \hat{b}_s(t_2) \hat{b}_s^\dagger(t_4) \rangle$, but only to zeroth order in Ω/β . By direct multiplication of Eq. (30) and evaluation of expectations with the help of Eq. (34), we have, up to terms of order Ω/β ,

$$\begin{aligned} \langle \hat{b}_s(t) \hat{b}_s^\dagger(t+\tau) \rangle &= e^{-2\beta t} e^{(-\beta - i\gamma)\tau} + \left(\frac{\omega_0}{\hbar} \right)^2 e^{-2\beta t} e^{(-\beta - i\gamma)\tau} \int_0^t dt' \int_0^{t+\tau} dt'' e^{(\beta - i\gamma)t'} e^{(\beta + i\gamma)t''} \langle \vec{\mu} \cdot \hat{A}_s^{(+)}(0, t') \vec{\mu} \cdot \hat{A}_s^{(-)}(0, t'') \rangle \\ &\quad + [\text{terms } O(\Omega^2/\beta^2) \text{ and higher}]. \end{aligned} \quad (61)$$

The first term is of order unity, but becomes negligible after a sufficiently long time t . The second term can be evaluated with the help of the commutator $[\mu_i \hat{A}_{S_i}^{(+)}(0, t'), \mu_j \hat{A}_{S_j}^{(-)}(0, t'')]$, which is examined in the Appendix. We find that under the integral in Eq. (58) the commutator may to a good approximation be written

$$[\mu_i \hat{A}_{S_i}^{(+)}(0, t'), \mu_j \hat{A}_{S_j}^{(-)}(0, t'')] \approx 2\beta(\hbar^2/\omega_0^2) \delta(t' - t''), \quad (62)$$

provided the range of t', t'' integration is very

great compared with the optical period $1/\omega_0$. When this result is inserted in Eq. (61) and we pass to the long-time limit, we obtain

$$\langle \hat{b}_s(t) \hat{b}_s^\dagger(t+\tau) \rangle \rightarrow \begin{cases} e^{(-\beta - i\gamma)\tau} + O(\Omega^2/\beta^2), & \text{if } \tau \geq 0, \\ e^{(\beta - i\gamma)\tau} + O(\Omega^2/\beta^2), & \text{if } \tau \leq 0, \end{cases} \quad (63)$$

Hence to the lowest order of Ω/β we have the result

$$\langle \hat{b}_s(t) \hat{b}_s^\dagger(t+\tau) \rangle \rightarrow e^{-\beta|\tau|} e^{-i\gamma\tau}, \quad (64)$$

which is independent of t but falls off exponentially with the time difference τ . It should be noted that the exponential falloff comes directly from the introduction of the commutator (62) in Eq. (61), and is therefore a true quantum-field effect.

The commutation relation (62) allows a simple heuristic interpretation of the result in Eq. (64) to be given. If we look at Eqs. (26) and (27), from which we started, as coupled Langevin-type equations for the dynamical variables $\hat{b}_S(t)$ and $\hat{R}_3(t)$, with quantum "noise sources"³² $\hat{A}_{Si}^{(+)}(0, t)$ and $\hat{A}_{Si}^{(-)}(0, t)$, then Eq. (62) implies that these quantum noise sources are δ correlated in some sense, and therefore represent a Markoffian random process. The exponential falloff of the correlation function $\langle \hat{b}_S(t) \hat{b}_S^\dagger(t + \tau) \rangle$ is then a reflection of this Markoffian process. This may be considered a rough justification for the Markoffian assumption that is introduced from the beginning in certain treatments,^{6, 15, 18} which can also be justified more formally via the so-called quantum-regression theorem.⁴²

We now turn to the evaluation of the second term in (60), and again make use of the commutation

relation (62). We then obtain

$$\begin{aligned} & \langle \hat{b}_S^\dagger(t_2) \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t_2) \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \hat{b}_S(t_4) \rangle \\ & - 2\beta(\hbar^2/\omega_0^2) \langle \hat{b}_S^\dagger(t_2) \hat{b}_S(t_2) \rangle \delta(t_2 - t_4) \\ & + \langle \hat{b}_S^\dagger(t_2) \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t_2) \hat{b}_S(t_4) \rangle. \end{aligned} \quad (65)$$

The second term on the right-hand side can be expanded with the help of Eq. (30), but its contribution is of order higher than Ω^2/β^2 . The first term on the right-hand side is given (in the long-time limit) by Eq. (42), so that we have, to order Ω^2/β^2 ,

$$\begin{aligned} & \langle \hat{b}_S^\dagger(t_2) \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t_2) \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \hat{b}_S(t_4) \rangle \\ & - \beta \frac{\hbar^2}{\omega_0^2} \left(\frac{\frac{1}{2}\Omega^2}{\frac{1}{2}\Omega^2 + \beta^2(1 + \theta^2)} \right) \delta(t_2 - t_4). \end{aligned} \quad (66)$$

Finally, we consider the third and fourth terms in (60), which are essentially conjugates of each other. Once again we have, by direct multiplication from Eq. (30),

$$\begin{aligned} & \vec{\mu} \cdot \vec{V}^*(t_2) \langle \hat{b}_S(t_2) \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \hat{b}_S(t_4) \rangle \\ & = \left(\frac{\omega_0}{\hbar} \right)^2 \vec{\mu} \cdot \vec{V}^*(t_2) e^{(-\beta + i\gamma)(t_2 + t_4)} \int_0^{t_2} dt' \int_0^{t_4} dt'' e^{(\beta - i\gamma)(t' + t'')} \langle \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t') \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t'') \rangle + O(\Omega^3). \end{aligned} \quad (67)$$

If $\mu_i \hat{A}_{Si}^{(+)}(0, t)$ and $\mu_i \hat{A}_{Si}^{(-)}(0, t)$ were treated as c numbers, the first term in Eq. (67) would be of order Ω^4/β^4 and therefore negligible to the order in which we are working. However, with the help of the commutation relation (62), we readily find in the long-time limit, to order Ω^2/β^2 ,

$$\begin{aligned} & \vec{\mu} \cdot \vec{V}^*(t_2) \langle \hat{b}_S(t_2) \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \hat{b}_S(t_4) \rangle \\ & - \frac{1}{2} \frac{\hbar^2}{\omega_0^2} \Omega^2 \frac{e^{-\beta(1-i\theta)t_2} e^{\beta(1-i\theta)t_4}}{1-i\theta} u(t_2 - t_4), \end{aligned} \quad (68)$$

where $u(\tau)$ is the unit step function, which vanishes

for $\tau < 0$ and is unity for $\tau \geq 0$. Similarly,

$$\begin{aligned} & \vec{\mu} \cdot \vec{V}(t_4) \langle \hat{b}_S^\dagger(t_2) \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t_2) \hat{b}_S^\dagger(t_4) \rangle \\ & - \frac{1}{2} \frac{\hbar^2}{\omega_0^2} \Omega^2 \frac{e^{-\beta(1+i\theta)t_4} e^{\beta(1+i\theta)t_2}}{1+i\theta} u(t_4 - t_2). \end{aligned} \quad (69)$$

