Coherent intermittency in the resonant fluorescence of a multilevel atom

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Time correlations in the fluorescence of a multilevel atom are studied in terms of the quantum statistics which describe the emission of the next photon. The *next* photon equations are substantially simpler than the *a* photon equations (or so-called optical Bloch equations) yet they contain phase information lost in the averaging procedure that yields the *a* photon equations. Employing the *next* photon equations we present a quantum formalism in which the time development of the wave function of the atom is dramatically modified by the observation of a dark period. This "collapse" of the quantum state due to measurements with a null result (such as the failure to record events in a photodetector) is the cause of intermittent atomic fluorescence even when the exciting field is arbitrarily coherent. The *next* photon and *a* photon formalisms are contrasted and applied to photon antibunching. The phenomenon of photon bunching which characterizes sideband correlations is also calculated. Observable effects particular to the coherent intermittency are emphasized.

I. INTRODUCTION AND SUMMARY

An atom driven by an external source of radiation will emit fluorescent photons with frequencies determined by its energy levels. In this paper we develop the theory of the correlations in time of these scattered photons. The correlations are determined by two fundamentally different types of measurements that can be made on the illuminated atom. First, one can observe the actual scattered photon. Second, one can make a null measurement; that is, a "switched-on" photodetector can observe the absence of scattered photons for some period of time. The first type of measurement is Markovian and destroys past history in the atomic wave function, whereas the second (or null-type) measurement leads to a non-Markovian change that can preserve phase information. In a multilevel atom with widely disparate lifetimes the ability to carry out null measurements leads to the possibility of observing intermittent periods of darkness in the fluorescence, during which the atomic state wave function is coherent.

To develop these ideas we consider the arrangement proposed by Dehmelt[1] and shown here in Fig. 1. In this so-called "V" system $|0\rangle$ is the atomic ground state and $|1\rangle$ and $|2\rangle$ are excited states. The lifetime of level $|1\rangle$ is a typical atomic lifetime of about 10^{-9} s ($\sim 1/\beta_1$), whereas the lifetime of $|2\rangle$ is very long, and can be on the order of 1 s ($\sim 1/\beta_2$). In the Dehmelt "shelving" scheme the presence of the "forbidden" transition $|2\rangle \leftrightarrow |0\rangle$ would affect the fluorescence from the $|1\rangle \leftrightarrow |0\rangle$ transition when both transitions are driven at resonance by external electric fields $\mathbf{E}_1$ and $\mathbf{E}_2$. That is, with $\mathbf{E}_2 = 0$ but E_1 very large photons will be emitted at a rate $\frac{1}{2}\beta_1$ from the $|1\rangle \leftrightarrow |0\rangle$ transition. If, however, E_2 is also turned on, the atom might at times be "shelved" in $|2\rangle$ so that the fluorescence is turned off for a period of time given approximately by $1/\beta_2$. This intermittency and the resulting telegraph [see Fig. 2(a)] in the strong transition emission of a single atom should be sufficiently intense to be seen with the unaided eye. According to Dehmelt this scheme should provide a means whereby the single quantum jumps $|0\rangle \leftrightarrow |2\rangle$ can be detected with great certainty, as the strong emission is effectively an amplifier for the shelving.

From a strictly quantum-mechanical point of view one is tempted to wonder whether the picture of the atom shelved into $|2\rangle$ is a bit too classical. For *coherent* illumination, should not the atom in fact be in a superposition of $|0\rangle$, $|1\rangle$, $|2\rangle$, so that there is at all times a strong probability for emission from $|1\rangle$ to $|0\rangle$? From this point of view photons from the $|2\rangle$-$|0\rangle$ transition could occasionally appear but intermittency would be absent, and the emission would be characterized by Fig. 2(b).

We will show that for steady, arbitrarily coherent illumination intermittency is required[2] yet during the dark periods the atom is indeed in a coherent superposition of $|0\rangle$, $|1\rangle$, and $|2\rangle$.[3] The essential manifestation of this superposition is that, as indicated in Fig. 2(a), a large proportion of the dark periods must end with the emission of a $|1\rangle \rightarrow |0\rangle$ photon as opposed to a $|2\rangle \rightarrow |0\rangle$ photon.

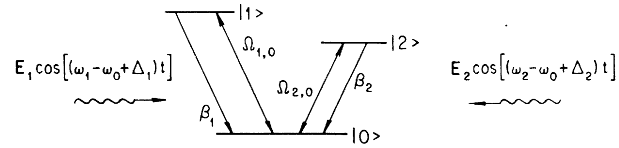

FIG. 1. V system or three-level atom where all transitions take place between the excited states $|1\rangle$, $|2\rangle$, and the ground state $|0\rangle$. The externally imposed fields $\mathbf{E}_1$ and $\mathbf{E}_2$ are approximately tuned to the strong $|0\rangle$-$|1\rangle$ and "forbidden" ($|0\rangle$-$|2\rangle$) transitions. The Rabi flopping frequencies are designated by $\Omega_{1,0}$ and $\Omega_{2,0}$. The spontaneous decay rates are β_1 and β_2.

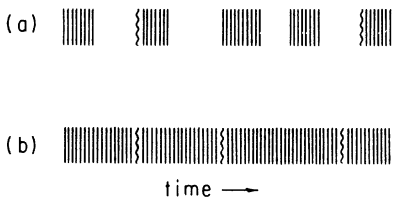

FIG. 2. Examples of fluorescence as a function of time for the V system. Each straight line represents many ($\sim 10^8$) scattered photons from the $|1\rangle$-$|0\rangle$ transition, whereas each wavy line represents a single scattered photon from the $|2\rangle$-$|0\rangle$ transition. The unitary interpretation of a null measurement yields (b) (i.e., no intermittency). The interpretation of a null measurement as collapsing the wave function of atoms plus field into the state with no outgoing scattered photons yields (a) (i.e., the quantum telegraph). Note that some dark periods end with the emission of a $|1\rangle$-$|0\rangle$ photon and not a $|2\rangle$-$|0\rangle$ photon. This is an indication of the coherence of the V system dark periods.