We may now proceed to use Eqs. (64), (66), (68), and (69) in order to evaluate the contribution of each term in (60) to the fourfold integral in Eq. (58). A straightforward but somewhat lengthy and tedious calculation leads to the following results in the long-term limit to order $(\Omega/\beta)^4$:

$$\begin{aligned} & \left(\frac{\Omega \omega_0}{\hbar} \right)^2 e^{(-\beta + i\gamma)\tau} e^{-2\beta t} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2 + \tau} dt_3 \int_0^{t_3} dt_4 e^{-\beta(1-i\theta)t_1} e^{-\beta(1+i\theta)t_3} e^{2\beta(t_4 + t_2)} \vec{\mu} \cdot \vec{V}^*(t_2) \langle \hat{b}_S(t_2) \hat{b}_S^\dagger(t_4) \rangle \vec{\mu} \cdot \vec{V}(t_4) \\ & - \frac{\Omega^4 e^{-\beta t} e^{i\gamma \tau}}{16\beta^4(1+\theta^2)(9+\theta^2)} \left(1 + e^{-2i\beta\theta\tau} + \frac{3}{i\theta} (1 - e^{-2i\beta\theta\tau}) [u(\tau) - u(-\tau)] + 2e^{-\beta t} e^{-i\beta\theta\tau} \right), \end{aligned} \quad (70)$$

$$\begin{aligned} & \left(\frac{\Omega \omega_0}{\hbar} \right)^2 e^{(-\beta + i\gamma)\tau} e^{-2\beta t} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2 + \tau} dt_3 \int_0^{t_3} dt_4 e^{-\beta(1-i\theta)t_1} e^{-\beta(1+i\theta)t_3} e^{2\beta(t_4 + t_2)} \langle \hat{b}_S^\dagger(t_2) \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t_2) \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \hat{b}_S(t_4) \rangle \\ & - \frac{\Omega^4}{8\beta^4(1+\theta^2)} \left[\left(\frac{2e^{(-\beta + i\gamma)\tau}}{(1+i\theta)(3-i\theta)} - \frac{e^{[-2\beta + i(\omega_0 - \omega_1)]\tau}}{(1+i\theta)(3+i\theta)} \right) u(\tau) + \left(\frac{2e^{(\beta + i\gamma)\tau}}{(1-i\theta)(3+i\theta)} - \frac{e^{[2\beta + i(\omega_0 - \omega_1)]\tau}}{(1-i\theta)(3-i\theta)} \right) u(-\tau) \right], \end{aligned} \quad (71)$$

$$\begin{aligned}
& \left(\frac{\Omega\omega_0}{\hbar}\right)^2 e^{(-\beta+i\gamma)\tau} e^{-2\beta t} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 e^{-\beta(1-i\theta)t_1} e^{-\beta(1+i\theta)t_3} e^{2\beta(t_4+t_2)} \\
& \quad \times [\vec{\mu} \cdot \vec{V}^*(t_2) \langle \hat{b}_S(t_2) \vec{\mu} \cdot \hat{A}_S^{(-)}(0, t_4) \hat{b}_S(t_4) \rangle + \langle \hat{b}_S^\dagger(t_2) \vec{\mu} \cdot \hat{A}_S^{(+)}(0, t_2) \hat{b}_S^\dagger(t_4) \rangle \vec{\mu} \cdot \vec{V}(t_4)] \\
& \quad - \frac{\Omega^4 e^{-\beta\tau} e^{i\gamma\tau}}{8\beta^4(1+\theta^2)} \left(\frac{1-i\theta-2\theta^2}{i\theta(3-i\theta)(1+\theta^2)} + \frac{2(1-i\theta)e^{-\beta\tau} e^{-i\beta\theta\tau}}{(1+i\theta)(9+\theta^2)} - \frac{e^{-2i\beta\theta\tau}}{i\theta(1+i\theta)(3+i\theta)} \right) u(\tau) \\
& \quad + \frac{\Omega^4 e^{\beta\tau} e^{i\gamma\tau}}{8\beta^4(1+\theta^2)} \left(\frac{1+i\theta-2\theta^2}{(-i\theta)(3+i\theta)(1+\theta^2)} + \frac{2(1+i\theta)e^{\beta\tau} e^{-i\beta\theta\tau}}{(1-i\theta)(9+\theta^2)} + \frac{e^{-2i\beta\theta\tau}}{i\theta(1-i\theta)(3-i\theta)} \right) u(-\tau). \quad (72)
\end{aligned}$$

Finally, we combine the results given by Eqs. (70)–(72), so as to yield the total contribution of the last term in Eq. (58). Together with the contribution of the first three terms, given by Eq. (59), we then have the following result for the atomic correlation function to order $(\Omega/\beta)^4$, in the long-term limit:

$$\begin{aligned}
\langle \hat{b}_S^\dagger(t) \hat{b}_S(t+\tau) \rangle & \rightarrow \frac{\Omega^2 e^{i(\omega_0-\omega_1)\tau}}{4\beta^2(1+\theta^2)} \left(1 - \frac{\Omega^2}{\beta^2(1+\theta^2)} \right) \\
& \quad + \frac{\Omega^4 e^{-\beta|\tau|} e^{i(\omega_0-\omega_1)\tau}}{8\beta^4\theta(1+\theta^2)^2} \\
& \quad \times [\sin(\beta\theta|\tau|) + \theta \cos\beta\theta\tau]. \quad (73)
\end{aligned}$$

A number of interesting general conclusions follow from this equation. In the first place, we note that the answer is independent of time t for large t , which shows that the system eventually settles down to a quasistationary state, at least in the wide sense. Of course the system is not strictly stationary, because of the presence of the monochromatic exciting field in a coherent state. Its presence is demonstrated by Eq. (43), which shows explicitly that $\langle \hat{b}_S(t) \rangle$ does not become t independent, and is reflected in the strictly oscillatory contribution to $\langle \hat{b}_S^\dagger(t) \hat{b}_S(t+\tau) \rangle$. Secondly, we note that Eq. (73) has the proper symmetry property for the correlation function of a stationary process, i.e.,

$$\langle \hat{b}_S^\dagger(t) \hat{b}_S(t-\tau) \rangle = \langle \hat{b}_S^\dagger(t) \hat{b}_S(t+\tau) \rangle^*,$$

and that it reduces to Eq. (42) to order $(\Omega/\beta)^4$ when $\tau=0$, as required. Moreover, the τ symmetry comes explicitly out of the calculation, rather than indirectly from stationarity. Finally, we note the presence of terms in Eq. (73) that fall off exponentially with τ in both directions, and we recall that they come from those contributions to the last term in Eq. (58) that involve the free-field commutation relations (62). If $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ had been treated as c numbers, this τ -symmetric exponentially decaying contribution, which of course leads to a finite spectral width, would be entirely absent. The quantum properties of the free field therefore show up much more explicitly in the two-time correlation function for a driven atom than in the transient behavior we considered

previously. In the language of the theory of random processes, we may say that the exponentially decaying contribution to the steady-state correlation function is a manifestation of a δ -correlated—or Markoffian—random process, that in this case reflects the presence of quantum fluctuations both of the field and of the atomic source. The two are of course intimately connected, and are inseparable in a consistent theory.

VI. SPECTRAL DENSITY OF THE RESONANCE FLUORESCENCE IN A WEAK FIELD

We have already seen from Eq. (57) that, apart from retardation effects, $\langle \hat{b}_S^\dagger(t) \hat{b}_S(t+\tau) \rangle$ is directly proportional to the normally ordered correlation function $\langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t+\tau) \rangle$ of the fluorescent light at some suitably chosen point \vec{r} in the far-field. By taking the Fourier transform of $\langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t+\tau) \rangle$ with respect to τ , and making use of Eq. (73), we obtain the spectral density $\Phi(\vec{r}, \omega)$ of the fluorescent electromagnetic field at \vec{r} . We then find to order $(\Omega/\beta)^4$, in the long-time limit,

$$\begin{aligned}
\Phi(\vec{r}, \omega) & = \int_{-\infty}^{\infty} d\tau \langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t+\tau) \rangle e^{i\omega\tau} \\
& = \left(\frac{\omega_0^2 \mu}{4\pi\epsilon_0 c^2} \right)^2 \frac{\Omega^2}{2\beta^2(1+\theta^2)} \frac{\sin^2\psi}{r^2} \\
& \quad \times \left[\pi \left(1 - \frac{\Omega^2}{\beta^2(1+\theta^2)} \right) \delta(\omega - \omega_1) \right. \\
& \quad \left. + \frac{\beta\Omega^2}{[\beta^2 + (\omega - \omega_0 + \gamma)^2][\beta^2 + (\omega + \omega_0 - \gamma - 2\omega_1)^2]} \right], \quad (74)
\end{aligned}$$

where ψ , as before, is the angle between the \vec{r} and $\vec{\mu}$ vectors. The contribution of the first term obviously corresponds to light that is elastically scattered by the atom. The second term, whose integral over all ω is smaller by the factor Ω^2/β^2 , contributes two approximately Lorentzian distributions symmetrically placed with respect to the driving frequency ω_1 , and displaced from it by $\pm|\gamma + \omega_1 - \omega_0|$. This corresponds to inelastic scattering, mediated by the quantum nature of the pro-

cess. This contribution is absent in a semiclassical theory, since it depends directly on the free-field commutation relation. Experimental demonstration of the existence of these peaks therefore represents a more searching test of QED than the study of spontaneous emission, and some experimental results have already been published.³⁴ It is not difficult to see why the existence of a contribution of finite bandwidth to the spectral density has a significance here that is altogether different from its significance in spontaneous emission in the vacuum. An excited atom has only a finite amount of stored energy and can radiate only for a finite time, so that the width of the spectral density has to be finite in spontaneous emission. However, in this problem we are dealing with an atom that is continuously excited by a strictly monochromatic field; in this case the finite bandwidth of the fluorescence is attributable to quantum fluctuations. Equation (74) is in essential agreement with the results of Mollow,^{5,16} Agarwal,⁶ and Walls and his co-workers.¹⁵ We emphasize that our results were obtained by direct evaluation of the correlation function, without the introduction of any classical driving fields or initial assumptions about stationarity or Markofficity, although both of these assumptions are now seen to be justified in a sense.

VII. TWO-TIME CORRELATION FUNCTIONS IN GENERAL

We now turn to the more general problem of evaluating the two-time correlation function of the resonance fluorescence produced in an external field, without restriction on the strength of the field or on the time. This time we shall not find it possible to proceed in quite such a straightforward manner as before, and evaluate the correlation directly from Eq. (58). Instead we need to solve several coupled equations of motion for several different correlation functions. Although the weak-field case we have just treated is a special case of this more general problem, it provides insights that are not available in the general case, in that we were able to identify the quantum-field fluctuations as the source of the finite linewidth. This is much less obvious in the following more general treatment.

We start by integrating Eqs. (26) and (27) over a finite time interval from t to $t + \tau$, with $\tau \geq 0$, and, as before, we discard the integral of terms oscillating at double the optical frequency. We then obtain

$$\begin{aligned} \hat{b}_S^\dagger(t + \tau) &= \hat{b}_S^\dagger(t) e^{-(\beta + i\gamma)\tau} \\ &+ \frac{2\omega_0}{\hbar} \int_0^\tau dt' \hat{R}_3(t + t') \mu_i \hat{A}_{Si}^{(+)}(0, t + t') \\ &\times e^{(i\beta - i\gamma)(t' - \tau)}, \end{aligned} \quad (75)$$

$$\begin{aligned} \hat{R}_3(t + \tau) + \frac{1}{2} &= [\hat{R}_3(t) + \frac{1}{2}] e^{-2\beta\tau} \\ &- \frac{\omega_0}{\hbar} \int_0^\tau dt' [\hat{b}_S^\dagger(t + t') \mu_i \hat{A}_{Si}^{(+)}(0, t + t') \\ &+ \text{H.c.}] e^{2\beta(t' - \tau)}. \end{aligned} \quad (76)$$

We now follow a procedure first used by Milonni¹³ of forming several different correlation functions, and solving for them simultaneously, although our method of solution is different from his, and our treatment is more general in some respects. By multiplying equations (75) and (76) and their conjugates by $\hat{b}_S^\dagger(t)$ on the left-hand side and taking expectation values, we obtain several different correlation functions.

It is convenient to introduce the following notation for the case where the exciting electromagnetic field is in the coherent state given by Eqs. (34) and (41):

$$\langle \hat{b}_S^\dagger(t) \hat{b}_S(t + \tau) \rangle e^{i(\omega_1 - \omega_0)\tau} \equiv g(t, \tau), \quad (77)$$

$$\langle \hat{b}_S^\dagger(t) \hat{b}_S^\dagger(t + \tau) \rangle e^{i(\omega_0 - \omega_1)(2t + \tau)} e^{2i\phi} \equiv f(t, \tau), \quad (78)$$

$$\langle \hat{b}_S^\dagger(t) \hat{R}_3(t + \tau) \rangle e^{i(\omega_0 - \omega_1)t} e^{i\phi} \equiv h(t, \tau). \quad (79)$$

These correlation functions are similar to, but not identical with, the functions introduced by Milonni.¹³ The main difference is that $g(t, \tau)$, $f(t, \tau)$ and $h(t, \tau)$ do not oscillate at the difference frequency $\omega_1 - \omega_0$; moreover, they become independent of t and ϕ for large t , as we shall see. From Eqs. (75) and (76) we then obtain the following relations among $g(t, \tau)$, $f(t, \tau)$, and $h(t, \tau)$ for $\tau \geq 0$:

$$\begin{aligned} g(t, \tau) &= [\langle \hat{R}_3(t) \rangle + \frac{1}{2}] e^{-\beta(1 - i\theta)\tau} \\ &+ \Omega \int_0^\tau dt' h(t, t') e^{\beta(1 - i\theta)(t' - \tau)}, \end{aligned} \quad (80)$$

$$f(t, \tau) = \Omega \int_0^\tau dt' h(t, t') e^{\beta(1 + i\theta)(t' - \tau)}, \quad (81)$$

$$\begin{aligned} h(t, \tau) &= -\frac{1}{2} \langle \hat{b}_S^\dagger(t) \rangle e^{i(\omega_0 - \omega_1)t} e^{i\phi} \\ &- \frac{1}{2} \Omega \int_0^\tau dt' [f(t, t') + g(t, t')] e^{2\beta(t' - \tau)}. \end{aligned} \quad (82)$$

In the derivation of these relations we have made use of the following two operator properties:

$$\hat{b}_S^\dagger(t) \hat{b}_S^\dagger(t) = 0 = \hat{b}_S^\dagger(t) [\hat{R}_3(t) + \frac{1}{2}] \quad (83a)$$

and

$$[\hat{b}_S^\dagger(t), \mu_i \hat{A}_{Si}^{(+)}(0, t + \tau)] = 0 = [\hat{b}_S^\dagger(t), \mu_i \hat{A}_{Si}^{(-)}(0, t + \tau)], \quad (83b)$$

for $\tau \geq 0$. That Eqs. (83b) hold for $\tau = 0$ follows immediately from Eqs. (21), for the total-field terms $\mu_i \hat{A}_i^{(+)}(0, t)$, $\mu_i \hat{A}_i^{(-)}(0, t)$ commute with all atomic operators at the same time t . The validity of the equations for $\tau > 0$ is less obvious, although it might be expected in an intuitive manner from

the fact that $\hat{A}_{S_i}^{(+)}(0, t + \tau)$ evolves from $\hat{A}_{S_i}^{(+)}(0, t)$ as a free field. A rigorous proof, based on advanced and retarded electromagnetic Green's functions, of the commutation relation

$$[\hat{J}(t), \hat{E}_{\text{free}}(\vec{r}, t + \tau)] = 0, \quad \text{for } \tau > r/c, \quad (84)$$

where $\hat{J}(t)$ is any atomic operator, has recently been given by Mollow.⁴³ The corresponding commutator for the positive and negative frequency parts of \hat{E}_{free} should then vanish also, provided, as always, we restrict ourselves to time intervals long compared with the optical period. Equations (83b) follow directly from Eq. (84) as a special case, after integration with respect to τ . However, it should be noted that Eqs. (83b) do not hold for $\tau < 0$, which is also to be expected on intuitive grounds from the fact that the presence of the free field affects the later time evolution of the atom. We shall return to this point shortly. The same problem has also been discussed very recently by Renaud, Whitley and Stroud.⁴¹