Indeed, if Γ is the percentage of dark periods which ends with the emission of a strong transition photon, then the percentage of time p_D that the V system fluorescence is dark is given by (see Sec. VI)

$$p_D=\frac{\Gamma}{2+\Gamma}\,, \tag{1.1}$$

in the limit where E_1 is so large that the induced transition rate (or so-called Rabi flopping frequency) $|\Omega_{1,0}|$ is greater than β_1. For steady coherent illumination the intermittency vanishes in the limit where all dark periods end with a $|2\rangle \to |0\rangle$ emission. Thus the dark period are not due to the absorption of a photon from the field E_2 that is tuned to the forbidden transition. Instead, during a dark period, the atom is in a time-dependent superposition that does not radiate. If additional forbidden transitions are available (say, to a level $|3\rangle$) then the observation of a dark period can put the atom into a long-term slowly changing superposition of $|2\rangle$ and $|3\rangle$. In view of these possibilities we develop the theory of resonance fluorescence in a general multilevel atom. Detailed comparisons of this theory with experimental observations[4–7] are contained in a separate paper.[8]

To motivate the role of null observations in the coherent intermittency, consider first the simplified problem of the unitary development of a three-level system driven by an electric field of the form

$$\mathbf{E}=\mathbf{E}_1\cos(\omega_1-\omega_0+\Delta_1)t+\mathbf{E}_2\cos(\omega_2-\omega_0+\Delta_2)t\ . \tag{1.2}$$

The wave function of the atom can therefore be written as

$$|\psi\rangle=\sum_{j=0}^{2}\bar{C}_j e^{-i\omega_j t}|j\rangle\ . \tag{1.3}$$

Schrödinger's equation for the atom is

$$i\hbar\frac{d\bar{C}_i}{dt}=\sum_{j=0}^{2}H_{ij}\bar{C}_j\,, \tag{1.4}$$

where H is Hermitian and, in the rotating-wave approximation (RWA)

$$H_{10}^{*}=H_{01}=-\hbar\Omega_{1,0}^{*}e^{i\Delta_1 t}\,,$$

and

$$H_{02}=-\hbar\Omega_{2,0}^{*}e^{i\Delta_2 t}=H_{20}^{*}\ . \tag{1.5}$$

All other elements of H are zero, and the Rabi flopping frequency is

$$\Omega_{i,0}=\mu_{i,0}E_i/2\hbar\,, \tag{1.6}$$

where $\mu_{i,0}$ are the dipole transition matrix elements between the ground state and the excited states. For the V system we have taken $\mu_{1,2}=0$.

For this closed three-level system $|\bar{C}_0|^2+|\bar{C}_1|^2+|\bar{C}_2|^2=1$. Given that the atom was in the ground state at $t=0$ the amplitudes to be in $|1\rangle$, $|2\rangle$ at time t are, to leading order in $|\Omega_{2,0}|/|\Omega_{1,0}| \ll 1$,

$$\bar{C}_1(t)=\tfrac{1}{2}[\exp(i|\Omega_{1,0}|t)\cos(|\Omega_{2,0}|t/\sqrt{2}) -\exp(-i|\Omega_{1,0}|t)]\,, \tag{1.7}$$

$$\bar{C}_2(t)=\frac{\sqrt{2}}{2}i\sin(|\Omega_{2,0}|t/\sqrt{2}) +\frac{\Omega_{2,0}}{4\Omega_{1,0}}[\cos(|\Omega_{2,0}|t/\sqrt{2})-\exp(-2i|\Omega_{1,0}|t)]\,, \tag{1.8}$$

where we have taken $\Delta_2=|\Omega_{1,0}|$ and $\Delta_1=0$.

For the closed system (1.4) the atom returns to the ground state by clocklike induced oscillations determined by $\Omega_{1,0}$ and $\Omega_{2,0}$. When an actual atom is in level $|1\rangle$ or $|2\rangle$ there is also a probability per unit time of spontaneously emitting a photon and returning to the ground state. If the process of driven time development and emission are separated then the chance of seeing a dark period is incredibly small. Specifically, if between photon observations the atom is regarded as developing according to (1.4), (1.7) and (1.8) with the expected rate of strong and forbidden emission given by $\beta_1|\bar{C}_1|^2$ and $\beta_2|\bar{C}_2|^2$, then the probability that a dark period longer than T will commence after the detection of a photon is

$$W(T)=\exp(-\beta_1 T/2)\ . \tag{1.9}$$

In this picture the observation of a photon resets the atom to the ground state[9] so that in (1.7) and (1.8) the time t is always measured from the previous emission. The probability of a strong emission between t and $t+dt$ is then about $(\beta_1/2)dt$ because the probability to be in $|1\rangle$ quickly builds up to approximately $\frac{1}{2}$. Even though there are β_1 attempts or resets per second the probability of seeing a dark period begin in the time T is

$$\beta_1 T\exp(-\beta_1 T/2)\,,$$

which is still fantastically small for, say, $\beta_1 T=10^7$. So if the atom is interpreted as being in a superposition (1.3) during the radiationless periods between photon observations, the fluorescence would be described by Fig. 2(b).