We now proceed to eliminate $h(t, \tau)$ among Eqs. (80)–(82) and to derive an integral equation for the combination $g(t, \tau) + h(t, \tau)$. Thus from Eqs. (80) and (81) we obtain

$$g(t, \tau) + f(t, \tau) = [\langle \hat{R}_3(t) \rangle + \frac{1}{2}] e^{-\beta(1-i\theta)\tau} + 2\Omega \int_0^\tau dt' h(t, t') e^{\beta(t'-\tau)} \cos\beta\theta(t' - \tau), \quad (85)$$

$$g(t, \tau) + f(t, \tau) = \frac{-2\Omega}{\beta(2 + 2\theta^2 + \Omega^2/\beta^2)} \langle \hat{b}_S^\dagger(t) \rangle e^{i(\omega_0 - \omega_1)t} e^{i\phi} + \sum_{\substack{i=1 \\ i \neq j \neq k}}^3 \left[-\Omega \langle \hat{b}_S^\dagger(t) \rangle e^{i(\omega_0 - \omega_1)t} e^{i\phi} \left(1 + \frac{\beta}{p_i} \right) + [\langle \hat{R}_3(t) \rangle + \frac{1}{2}] [p_i + \beta(1 + i\theta)] \right] \frac{(2\beta + p_i) e^{\beta i \tau}}{(p_i - p_j)(p_i - p_k)}, \quad (89)$$

in which p_1, p_2, p_3 (assumed to be unequal) are given by Eqs. (55a) and (55b).

We have already shown that for long times t $\langle \hat{R}_3(t) \rangle$ and $\langle \hat{b}_S^\dagger(t) \rangle e^{i(\omega_0 - \omega_1)t} e^{i\phi}$ become independent of both t and ϕ [cf. Eqs. (42) and (43)]. It follows from Eq. (87) that $m(t, \tau)$ also becomes independent of t and ϕ for large t , and from Eq. (86) so does $g(t, \tau) + f(t, \tau)$, since the kernel $K(\tau)$ does not contain t or ϕ . Equations (80)–(82) then show that all three correlation functions $g(t, \tau)$, $f(t, \tau)$, and $h(t, \tau)$ separately become independent of t and ϕ

and with the help of Eq. (82) we have, after integration by parts,

$$g(t, \tau) + f(t, \tau) = m(t, \tau) + \int_0^\tau dt' K(\tau - t') [g(t, t') + f(t, t')], \quad (86)$$

for $\tau \geq 0$, where

$$m(t, \tau) \equiv [\langle \hat{R}_3(t) \rangle + \frac{1}{2}] e^{-\beta(1-i\theta)\tau} - \langle \hat{b}_S^\dagger(t) \rangle e^{i(\omega_0 - \omega_1)t} e^{i\phi} [\Omega/\beta(1 + \theta^2)] \times [1 - e^{-\beta\tau}(\cos\beta\theta\tau - \theta \sin\beta\theta\tau)], \quad (87)$$

and

$$K(\tau) \equiv [\Omega^2/\beta(1 + \theta^2)] [e^{-2\beta\tau} - e^{-\beta\tau}(\cos\beta\theta\tau + \theta \sin\beta\theta\tau)]. \quad (88)$$

Equation (86) is a Volterra integral equation of the same kind as our earlier integral Eq. (36) for $\langle \hat{R}_3(t) \rangle$, in which, moreover, the kernel is exactly the same [cf. Eq. (46)], although the inhomogeneous term $m(t, \tau)$ is different. The general solution can be written as before, and takes the form

for large times t , so that a quasistationary state is reached. This quasistationarity is again a consequence of the quantum fluctuations, although the conclusion may be less transparent than before.

Once $g(t, \tau) + f(t, \tau)$ has been found, it is only a matter of substitution to derive $h(t, \tau)$ from Eq. (82), $f(t, \tau)$ from Eq. (81), and $g(t, \tau)$ from Eq. (80). Thus we find for the atomic correlation function $g(t, \tau)$, at any time t , for any initial atomic state, and for $\tau \geq 0$,

$$g(t, \tau) = \frac{-\frac{1}{2}\Omega e^{i(\omega_0 - \omega_1)t} e^{i\phi}}{\beta(\frac{1}{2}\Omega^2/\beta^2 + 1 + \theta^2)} \langle \hat{b}_S^\dagger(t) \rangle (1 + i\theta) + \sum_{\substack{i=1 \\ i \neq j \neq k}}^3 \frac{e^{\beta i \tau}}{(p_i - p_j)(p_i - p_k)} \left([\langle \hat{R}_3(t) \rangle + \frac{1}{2}] [(p_i + 2\beta)(p_i + \beta + i\beta\theta) + \frac{1}{2}\Omega^2] - \frac{1}{2} \frac{\Omega}{p_i} (p_i + 2\beta)(p_i + \beta + i\beta\theta) \langle \hat{b}_S^\dagger(t) \rangle e^{i(\omega_0 - \omega_1)t} e^{i\phi} \right), \quad (90)$$

in which $\langle \hat{R}_3(t) \rangle$ is given by Eq. (53), and $\langle \hat{b}_s^\dagger(t) \rangle$ by Eq. (28) when the value of $\langle \hat{R}_3(t) \rangle$ is substituted under the integral.

So far our calculation has been confined to non-negative values of τ , and for good reason. The commutation relations (83b) do not hold for $\tau < 0$, and when the commutators are nonzero the foregoing simple procedure for deriving $g(t, \tau)$ breaks down. However, it follows from the definition (77) that

$$g(t, -\tau) = g^*(t - \tau, \tau), \quad (91)$$

and since $g(t, \tau)$ has been shown to become independent of the first argument for large t , we have immediately

$$g(t, -\tau) \rightarrow g^*(t, \tau) \text{ as } t \rightarrow \infty. \quad (92)$$

The function $g(t, \tau)$ is therefore known for both positive and negative values of its second argument τ , once it is known for $\tau \geq 0$ and for all values of its first argument.

It is possible, in principle, to associate a time-dependent spectral density $\Psi(t, \omega)$ with the time-dependent correlation function $g(t, \tau)$ given by Eq. (90), through a finite Fourier integral. Thus we might define

$$\Psi(t, \omega) = K \int_0^t d\tau g(t - \tau, \tau) e^{i(\omega - \omega_1)\tau} + \text{c.c.}, \quad (93)$$

where K is some constant, and, in view of the close relationship (57) between the atomic and the field correlation functions, we might interpret this quantity as the rate of production of photons of frequency ω at time t . Had we started from a mode expansion of the electromagnetic field, we would have been led to an equation of the form (93) for the rate of change of the number of photons. But, despite this interpretation, $\Psi(t, \omega)$ is not a physically meaningful quantity in the sense of being accessible to direct measurement. During the time that $\Psi(t, \omega)$ is changing with respect to its first argument t , the time scale for the variation is of the same order as or less than the reciprocal bandwidth, and a spectral density $\Psi(t, \omega)$ that changes in times short compared with its reciprocal bandwidth is not really physically meaningful.

Let us then turn to the long-time limit, when $g(t, \tau)$ becomes independent of its first argument. In that case we may make use of the asymptotic expressions for $\langle \hat{R}_3(t) \rangle + \frac{1}{2}$ and $\langle \hat{b}_s^\dagger(t) \rangle$ given by Eqs. (42) and (43) and substitute them in Eq. (90). We obtain for $\tau \geq 0$

$$\begin{aligned} \lim_{t \rightarrow \infty} g(t, \tau) &= \frac{\frac{1}{2}\Omega^2(1 + \theta^2)}{\beta^2(\frac{1}{2}\Omega^2/\beta^2 + 1 + \theta^2)^2} \\ &\quad - \frac{\frac{1}{8}\Omega^4}{\beta^2(\frac{1}{2}\Omega^2 + 1 + \theta^2)} \sum_{\substack{i=1 \\ i \neq j \neq k}}^3 \frac{(p_i + 2\beta)e^{p_i\tau}}{p_i(p_i - p_j)(p_i - p_k)}, \end{aligned} \quad (94)$$

which can be shown to reduce to our earlier Eq. (73) to order $(\Omega/\beta)^4$ in the weak-field limit $\Omega/\beta \ll 1$, when $\tau \geq 0$.

VIII. SPECTRAL DENSITY OF THE FLUORESCENCE

The spectral density $\Phi(\vec{r}, \omega)$ of the fluorescence observed at some point \vec{r} is, as before, equal to the Fourier transform with respect to τ of the function $\langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t + \tau) \rangle$ given by Eq. (57a) in the long-time limit. With the help of Eqs. (94) and (92) we then find, after some rearrangement of terms,

$$\begin{aligned} \Phi(\vec{r}, \omega) &= \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi\epsilon_0 c^2 r} \right)^2 \int_{-\infty}^{\infty} d\tau g(\infty, \tau) e^{i(\omega - \omega_1)\tau} \\ &= \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi\epsilon_0 c^2 r} \right)^2 \int_0^{\infty} d\tau g(\infty, \tau) e^{i(\omega - \omega_1)\tau} + \text{c.c.} \\ &= \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi\epsilon_0 c^2 r} \right)^2 \frac{\frac{1}{2}\Omega^2/\beta^2}{(\frac{1}{2}\Omega^2/\beta^2 + 1 + \theta^2)} \\ &\quad \times \left[\left(\frac{1 + \theta^2}{\frac{1}{2}\Omega^2/\beta^2 + 1 + \theta^2} \right) \pi \delta(\omega - \omega_1) \right. \\ &\quad \left. + \frac{\beta\Omega^2 [4\beta^2 + \frac{1}{2}\Omega^2 + (\omega_1 - \omega)^2]}{|C(i(\omega_1 - \omega))|^2} \right], \end{aligned} \quad (95)$$

where $C(p)$ is the cubic function of p defined by the left-hand side of Eq. (54),

$$\begin{aligned} C(p) &\equiv p^3 + 4\beta p^2 + (5\beta^2 + \beta^2\theta^2 + \Omega^2)p \\ &\quad + (2\beta^2 + 2\beta^2\theta^2 + \Omega^2)\beta. \end{aligned} \quad (96)$$

This spectral density $\Phi(\vec{r}, \omega)$ contains a δ -function contribution or sharp line at frequency ω_1 , corresponding to elastic scattering of the external field by the atom, and in general several peaks of finite width associated with fluorescence. For a small amount of detuning θ , the area under the sharp line at ω_1 becomes increasingly unimportant as the strength of the external field increases, i.e., as $\Omega/\beta \rightarrow \infty$. In the limit of an extremely strong external field, $\Omega/\beta \gg 1$ and $\Omega/\beta \gg \theta$, Eq. (95) leads to

$$\Phi(\vec{r}, \omega) = \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi\epsilon_0 c^2 r} \right)^2 \times \left(\frac{2\pi(1+\theta^2)}{\Omega^2/\beta^2} \delta(\omega - \omega_1) + \frac{\frac{1}{2}\beta}{(\omega - \omega_1)^2 + \beta^2} + \frac{\frac{3}{8}\beta}{(\omega - \omega_1 - \Omega)^2 + \frac{9}{4}\beta^2} + \frac{\frac{3}{8}\beta}{(\omega - \omega_1 + \Omega)^2 + \frac{9}{4}\beta^2} \right), \quad (97a)$$

in which there are three peaks of finite width, with the heights of the central peak and of the side peaks in the ratio 3:1. The side peaks are shifted roughly by the Rabi frequency Ω from the line center, and they reflect the Rabi oscillations of the atom. The phenomenon is sometimes called the ac Stark effect. On the other hand, in the limit of a strong field but with very strong detuning, $\Omega/\beta \gg 1$ and $\theta \gg \Omega/\beta$, we find from Eq. (95)

$$\Phi(\vec{r}, \omega) = \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi\epsilon_0 c^2 r} \right)^2 \times \left(\frac{\pi\Omega^2/\beta^2}{2\theta^2} \delta(\omega - \omega_1) + \frac{\frac{1}{4}\beta(\Omega/\beta\theta)^6}{(\omega - \omega_1)^2 + 4\beta^2} + \frac{\frac{1}{8}\beta(\Omega/\beta\theta)^4}{(\omega - \omega_1 - \beta\theta)^2 + \beta^2} + \frac{\frac{1}{8}\beta(\Omega/\beta\theta)^4}{(\omega - \omega_1 + \beta\theta)^2 + \beta^2} \right). \quad (97b)$$

Equations (95) and (97) are in agreement with the results obtained by Mollow,^{5,16} Milonni,¹³ Agarwal,⁶ Carmichael and Walls,¹⁵ and the latest calculations of Smithers and Freedhoff,⁴⁴ but do not agree with the conclusions reached earlier by Stroud⁸ and Smithers and Freedhoff¹⁴ on the basis of different assumptions. The last two treatments were limited to a finite number of emitted photons, whereas no such limitations were imposed here, and some difference in the conclusions is therefore to be expected. This point has also been discussed by Carmichael and Walls¹⁵ and Smithers and Freedhoff.⁴⁴

Some graphs of the form of $\Phi(\vec{r}, \omega)$ for various field strengths and various detunings are illustrated in Figs. 5–7, but with the elastically scattered δ -function contribution omitted. It will be seen that the effect of increasing detuning (i.e., increasing θ) is to lower the absolute heights of the peaks, but that the height ratio of the side peaks to the central peak of finite width increases. This is also evident by inspection of Eq. (97b). Curves of the general shape shown in Figs. 5–7 have been obtained by Schuda, Stroud, and Hercher,³⁴ and more recently by Wu, Grove and Ezekiel³⁴ from direct measurement of the resonance fluorescence in an atomic beam of sodium.

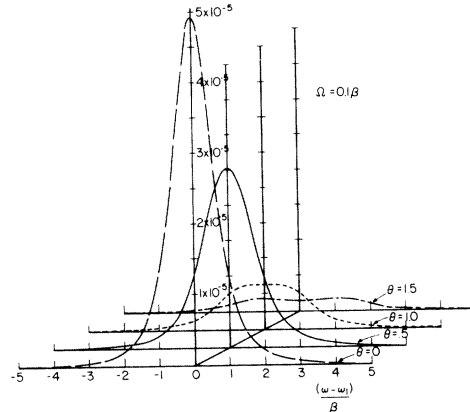


FIG. 5. Spectral density $\Phi(\vec{r}, \omega)(\omega_0 \mu \sin \psi / 4\pi\epsilon_0 c^2 r)^{-2}$ of the fluorescence at position \vec{r} in the long-time limit, for various detunings $\theta = (\omega_1 - \omega_0 + \gamma)/\beta$ of the exciting field. The Rabi frequency $\Omega = 0.1\beta$. The elastically scattered δ -function contribution is not shown.