Including the possibility that the wave function of atom plus field can be modified through the observation of no scattered photons yields a probability of darkness tremendously greater than that given by (1.9). The resultant dark time is determined by (1.1) and the value of Γ calculated in Sec. VI as

$$\Gamma=\left[1+\frac{\beta_1\beta_2}{4|\Omega_{2,0}|^2}\right]^{-1} . \quad (1.10)$$

The prediction[2] that coherent illumination would in fact lead to a telegraph has apparently been verified by experiment.[4,6,7] Of course no experiments have used a field E_2 with a bandwidth small compared to β_2. So it is still possible for a strict phenomenologist to question whether such a futuristic source would eliminate the intermittency. As the intermittency is a fundamentally coherent phenomenon, we claim that an arbitrarily coherent **E** field will still yield this effect.[3]

Restating the key issue we note that in the period between photon detections no energy is scattered by the driven atom. If such a period of observed null emission is interpreted as indicating that the system is developing in a unitary fashion (1.4), then the distribution (1.9) will prevail and there will be no intermittency. Instead, our formulation of orthodox quantum mechanics will interpret such a null measurement as "collapsing" the wave function of atom plus field into a state of no scattered photons.[3]

In order to apply this interpretation of null measurement we introduce the wave function of atom plus scattered field

$$|\Psi\rangle=\sum_{i,\{n\}} C_{i\{n\}}|i,\{n\}\rangle , \quad (1.11)$$

where $\{n\}=n_{k_1},n_{k_2},\ldots$ is the number of scattered photons in each of the outgoing states labeled by wave numbers k_i. Following Kimble and Mandel,[9] and Mollow,[10] the model Hamiltonian for an atom coupled to the electromagnetic field is taken as

$$H=H_0+H_I , \quad (1.12)$$

with

$$H_0=\tfrac{1}{2}\int\left[\epsilon_0E^2+\frac{1}{\mu_0}B^2\right]d^3r+\sum_i \hbar\omega_i\Lambda_{ii} , \quad (1.13)$$

$$H_I=-\frac{i}{2}\sum_{i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\cdot[\mathbf{A}^-(0,t)\Lambda_{ij}(t)+\Lambda_{ij}(t)\mathbf{A}^+(0,t)] . \quad (1.14)$$

Here $\mathbf{A}^-$ and $\mathbf{A}^+$ are the negative and positive frequency components of the vector potential **A**, and

$$\Lambda_{ij}(0)=|i\rangle\langle j|=\sum_{\{n\}}\Lambda_{ij}^{\{n\}}(0) , \quad (1.15)$$

where

$$\Lambda_{ij}^{\{n\}}(0)=|i,\{n\}\rangle\langle\{n\},j| . \quad (1.16)$$

In Sec. II the Heisenberg equations for $\Lambda_{ij}^{\{n\}}$ are developed and the current **J** and scattered electric field $\mathbf{E}(r,t)$ are found in terms of Λ_{ij}:

$$\mathbf{J}(t)=\frac{1}{2i}\sum_{i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\Lambda_{ij}(t) , \quad (1.17)$$

$$\mathbf{E}(\mathbf{r},t)=\frac{1}{8\pi\epsilon_0c^2}\sum_{i,j}(\omega_i-\omega_j)^2\left[\frac{\boldsymbol{\mu}_{ij}}{r}-\frac{(\boldsymbol{\mu}_{ij}\cdot\mathbf{r})\mathbf{r}}{r^3}\right]\times\Lambda_{ij}\left[t-\frac{r}{c}\right] , \quad (1.18)$$

where $t\geq r/c$. Since all measurements of the photon field deal with various correlations of **E** it is evident that the equations of motion for $\Lambda_{ij}^{\{n\}}$ permit the determination of all measurable parameters. Comparison of (1.17) and (1.14) shows that H_I is a reasonable model Hamiltonian.

The general state of atom plus scattered field is described by the general density matrix

$$\rho(t)=\sum_{i,j,\{n\},\{n'\}}|i,\{n\}\rangle\rho_{ij\{n\}\{n'\}}(t)\langle\{n'\},j| . \quad (1.19)$$

In Sec. III we show that there are two key situations in which the equations of motion for the reduced atomic density matrix are closed. These two cases are

$$\sigma(0)=\sum_{\{n\}}\langle\{n\}|\rho(0)|\{n\}\rangle \quad (1.20)$$

and

$$\sigma_0(0)=\langle\{0\}|\rho(0)|\{0\}\rangle(|\{0\}\rangle\langle\{0\}|) . \quad (1.21)$$

In view of (1.19), and (1.15) and (1.16),

$$\sigma(t)=\sum_{i,j}\rho_{ij}(t)\Lambda_{ij}(0) , \quad (1.22)$$

$$\sigma_0(t)=\sum_{i,j}\rho_{ij}^0(t)\Lambda_{ij}^0(0) , \quad (1.23)$$

where

$$\rho_{ij}(t)=\sum_{\{n\}}\rho_{ij\{n\}\{n\}}(t) , \quad (1.24)$$

$$\rho_{ij}^0(t)=\rho_{ij00}(t) . \quad (1.25)$$

In the first case the atomic density matrix (1.24) is the sum over all possible numbers of scattered photons and the resulting equations are the so-called optical Bloch equations.[11] For the V system they are derived in Sec. III in the RWA as[12,13]

$$\frac{d\rho_{11}}{dt}=-\beta_1\rho_{11}+i\Omega_{1,0}\bar{\rho}_{01}-i\Omega_{1,0}^*\bar{\rho}_{10} , \quad (1.26)$$

$$\frac{d\rho_{22}}{dt}=-\beta_2\rho_{22}+i\Omega_{2,0}\bar{\rho}_{02}-i\Omega_{2,0}^*\bar{\rho}_{20} , \quad (1.27)$$

$$\frac{d\bar{\rho}_{01}}{dt}=-(i\Delta_1+\tfrac{1}{2}\beta_1)\bar{\rho}_{01}+i\Omega^*_{1,0}(\rho_{11}-\rho_{00})+i\Omega^*_{2,0}\bar{\rho}_{21} , \tag{1.28}$$

$$\frac{d\bar{\rho}_{02}}{dt}=-(i\Delta_2+\tfrac{1}{2}\beta_2)\bar{\rho}_{02}+i\Omega^*_{2,0}(\rho_{22}-\rho_{00})+i\Omega^*_{1,0}\bar{\rho}_{12} , \tag{1.29}$$

$$\frac{d\bar{\rho}_{12}}{dt}=-(i\Delta_1+i\Delta_2+\tfrac{1}{2}\beta_1+\tfrac{1}{2}\beta_2)\bar{\rho}_{12}+i\Omega_{1,0}\bar{\rho}_{02}-i\Omega^*_{2,0}\bar{\rho}_{10} , \tag{1.30}$$