On the other hand, in these experiments the height ratio of side peaks to central peak is found to become progressively smaller as the detuning increases, in apparent contradiction with Figs. 5–7. However, it must be remembered that the figures do not include the elastically scattered component of the light, which contributes to the measurements. Recall that the exciting field is not strictly monochromatic in the experiments; even if it were, this would show up as a peak of finite width because of the finite spectral resolution. If we calculate the ratio of the areas of the central peak to the side peaks from Eq. (97b), and include the elastically scattered contribution, we find that

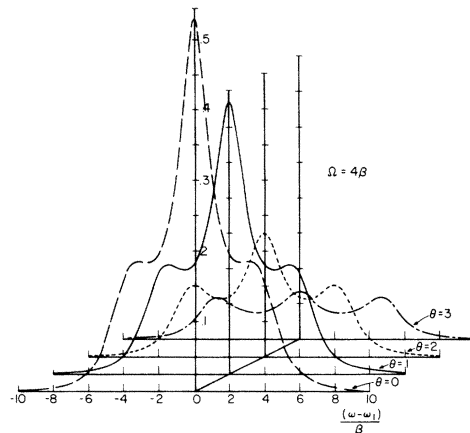


FIG. 6. Spectral density $\Phi(\vec{r}, \omega)(\omega_0 \mu \sin \psi / 4\pi\epsilon_0 c^2 r)^{-2}$ of the fluorescence at position \vec{r} in the long-time limit, for various detunings $\theta = (\omega_1 - \omega_0 + \gamma)/\beta$ of the exciting field. The Rabi frequency $\Omega = 4\beta$. The elastically scattered δ -function contribution is not shown.

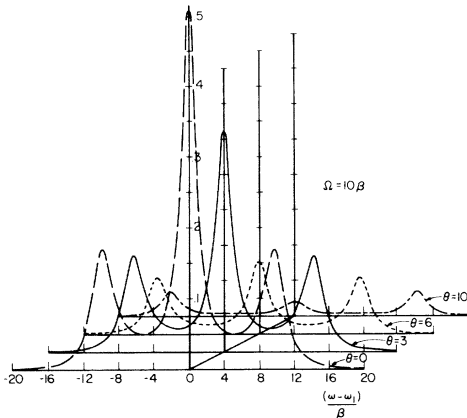


FIG. 7. Spectral density $\Phi(\vec{r}, \omega)(\omega_0 \mu \sin \psi / 4 \pi \epsilon_0 c^2 \gamma)^{-2}$ of the fluorescence at position \vec{r} in the long-time limit, for various detunings $\theta = (\omega_1 - \omega_0 + \gamma) / \beta$ of the exciting field. The Rabi frequency $\Omega = 10\beta$. The elastically scattered δ -function contribution is not shown.

it increases as $4(\beta\theta/\Omega)^2$ with increasing detuning θ . When $\Omega/\beta = \theta \gg 1$, the ratio of the areas given by Eq. (95) is about 6:1, in rough agreement with the measurements. The narrowing of the spectral line in a weak field, because of the presence of the elastically scattered component, has also been observed by Gibbs and Venkatesan.⁴⁵ Figure 8 shows a comparison of the theoretical spectral distribution with some results obtained by Wu *et al.*,³⁴ after convolution of the theoretical curve with an instrumental profile of Gaussian shape and of width $2\beta \approx 12$ MHz, which gives best agreement. The validity of the theory therefore seems to be confirmed. However, we should point out that

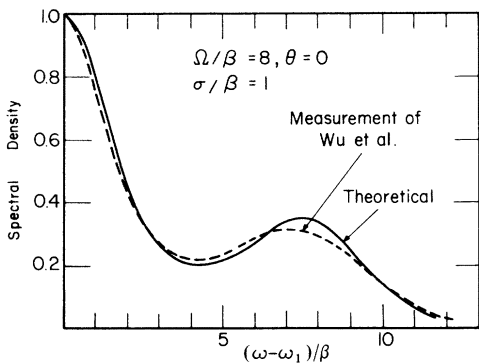


FIG. 8. Comparison of the theoretical spectral density $\Phi(\vec{r}, \omega)$ for $\Omega/\beta = 8$ and $\theta = 0$, after convolution with a Gaussian instrumental profile of standard deviation $\sigma = \beta$, with experimental results of Wu, Grove, and Ezekiel (Ref. 34). Both curves have been normalized to unity, and the experimental curve has been smoothed slightly.

Wu *et al.* claimed an instrumental resolution of better than 3 MHz, which does not give good agreement, and that the two-level atom approximation may not be an adequate approximation for the transitions occurring in their atomic beam.

If spectral measurements of the fluorescence that are free from the contribution of the elastically scattered component could be carried out, they should result in the spectral distributions shown in Figs. 5–7, and provide a somewhat cleaner test of the theory. An interesting possibility for such measurements is provided by the heterodyne detection technique, in which the light from the atom is allowed to beat against a portion of the exciting beam, and the beats occurring in the output of the photoelectric detector are analyzed. The photoelectric variations in general reflect only the broadening of the fluorescence relative to the exciting beam.

A superficial examination of the analysis leading from Eqs. (75) and (76) to the spectral distribution given by Eq. (95) may suggest that the operator character of the free field $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ played very little role in the calculation, because of the zero commutator in Eq. (83b). That is why some calculations in which the operator character of $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ was effectively ignored yielded essentially the same answers.^{11,13} This is in apparent contradiction with the conclusions reached in Sec. V, where it was found that the operator character of $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ was essential for a finite spectral width. Indeed, we concluded there that the finite linewidth could, in a sense, be attributed to the fluctuations of the quantum field, although the fluctuations of the source and the field are inseparable in a consistent treatment. But the contradiction is only an apparent one, for $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ cannot be c numbers if the symmetry property (92) of the correlation function $g(t, \tau)$, which is needed for a real spectral density, is to be satisfied. By repeating the calculation leading to Eq. (90), but with $\tau < 0$, we can readily show that a solution decaying exponentially with $|\tau|$ cannot be obtained when $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ are c numbers and are replaced by their eigenvalues. The operator character of the free field and the commutation relations obeyed by $\hat{A}_{S_i}^{(+)}(0, t)$ and $\hat{A}_{S_i}^{(-)}(0, t)$ are therefore essential to account for the full behavior of $g(t, \tau)$ and of the spectral density of the fluorescence.

IX. INTENSITY CORRELATION OF THE FLUORESCENT LIGHT

So far our discussion has been limited to second-order correlation functions of the field and of the corresponding atomic operators. However, as is

well known, it is possible to make correlation measurements with photoelectric detectors, in which the arrival times of photoelectric pulses at some point \vec{r} at two different times t and $t + \tau$ are registered directly. As has been shown by Glauber,³³ the joint probability density of photodetection $P_2(\vec{r}, t, t + \tau)$ is proportional to the normally ordered correlation function of the fourth order in the field, or of the second order in the light intensity, of the form

$$\begin{aligned} \Gamma^{(2,2)}(\vec{r}, t, \tau) &= \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi \epsilon_0 c^2 r} \right)^4 \left\langle \hat{b}_s^\dagger \left(t - \frac{r}{c} \right) \hat{b}_s^\dagger \left(t - \frac{r}{c} + \tau \right) \hat{b}_s \left(t - \frac{r}{c} + \tau \right) \hat{b}_s \left(t - \frac{r}{c} \right) \right\rangle \\ &= \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi \epsilon_0 c^2 r} \right)^4 \left[\left\langle \hat{b}_s^\dagger \left(t - \frac{r}{c} \right) \hat{R}_3 \left(t - \frac{r}{c} + \tau \right) \hat{b}_s \left(t - \frac{r}{c} \right) \right\rangle + \frac{1}{2} \left\langle \hat{R}_3 \left(t - \frac{r}{c} \right) \right\rangle + \frac{1}{4} \right]. \end{aligned} \quad (98)$$

When $\tau < 0$, the time $t + \tau$ precedes t , and the joint probability for photodetection leads to a correlation function of the form $\Gamma^{(2,2)}(\vec{r}, t - |\tau|, |\tau|)$. Instead of explicitly studying the symmetry properties of $\Gamma^{(2,2)}(\vec{r}, t, \tau)$ with respect to τ , we shall therefore be content to show that $\Gamma^{(2,2)}(\vec{r}, t, \tau)$ given by Eq. (99) becomes independent of the second argument for large times t . Once again it will be quantum fluctuations of the field that cause the correlations to die out smoothly as $\tau \rightarrow \infty$.