$$\frac{d\bar{\rho}_{00}}{dt}=\beta_1\rho_{11}+\beta_2\rho_{22}+i\Omega^*_{1,0}\bar{\rho}_{10}-i\Omega_{1,0}\bar{\rho}_{01}+i\Omega^*_{2,0}\bar{\rho}_{20}-i\Omega_{2,0}\bar{\rho}_{02} , \tag{1.31}$$

where

$$\rho_{ij}=\rho^*_{ji} ,$$

$$\bar{\rho}_{10}=\rho_{10}\exp[i(\omega_1-\omega_0+\Delta_1)t] , \tag{1.32}$$

$$\bar{\rho}_{20}=\rho_{20}\exp[i(\omega_2-\omega_0+\Delta_2)t] . \tag{1.33}$$

In the case under consideration $\rho_{ii}(t)$ is the probability that the atom is in state $|i\rangle$ independent of the state of the scattered field. In addition to induced transitions these equations allow for the return of the atom to the ground state through spontaneous decay, as evidenced by the terms $+\beta_1\rho_{11}$ and $+\beta_2\rho_{22}$ in (1.31). From the solution to these equations $\beta_1\rho_{11}(t)$ and $\beta_2\rho_{22}(t)$ yield the probability per second that a $|1\rangle\to|0\rangle$ or $|2\rangle\to|0\rangle$ photon will be emitted at time t.

In contrast to the *a* photon equations (1.26)–(1.31) the equation of motion for $\rho^0_{ij}(t)$ [the second case, Eq. (1.25)] yields the probability distribution for the *next* photon. In Sec. III we find that the equation of motion for ρ^0_{ij} factors, so that

$$\rho^0_{ij}=C_{i,0}C^*_{j,0} ,$$

and

$$\frac{d\bar{c}_2}{dt}=(i\Delta_2-\tfrac{1}{2}\beta_2)\bar{c}_2+i\Omega_{2,0}\bar{c}_0 , \tag{1.34}$$

$$\frac{d\bar{c}_1}{dt}=(i\Delta_1-\tfrac{1}{2}\beta_1)\bar{c}_1+i\Omega_{1,0}\bar{c}_0 , \tag{1.35}$$

$$\frac{d\bar{c}_0}{dt}=i\Omega^*_{1,0}\bar{c}_1+i\Omega^*_{2,0}\bar{c}_2 , \tag{1.36}$$

where

$$\bar{c}_i=C_{i,0}\exp[i(\omega_i+\Delta_i)t] \tag{1.37}$$

and $\Delta_0=0$. Equations (1.34)–(1.36), for the next photon amplitudes, were first presented in the pioneering work of Cohen-Tannoudji and Dalibard.[14] The factorization of the density matrix indicates that for the case of zero emitted photons the $C_{i,0}(t)$ are probability amplitudes. In view of (1.11) they are now in fact the amplitudes $C_{i\{0\}}(t)$ that at time t the atom will be in level $|i\rangle$ and that no photons will have been emitted between some reference time and time t. Similar to the closed system described by (1.4) the equations of motion for the $C_{i,0}$ are Hamiltonian, but in sharp contrast the Hamiltonian for the $C_{i,0}$ is not Hermitian. Thus the quantity

$$W(t)=\sum_i |C_{i,0}(t)|^2 \tag{1.38}$$

is not conserved. Furthermore, it is the probability that the next photon will be emitted after time t. An intuitive yet entirely accurate route to the *next* photon equations from the *a* photon equations is provided by dropping the regeneration term $\beta_1\rho_{11}+\beta_2\rho_{22}$ in (1.31). The resultant equations so obtained factor directly into (1.34)–(1.36). The regeneration terms correspond to an increase in the probability for the atom to be in the ground state as a result of spontaneous decay. As these terms represent effects which contribute to the $C_{i\{n\}}$ for $\{n\}\neq 0$, they do not contribute to the dynamics of the darkness amplitudes $C_{i,0}$.

Another route to the equations for $C_{i,0}$ comes from reasoning associated with off-equilibrium linear-response theory. From this approach we note that quantum theory requires the equations for $C_{i,0}$ be linear first-order differential equations. Use of the retarded potential requires that the variables $C_{i,0}$ form a complete set and that dW/dt be a negative definite quadratic form. The most general set of equations consistent with these restrictions has the form (1.34)–(1.36) when only the leading-order secularities are retained (the RWA).

From $W(t)$ the probability distribution of next photons can be directly obtained. The probability that the next photon is recorded between t and $t+dt$, provided that the atom was in $|0\rangle$ at $t=0$, is

$$D(t)dt=-(dW/dt)dt$$
$$=-[(\beta_1|C_{1,0}(t)|^2+\beta_2|C_{2,0}(t)|^2)]dt . \tag{1.39}$$

Thus a solution of (1.34)–(1.36) yields the statistics of the time between photons. These equations are much simpler than the *a* photon equations because they have three eigenvalues rather than eight eigenvalues. Yet these coherent next photon equations contain information lost in the incoherent averaging procedure that yields the optical Bloch equations.

In Sec. V the *next* photon and *a* photon approaches are compared as regards the long-studied[9,10] problem of two-level resonance fluorescence. The spectral intensities were first derived by Kimble and Mandel[9] and Mollow.[10] The antibunching[9] takes a simple form when described in terms of the next photon.

In Sec. VI the dark period statistics of the V system is calculated in detail. That the time of darkness is a key statistic has previously been emphasized.[3,14–16]

The solution to (1.34)–(1.36) which begins in the ground state at $t=0$ is shown in Sec. VI to be

$$\bar{c}_0=\cos(|\Omega_{1,0}|t)e^{-\beta_1 t/4}+4\left[\frac{\Omega_{2,0}}{\beta_1}\right]^2 e^{i|\Omega_{1,0}|t}(e^{-\beta_1 t/4}-e^{-\beta_l t}) , \tag{1.40}$$

$$\bar{c}_1=i\sin(|\Omega_{1,0}|t)e^{-\beta_1 t/4}+4\left[\frac{\Omega_{2,0}}{\beta_1}\right]^2 e^{i|\Omega_{1,0}|t}(e^{-\beta_1 t/4}-e^{-\beta_l t}) , \tag{1.41}$$

$$\bar{c}_2=2i(\Omega_{2,0}/\beta_1)(e^{-\beta_l t}-e^{-\beta_1 t/4})e^{i|\Omega_{1,0}|t} , \tag{1.42}$$

where we have taken $\Delta=|\Omega_{1,0}|$, and in each independent exponent we have kept only the leading terms in $|\Omega_{2,0}|/\beta_1$. The long-time scale for next photon emission is determined by

$$\beta_l\equiv\tfrac{1}{2}\beta_2+\frac{2|\Omega_{2,0}|^2}{\beta_1} . \tag{1.43}$$

When the strong transition is saturated $|\Omega_{1,0}|\geq\beta_1$. For this case the wide separation of time scales, $\beta_l\ll\beta_1$, occurs only if

$$|\Omega_{2,0}|/\beta_1\ll 1 , \tag{1.44}$$

which is a key telegraph inequality.

After the reset of the atom to the ground state the probability that there will be a dark period longer than t (where $t\gg 1/\beta_1$) follows from (1.38):

$$W(t)=4(|\Omega_{2,0}|/\beta_1)^2 e^{-2\beta_l t} . \tag{1.45}$$

A comparison of this result with the corresponding result (1.9), which follows from the unitary interpretation of darkness, indicates that the modification of $|\Psi\rangle$ due to null measurements tremendously enhances the probability of periods of darkness. In our picture the measurement of a period of time during which no photons are recorded changes our information about the system and thus the wave function. This null measurement increases the probability of successive periods of darkness. In addition to the failure of the photodetector to observe fluorescence events, the collapse of $|\Psi\rangle$ also requires a measurement determining that the laser is in fact turned on. The measurement of darkness with the laser off will obviously not lead to a change of the wave function of the ground-state atom.

The projection operator which effects the collapse of the wave function due to null fluorescence is[3]

$$\mathcal{P}=\sum_i |i,0\rangle\langle 0,i| . \tag{1.46}$$

If no scattered photons are observed for a time t after a reset to the ground state, the collapsed wave function at time t is then

$$|\psi(t)\rangle_c=\frac{\mathcal{P}}{\mathrm{Tr}\mathcal{P}}|\Psi(t)\rangle , \tag{1.47}$$

where $|\Psi(0)\rangle=|0,0\rangle$ and where $\mathrm{Tr}\mathcal{P}$ has been included for normalization. If at time t the atom plus field is in a state $|\Psi(t)\rangle$ and no scattered photons are observed between t and $t+\delta t$, the state of the system at time $t+\delta t$ is

$$|\Psi(t+\delta t)\rangle_c=\frac{\hat{\mathcal{P}}|\Psi(t+\delta t)\rangle}{\mathrm{Tr}\hat{\mathcal{P}}} , \tag{1.48}$$

where

$$\hat{\mathcal{P}}=[\exp(iH\delta t/\hbar)](I-\mathcal{P}) , \tag{1.49}$$

where I is the identity. The operator $I-\mathcal{P}$ projects out the states with nonzero scattered photons. The state of the system at time $t+\delta t$ that is created by the null measurements described by $\hat{\mathcal{P}}$ still depends upon its state at the earlier time t. The observation of darkness is thus a non-Markovian observation.

Comparing (1.42) with (1.41) we see that the critical collapse time T_c for which the observation of darkness causes $|\bar{c}_2|^2>|\bar{c}_1|^2$ is given by

$$T_c=(4/\beta_1)\ln(\beta_1/2|\Omega_{2,0}|) . \tag{1.50}$$

Hence when the emission of about $2\ln(\beta_1/2|\Omega_{2,0}|)$ photons is "missed" the atom has a substantial probability to be in $|2\rangle$. The weak (logarithmic) decoupling of the forbidden transition indicated by (1.50) suggests that isolated systems are difficult to achieve. This effect may provide a source of low-frequency noise.

During the period of darkness the V system has nonzero values of $\bar{c}_0$ and $\bar{c}_1$, so that the atom is indeed in a coherent superposition. From (1.39) one calculates that the probability of a dark period ending with an emission from $|1\rangle\to|0\rangle$, as opposed to $|2\rangle\to|0\rangle$, is

$$\lim_{t\to\infty}\frac{\beta_1|\bar{c}_1(t)|^2}{\beta_2|\bar{c}_2(t)|^2}=\frac{4|\Omega_{2,0}|^2}{\beta_1\beta_2}=\frac{\Gamma}{1-\Gamma} . \tag{1.51}$$

Figure 3 shows the probability distribution that after a reset the next photon will be emitted at time t with frequency $\omega_1-\omega_0$.

The coherence of the V system intermittency can be contrasted with the incoherent telegraph generated by the single laser optical pumping arrangement shown in Fig. 4. If the lifetime of level $|2\rangle$ in this arrangement is long compared to the lifetime of level $|1\rangle$, then the branching transition $|1\rangle\to|2\rangle$ can lead to a cessation of the fluorescence in the $|1\rangle$-$|0\rangle$ transition. In this case, however, the shelving in level $|2\rangle$ and hence the dark period is always preceded by the irreversible (incoherent) emission of a $|1\rangle\to|2\rangle$ photon. Also these dark periods always terminate with the emission of a $|2\rangle\to|0\rangle$ photon. This arrangement was discussed by Javanainen[12] from the point of view of the a photon equations.