In order to calculate $\Gamma^{(2,2)}(\vec{r}, t, \tau)$, we again make use of the two integral relations (75) and (76). It is convenient to define the two atomic correlation functions

$$\mathcal{K}(t, \tau) \equiv \langle \hat{b}_s^\dagger(t) \hat{R}_3(t + \tau) \hat{b}_s(t) \rangle, \quad (100)$$

$$\mathcal{F}(t, \tau) \equiv \langle \hat{b}_s^\dagger(t) \hat{b}_s^\dagger(t + \tau) \hat{b}_s(t) \rangle e^{i(\omega_0 - \omega_1)(t + \tau)} e^{i\phi}, \quad (101)$$

which, as we shall see, become independent of both t and ϕ in the long-time limit. From Eq. (76) we then obtain by direct multiplication, with the help of Eqs. (83b), (34), and (41),

$$\begin{aligned} \mathcal{K}(t, \tau) &= -\frac{1}{2} \langle \hat{R}_3(t) \rangle - \frac{1}{4} \\ &\quad - \frac{\Omega}{2} \int_0^\tau dt' [\mathcal{F}(t, t') + \mathcal{F}^*(t, t')] e^{2\beta(t' - \tau)}, \end{aligned} \quad (102)$$

and from Eq. (75),

$$\mathcal{F}(t, \tau) = \Omega \int_0^\tau dt' \mathcal{K}(t, t') e^{(\beta + i\theta)(t' - \tau)}. \quad (103)$$

We now use Eq. (103) to substitute for $\mathcal{F}(t, t')$ and $\mathcal{F}^*(t, t')$ in Eq. (102):

$$\begin{aligned} P_2(\vec{r}, t, t + \tau) &\propto \langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_j^{(-)}(\vec{r}, t + \tau) \hat{E}_j^{(+)}(\vec{r}, t + \tau) \hat{E}_i^{(+)}(\vec{r}, t) \rangle \\ &\equiv \Gamma^{(2,2)}(\vec{r}, t, \tau), \end{aligned} \quad (98)$$

for $\tau \geq 0$. If we choose the point \vec{r} to lie in the far field outside the exciting beam, as before, so that Eq. (57b) is satisfied, and make use of Eq. (16) for the far field together with the commutation relation (84), we obtain, for $\tau \geq 0$,

$$\begin{aligned} \mathcal{K}(t, \tau) &= -\frac{1}{2} \langle \hat{R}_3(t) \rangle - \frac{1}{4} \\ &\quad - \frac{\Omega^2 e^{-2\beta\tau}}{2} \int_0^\tau dt' \int_0^{t'} dt'' \mathcal{K}(t, t'') \\ &\quad \times [e^{\beta(1+i\theta)t''} e^{\beta(1-i\theta)t'} + \text{c.c.}]. \end{aligned}$$

After integrating once by parts we arrive at the following integral equation:

$$\mathcal{K}(t, \tau) = -\frac{1}{2} \langle \hat{R}_3(t) \rangle - \frac{1}{4} + \int_0^\tau dt' \mathcal{K}(t, t') K(\tau - t'), \quad (104)$$

where $K(\tau)$ is the integral kernel that we encountered before in Eqs. (46) and (88), in the course of evaluating $\langle \hat{R}_3(\tau) \rangle$ and $g(t, \tau)$. Each of the problems of determining $\langle \hat{R}_3(\tau) \rangle$, $g(t, \tau)$, and $\mathcal{K}(t, \tau)$ therefore leads to the same integral equation with the same kernel, but with a different inhomogeneous term, as is also suggested by the application of the quantum-regression theorem^{16, 42} to the problem. We have already shown that $-\frac{1}{2} \langle \hat{R}_3(t) \rangle - \frac{1}{4}$ becomes independent of both t and ϕ in the long-time limit, and since $K(\tau)$ does not depend on t and ϕ , both $\mathcal{K}(t, \tau)$ and $\mathcal{F}(t, \tau)$ must also become independent of t and ϕ as $t \rightarrow \infty$.

If we examine the inhomogeneous term given by Eq. (45) in the integral equation (36) for $\langle \hat{R}_3(\tau) \rangle$, we observe that it reduces to the constant $-\frac{1}{2}$ in the special case in which the atom starts in the lower or ground state at time zero. Let us denote the corresponding solution [which is given by Eq. (53) in general] at time τ by $\langle \hat{R}_3(\tau) \rangle_G$, where G indicates the atomic ground state. The integral equations for $\mathcal{K}(t, \tau)$ and $\langle \hat{R}_3(\tau) \rangle_G$ are then almost identical, except that the inhomogeneous term is larger by the factor $\langle \hat{R}_3(t) \rangle + \frac{1}{2}$ in the former case. Inspection of Eq. (50) then shows that the solutions

for $\mathcal{K}(t, \tau)$ and $\langle \hat{R}_3(\tau) \rangle_G$ must be similar also, except for the factor $\langle \hat{R}_3(t) \rangle + \frac{1}{2}$, so that we may write directly

$$\mathcal{K}(t, \tau) = \langle \hat{R}_3(\tau) \rangle_G [\langle \hat{R}_3(t) \rangle + \frac{1}{2}]. \quad (105)$$

From Eq. (99) we then have for any atomic state, and for the coherent state of the field with eigenvalue given by Eq. (41),

$$\begin{aligned} \Gamma^{(2,2)}(\vec{r}, t+r/c, \tau) \\ = \left(\frac{\omega_0^2 \mu \sin \psi}{4\pi \epsilon_0 c^2 r} \right)^4 [\langle \hat{R}_3(\tau) \rangle_G + \frac{1}{2}] [\langle \hat{R}_3(t) \rangle + \frac{1}{2}], \end{aligned} \quad (106)$$

where $\langle \hat{R}_3(t) \rangle$ and $\langle \hat{R}_3(\tau) \rangle_G$ are both given by Eq. (53). If we recall that $\langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t) \rangle$ is proportional to the photoelectric detection probability density $P_1(\vec{r}, t)$ at position \vec{r} at time t , and make use of Eqs. (40), (98), and (106), we can express the joint probability density $P_2(\vec{r}, t, t+\tau)$ at two times t and $t+\tau$ in the form of a product

$$P_2(\vec{r}, t, t+\tau) = P_1(\vec{r}, \tau+r/c) P_1(\vec{r}, t), \quad t > r/c. \quad (107)$$

This is a solution of remarkable formal simplicity. In the special case of no detuning ($\theta=0$) and for an atom initially in the ground state, it agrees with the solution recently obtained by Carmichael and Walls,¹⁵ although our Eq. (106) holds more generally. We note that since $\langle \hat{R}_3(0) \rangle_G = -\frac{1}{2}$, $\Gamma^{(2,2)}(\vec{r}, t, \tau)$ vanishes identically for $\tau=0$, which is a reflection of the fact that the two-level atom cannot emit two photons simultaneously. As τ increases $\Gamma^{(2,2)}(\vec{r}, t, \tau)$ generally may pass through a maximum, and the same curves illustrated in Figs. 1–3 that describe the evolution of the light intensity with τ also describe the form of the intensity correlation function $\Gamma^{(2,2)}(\vec{r}, t, \tau)$. As $\tau \rightarrow \infty$, $\langle \hat{R}_3(\tau) \rangle$ becomes independent of the initial atomic state, and for sufficiently large t and τ we then have

$$P_2(\vec{r}, t, \tau) \simeq P_1^2(\vec{r}, \infty). \quad (108)$$

This shows that in the steady state photoelectric detections or intensity fluctuations separated by sufficiently-long-time intervals become independent, as expected, because the correlations have died out. Evidently there is an underlying random process at work here, which we can again identify as the fluctuation of the quantum field.