Unlike the observation of a dark period the recording

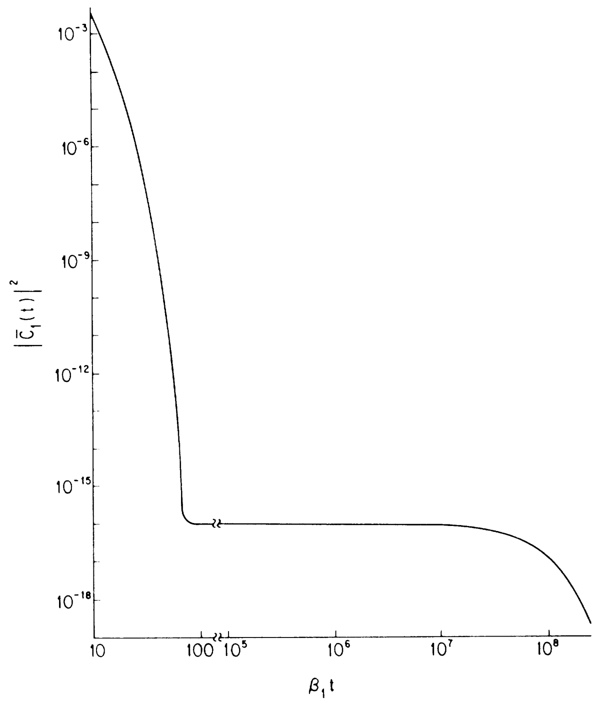

FIG. 3. Probability ($|\bar{c}_1(t)|^2$) that after a reset to $|0\rangle$ there will be no photons radiated for a time t and also that the atom will be in state $|1\rangle$ at t. The oscillations at $|\Omega_{1,0}|$ have been averaged out. For this plot we have taken $4|\Omega_{2,0}|^2/\beta_1\beta_2=1$, $\beta_2/\beta_1=10^{-8}$. The probability that the next photon will be a $|1\rangle$-$|0\rangle$ photon and that it will be recorded between t and $t+dt$ is $\beta_1|\bar{c}_1(t)|^2dt$. For the above parameters $\beta_1 T_c=37$. The extended plateau for $10^2<\beta_1 t<10^7$ indicates the presence of photons from the strongly coupled level $|1\rangle$ that take a long time to be emitted.

of a photon is a Markovian process which resets the atom. In Sec. IV the general theory of multiple photon correlations is reviewed. It is shown that the observation of a photon in the V arrangement resets the atom to the ground state provided that the measurement does not discriminate in frequency. This is the origin of antibunching,[17] which is calculated in Sec. V for a two-level

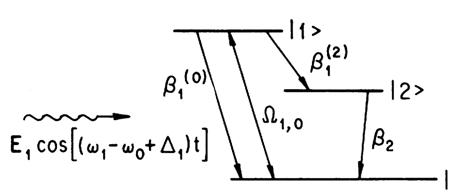

FIG. 4. Optical-pumping arrangement for a three-level atom driven by one laser tuned to the strong $|0\rangle$-$|1\rangle$ transition. The $|2\rangle$-$|0\rangle$ transition is forbidden so that $\beta_2/\beta_1 \ll 1$. This arrangement also shows intermittency but the dark periods are not coherent as evidenced by the fact that all dark periods now end with a $|2\rangle$-$|0\rangle$ photon in contrast with the V system described by Fig. 2(a). The decay rate of level $|1\rangle$ is $\beta_1=\beta_1^{(0)}+\beta_1^{(2)}$ so that the branching ratio from $|1\rangle$ to the (metastable) state $|2\rangle$ is $\beta_1^{(2)}/\beta_1^{(0)}$.

system. If the photon is forced to pass through a filter prior to being recorded then the measurement throws the system into more general mixed states. These mixed states are investigated in Sec. V, where it is also shown that filtered measurements lead to photon bunching and sideband correlations.[18] These effects are made observable when, for instance, a two-level system is driven by a field with frequency $\omega_L=\omega_1-\omega_0+\Delta$ where $\Delta>0$. As a consequence of the detuning, sidebands occur at $\omega_L-\Delta$ and $\omega_L+\Delta$ in addition to the central peak at ω_L. If a photon at $\omega_L+\Delta$ is observed we show in Sec. V that the probability of seeing a photon at $\omega_L-\Delta$ is about $|\Delta/\Omega|^4$ greater than in the steady state, provided that (a) the higher-frequency photon is seen first and (b) the interval between photons is less than $1/\beta$. This irreversibility in the order in which filtered photons are recorded also applies to the observation of V telegraph photons (Sec. VI) from the $|1\rangle\to|0\rangle$ and $|2\rangle\to|0\rangle$ transitions.

An approximation central to all the work presented in this paper is the rotating-wave approximation. When it is relaxed expressions such as (1.18) as well as the details of the Markovian resets[19] will have to be modified. We have also chosen a model wherein the dependence of H_I on $\mathbf{E}$ is linear. At intense electric fields, $\boldsymbol{\mu}$ can also depend upon $\mathbf{E}$, which could lead to substantially more intricate phenomena requiring more complicated analysis. Although issues related to the effect of degeneracy on fluorescence in a multilevel atom have been avoided, the theory developed in Sec. III is valid for this case and the consequences of the quantum Onsager cross terms should be pursued.

The power dependence of β_l and p_D are determined by the same key parameter (1.10). Their sensitivity to the coherence of E_2 relative to β_2 appears worthy of experimental investigation.

II. HEISENBERG EQUATIONS AND THE TWO-TIME-SCALES APPROXIMATION

The evolution of the N-level atom coupled to the second-quantized electro magnetic (EM) field is in principle completely described by the Heisenberg equations of motion. The EM field is characterized by the vector potential $\mathbf{A}(\mathbf{x},t)$ together with the gauge fixed by the condition

$$\nabla\cdot\mathbf{A}(\mathbf{x},t)=0 \; .$$

The scalar potential is taken to be zero. The general state of the atom plus field is described by the vector

$$|i,\{n_k\}\rangle=|i\rangle\otimes|\{n_k\}\rangle \; . \tag{2.1}$$

As mentioned in the Introduction i labels the atomic levels and

$$|\{n_k\}\rangle\equiv|\{n_{k_1},n_{k_2},\ldots\}\rangle$$

is the EM Fock space containing n_{k_i} photons with momentum k_i. The externally imposed coherent EM field has a vector potential with a positive frequency part given by $V(\mathbf{x},t)$. The expected value of the laser field is $\mathbf{V}+\mathbf{V}^*$. We will use the basis states (defined at some time, say, $t=0$)

$$|i,\{\tilde{n}_k\},\mathbf{V}(\mathbf{x},0)\rangle \equiv |i\rangle \otimes |\{\tilde{n}_k\}\rangle \otimes |\mathbf{V}(\mathbf{x},0)\rangle \ , \tag{2.2}$$

where

$$\Box \mathbf{V}(\mathbf{x},t)=\mathbf{0} \ , \tag{2.3}$$

so that the $\tilde{n}_{k_i}$ are to be interpreted as the number of photons with momentum k_i in the states orthogonal to the coherent EM state

$$|\mathbf{V}(\mathbf{x},0)\rangle \ . \tag{2.4}$$

Thus the complete EM Hilbert space can be written as the direct product

$$\mathcal{H}^{\mathrm{EM}}=\mathcal{H}^{\perp}\otimes|\mathbf{V}(\mathbf{x},0)\rangle \ . \tag{2.5}$$

In this way the EM field has been decomposed into a "quantized" field $\mathbf{A}_q$ (describing the scattered photons) and a classical coherent field $\mathbf{A}_f$ (describing the laser) so that

$$\mathbf{A}(\mathbf{x},t)=\mathbf{A}_q(\mathbf{x},t)+\mathbf{A}_f(\mathbf{x},t) \ , \tag{2.6}$$

where

$$\Box \mathbf{A}_f=\mathbf{0} \ . \tag{2.7}$$

The free-field operator can be separated into its positive and negative frequency parts [positive frequency means $\exp(-i\omega t)$ with $\omega>0$],

$$\mathbf{A}_f=\mathbf{A}_f^{+}+\mathbf{A}_f^{-} \ . \tag{2.8}$$

The connection between $\mathbf{V}$ and $\mathbf{A}$ is provided by the relations

$$\mathbf{A}_f^{+}(\mathbf{x},t)|\{n_k\},\mathbf{V}(\mathbf{x},0)\rangle=\mathbf{V}(\mathbf{x},t)|\{n_k\}\mathbf{V}(\mathbf{x},0)\rangle \ , \tag{2.9}$$

$$\langle \mathbf{V}(\mathbf{x},0),\{n_k\}|A_f^{-}(\mathbf{x},t)=\langle \mathbf{V}(\mathbf{x},0),\{n_k\}|\mathbf{V}^{*}(\mathbf{x},t) \ .$$

We will interpret the n_k as the set of occupation numbers for the field $\mathbf{A}_q$. So from here on we drop the tilde superscript from n in (2.2). The commutation relations for the electromagnetic field can be applied to the Fourier components of $\mathbf{A}_q$ which in turn is decomposed into positive and negative frequencies

$$\mathbf{A}_q=\mathbf{A}_q^{+}+\mathbf{A}_q^{-} \ ,$$

where

$$\mathbf{A}_q^{+}(\mathbf{x},0)=\sum_{\mathbf{k}} a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{x}} \ ,$$

$$\mathbf{A}_q^{-}(\mathbf{x},0)=\sum_{\mathbf{k}} a_{\mathbf{k}}^{+} e^{-i\mathbf{k}\cdot\mathbf{x}} \ .$$

In this representation $a,a^{\dagger}$ are the usual lowering and raising operating with

$$[a_{\mathbf{k}},a_{\mathbf{k}}^{\dagger}]=\delta_{\mathbf{k},\mathbf{k}'} \ .$$

The state of the atom plus scattered field is described by the operators

$$\Lambda_{ij}^{\{n\}}=|i,\{n_k\}\rangle\langle\{n_k\},j| \tag{2.10}$$

which obey the equal-time relations

$$\Lambda_{ij}^{\{n\}}\Lambda_{kl}^{\{n'\}}=\delta_{jk}\Lambda_{il}^{\{n\}} \quad \text{for } \{n\}=\{n'\} \ ,$$

so that

$$\begin{aligned}[\Lambda_{ij}^{\{n\}},\Lambda_{kl}^{\{n'\}}]&=\delta_{jk}\Lambda_{il}^{\{n\}}-\delta_{il}\Lambda_{jk}^{\{n\}} \quad \text{for } \{n\}=\{n'\}\\ &=0 \quad \text{for } \{n\}\neq\{n'\}\end{aligned} \tag{2.11}$$

and

$$\Lambda_{ij}=\sum_{\{n_k\}}\Lambda_{ij}^{\{n\}}=|i\rangle\langle j|\otimes I \ ,$$

where I is the identity operator on $\mathcal{H}^{\perp}$. The interaction Hamiltonian (1.14) is

$$H_I=-\frac{i}{2}\sum_{\{n_k\}i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\cdot[\mathbf{A}^{-}(0,t)\Lambda_{ij}^{\{n\}}(t)+\Lambda_{ij}^{\{n\}}(t)\mathbf{A}^{+}(0,t)] \ , \tag{2.12}$$

where $\mathbf{A}^{-}$, $\mathbf{A}^{+}$ are the negative and positive frequency contributions to $\mathbf{A}$ where

$$\mathbf{A}=\mathbf{A}^{-}+\mathbf{A}^{+} \ , \tag{2.13}$$

and H_I has been written in time-ordered form.

The time development of any operator (including its dependence on the coherent state) is determined by the commutator with H provided that the explicit time dependence, of H, and the decomposition (2.6) can be neglected. For fixed laser illumination this is the case, but if the amplitude or frequency of the laser is varied an explicit time dependence will appear. When these variations are slow compared to $\omega_i-\omega_j$ the adiabatic approximation is valid and the commutator of an operator with H still yields its equation of motion. For short pulses of duration $\Delta t\sim 1/(\omega_i-\omega_j)$ the separation (2.6) is not valid.