Equation (106) provides a simple and interesting example of a quantum field whose intensity fluctuations may have a negative correlation.⁴⁶ Examples of such fields have recently been discussed by Stoler.⁴⁷ If we denote the expectation of the light intensity $\langle \hat{E}_i^{(-)}(\vec{r}, t) \hat{E}_i^{(+)}(\vec{r}, t) \rangle$ by $\langle \hat{I}(\vec{r}, t) \rangle$, and

introduce a normalized intensity correlation function defined by

$$\lambda(\vec{r}, t, \tau) \equiv \frac{\Gamma^{(2,2)}(\vec{r}, t, \tau)}{\langle \hat{I}(\vec{r}, t) \rangle \langle \hat{I}(\vec{r}, t+\tau) \rangle} - 1, \quad t > r/c, \quad (109)$$

we find from Eq. (106)

$$\lambda(\vec{r}, t, \tau) = \frac{\langle \hat{I}(\vec{r}, \tau+r/c) \rangle_G}{\langle \hat{I}(\vec{r}, t+\tau) \rangle} - 1, \quad t > r/c. \quad (110)$$

For small τ , $\lambda(\vec{r}, t, \tau)$ is negative, because the detection of one photon at time t makes the detection of another one at time $t+\tau$ less likely. However, $\lambda(\vec{r}, t, \tau)$ may become positive as τ increases, and it always tends to zero as $\tau \rightarrow \infty$. The intensity correlation function therefore reflects quantum features of the field both in its behavior for small τ and for larger τ , and its measurement would appear to be a very direct test of some of these quantum properties.

X. SUMMARY

We have examined the interaction of a two-level atom with the quantized electromagnetic field, without the use of perturbation theory, without the introduction of classical fields or classical factorization conditions for the states, and without assumptions about loss of memory as the interaction proceeds. We find that it is possible to account for certain transient processes, such as those in spontaneous emission in the vacuum, without explicitly invoking the quantum properties of the free field. It is sufficient that the total field $\hat{A}_i^{(+)}(0, t)$ and $\hat{A}_i^{(-)}(0, t)$ commute with atomic operators at the same time. The quantum properties of the field are generally more manifest in the presence of an exciting field when a steady state is reached, as the effects of quantum fluctuations are then clearly distinguished from transient effects. Indeed, the fact that a quasistationary state is reached is itself a reflection of the quantum fluctuations. We have presented a general solution for the growth of the fluorescent light intensity, for an arbitrary state of the atom and for an arbitrary initial coherent state of excitation of the field. We have calculated two-time correlation functions of the second order and of the fourth order in the field, and have shown that the quantum fluctuations of the free field play an essential role in causing correlations to die out. Curves have been presented showing the spectral density of the fluorescent light under various conditions of excitation and detuning. The two-time intensity correlation function $\Gamma^{(2,2)}(\vec{r}, t, \tau)$, which is directly

accessible to measurement with a photodetector, reveals a further explicit quantum feature in that it starts from zero for zero separation of the two times. This reflects the fact that the two-level atom can emit only one photon at one time, and that the quantum field, in turn, can produce only one photoemission at one time. From the point of view of testing QED, this would appear to be one of the most attractive areas for experiment.

APPENDIX: EVALUATION OF THE FREE-FIELD COMMUTATOR

We consider the commutator

$$[\mu_i \hat{A}_{Si}^{(+)}(0, t_1), \mu_j \hat{A}_{Sj}^{(-)}(0, t_2)]$$

$$\begin{aligned} [\vec{\mu} \cdot \hat{\mathbf{A}}_S^{(+)}(0, t_1), \vec{\mu} \cdot \hat{\mathbf{A}}_S^{(-)}(0, t_2)] &= \frac{1}{L^3} \sum_{\mathbf{k}, s} \left(\frac{\hbar}{2\omega\epsilon_0} \right) \mu_i \mu_j (\vec{\epsilon}_{\mathbf{k}, s})_i (\vec{\epsilon}_{\mathbf{k}, s}^*)_j e^{i(\omega_0 - \omega)(t_1 - t_2)} \\ &= \frac{1}{L^3} \sum_{\mathbf{k}} \left(\frac{\hbar}{2\omega\epsilon_0} \right) \mu_i \mu_j \left(\delta_{ij} - \frac{k_i k_j}{k^2} \right) e^{i(\omega_0 - \omega)(t_1 - t_2)} \\ &= \frac{\hbar}{2\epsilon_0 c} \frac{1}{(2\pi)^3} \int \frac{d^3 k}{k} \left(\mu^2 - \frac{(\vec{\mu} \cdot \vec{k})^2}{k^2} \right) e^{i(\omega_0 - ck)(t_1 - t_2)}, \end{aligned} \quad (A3)$$

when we replace the sum by an integral in the usual way, and put $\omega = ck$. The integral is most easily evaluated in polar coordinates, if we choose the z axis in the direction of $\vec{\mu}$. We then have

$$\begin{aligned} [\vec{\mu} \cdot \hat{\mathbf{A}}_S^{(+)}(0, t_1), \vec{\mu} \cdot \hat{\mathbf{A}}_S^{(-)}(0, t_2)] &= \frac{\hbar \mu^2}{2\epsilon_0 c (2\pi)^3} \int_0^\infty k dk e^{i(\omega_0 - ck)(t_1 - t_2)} \\ &\quad \times \int_0^\pi \sin^3 \theta d\theta \int_0^{2\pi} d\phi \\ &= \frac{\hbar \mu^2}{6\pi^2 \epsilon_0 c^3} \int_{-\infty}^{\omega_0} d\omega' (\omega_0 - \omega') e^{i\omega'(t_1 - t_2)}. \end{aligned} \quad (A4)$$

The first term can be well approximated under a

that is encountered under the integral in Eq. (58) and elsewhere, when matrix elements are to be evaluated. Since $A_{Si}^{(+)}(\vec{r}, t)$ is a free-field operator, it can be given a mode expansion in plane waves in the usual form,

$$\hat{A}_{Si}^{(+)}(\vec{r}, t) = \frac{1}{L^{3/2}} \sum_{\mathbf{k}, s} \left(\frac{\hbar}{2\omega\epsilon_0} \right)^{1/2} \hat{a}_{\mathbf{k}, s} \vec{\epsilon}_{\mathbf{k}, s} e^{i[\vec{k} \cdot \vec{r} + (\omega_0 - \omega)t]} \quad (A1)$$

for $\omega = ck$, in which the operators $\hat{a}_{\mathbf{k}, s}$ and their conjugates $\hat{a}_{\mathbf{k}, s}^\dagger$ obey the commutation relations

$$\begin{aligned} [\hat{a}_{\mathbf{k}, s}, \hat{a}_{\mathbf{k}', s'}] &= 0 = [\hat{a}_{\mathbf{k}, s}^\dagger, \hat{a}_{\mathbf{k}', s'}^\dagger], \\ [\hat{a}_{\mathbf{k}, s}, \hat{a}_{\mathbf{k}', s'}^\dagger] &= \delta_{ss'} \delta_{\mathbf{k}\mathbf{k}'}. \end{aligned} \quad (A2)$$

With the help of Eqs. (A1) and (A2) we then have

t_1, t_2 double integral by a δ function, and the second term by a derivative of a δ function, provided the t_1, t_2 integrals extend over intervals that are very great compared with $1/\omega_0$, and provided the rest of the integrand contains only functions that vary slowly compared with optical oscillations. Accordingly, we may write, with the help of Eq. (18),

$$\begin{aligned} [\vec{\mu} \cdot \hat{\mathbf{A}}_S^{(+)}(0, t_1), \vec{\mu} \cdot \hat{\mathbf{A}}_S^{(-)}(0, t_2)] \\ \approx 2\beta (\hbar/\omega_0)^2 [\delta(t_1 - t_2) + (i/\omega_0)\delta'(t_1 - t_2)]. \end{aligned} \quad (A5)$$

When applied to a slowly varying test function, the contribution of the second term will evidently be much smaller than the first, and can be discarded.

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