The Heisenberg equations for Λ_{ij} follow from (1.12) and (2.12):

$$\begin{aligned}\dot{\Lambda}_{kl}^{\{n\}}(t)=&\,i(\omega_k-\omega_l)\Lambda_{kl}^{\{n\}}(t)+\frac{1}{2\hbar}\sum_{i,j}(\omega_i-\omega_j)\mu_{ij}\mathbf{A}^{-}(0,t)\Lambda_{ij}\Lambda_{kl}^{\{n\}}-\frac{1}{2\hbar}\sum_{i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\Lambda_{kl}^{\{n\}}\mathbf{A}^{-}(0,t)\Lambda_{ij}\\ &-\frac{1}{2\hbar}\sum_{i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\Lambda_{kl}^{\{n\}}\Lambda_{ij}\mathbf{A}^{+}(0,t)+\frac{1}{2\hbar}\sum_{i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\Lambda_{ij}\mathbf{A}^{+}(0,t)\Lambda_{kl}^{\{n\}} \ .\end{aligned} \tag{2.14}$$

The transverse EM current

$$J^{T}=J-\Delta^{-1}\nabla(\nabla\cdot\mathbf{J}) \tag{2.15}$$

is the source of the scattered field

$$\Box \mathbf{A}=\mathbf{J}^T . \tag{2.16}$$

Following Ref. 9, the total current is

$$\mathbf{J}=\frac{1}{2i}\sum_{i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\Lambda_{ij} . \tag{2.17}$$

From (2.15)–(2.17) one can obtain $\mathbf{A}(0,t)$ in terms of Λ_{ij}:

$$\mathbf{A}^-(0,t)=\frac{1}{4\pi\epsilon_0 c^2}\left[\frac{1}{3c}\sum_{i\geq j}(\omega_i-\omega_j)^2\boldsymbol{\mu}_{ij}\Lambda_{ij}+\frac{i}{3M}\sum_{i\geq j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}\Lambda_{ij}\right]+\mathbf{A}_f^-(0,t) , \tag{2.18}$$

which is valid for $t>0$ (M is a parameter related to the Lamb shift). Proper time ordering of the equations of motion (2.14) is most effectively carried out in terms of the N photon operators

$$\Lambda_{ij}^N\equiv\sum_{\{n\}}\Lambda_{ij}^{\{n\}} ,\qquad \sum_k n_k=N . \tag{2.19}$$

These operators obey the relation

$$\mathbf{A}_q^+\Lambda_{ij}^N=\Lambda_{ij}^{N-1}\mathbf{A}_q^+(1-\delta_{N,0}) . \tag{2.20}$$

Substitution of (2.18) and (2.20) into (2.14) yields

$$\begin{aligned}\dot{\Lambda}_{kl}^N(t)=\;&i\omega_k\Lambda_{kl}^N(t)+\tfrac{1}{2}a\sum_{n\geq i}(\omega_i-\omega_k)\boldsymbol{\mu}_{ik}(\omega_n-\omega_i)^2\boldsymbol{\mu}_{ni}\Lambda_{nl}^N(t)+\frac{ib}{2}\sum_{n\geq i}(\omega_i-\omega_k)\boldsymbol{\mu}_{ik}(\omega_n-\omega_i)\boldsymbol{\mu}_{ni}\Lambda_{nl}^N(t)\\&+\frac{1}{2\hbar}\sum_i(\omega_i-\omega_k)\boldsymbol{\mu}_{ik}\,\mathbf{A}_f^-(t)\Lambda_{il}^N(t)-\frac{1}{2\hbar}\sum_j(\omega_l-\omega_j)\boldsymbol{\mu}_{lj}\,\mathbf{A}_f^-(t)\Lambda_{kj}^N(t)\\&-\frac{a}{2}\sum_{j,n\geq k}(\omega_l-\omega_j)\boldsymbol{\mu}_{lj}\boldsymbol{\mu}_{nk}(\omega_n-\omega_k)^2\Lambda_{nj}^{N-1}(t)(1-\delta_{N,0})\\&-\frac{ib}{2}\sum_{j,n\geq k}(\omega_l-\omega_j)\boldsymbol{\mu}_{lj}\boldsymbol{\mu}_{nk}(\omega_n-\omega_k)\Lambda_{nj}^{N-1}(t)(1-\delta_{N,0})+\text{H.c.}(k\leftrightarrow l) ,\end{aligned} \tag{2.21}$$

where H.c. denotes the Hermitian conjugate, $k\leftrightarrow l$ indicates the interchange of k and l indices, and

$$a^{-1}=12\pi\epsilon_0 c^3\hbar ,$$
$$b^{-1}=12\pi\epsilon_0 c^2\hbar M .$$

Equation (2.21) can be cast into the form

$$\dot{\Lambda}_{kl}^N=\frac{i}{\hbar}\sum_j(H_{kj}^*\Lambda_{jl}^N-\Lambda_{kj}^N H_{lj})+(1-\delta_{N0})\left[\sum_{\beta,\alpha\geq k}L_{kl\alpha\beta}\Lambda_{\alpha\beta}^{N-1}+\text{H.c.}(k\leftrightarrow l)\right] , \tag{2.22}$$

where

$$H_{kj}^*=\hbar\omega_k\delta_{kj}-\tfrac{1}{2}i\hbar a\sum_{i\leq j}(\omega_i-\omega_k)(\omega_i-\omega_j)^2\boldsymbol{\mu}_{ik}\cdot\boldsymbol{\mu}_{ji}+\tfrac{1}{2}b\hbar\sum_{i\leq j}(\omega_i-\omega_k)(\omega_j-\omega_i)\boldsymbol{\mu}_{ik}\cdot\boldsymbol{\mu}_{ji}-\tfrac{1}{2}i(\omega_j-\omega_k)\boldsymbol{\mu}_{jk}\cdot(\mathbf{V}+\mathbf{V}^*)$$

and

$$L_{kl\alpha\beta}=\frac{a}{2}(\omega_l-\omega_\beta)(\omega_\alpha-\omega_k)^2\boldsymbol{\mu}_{l\beta}\cdot\boldsymbol{\mu}_{\alpha k}-\tfrac{1}{2}ib(\omega_l-\omega_\beta)(\omega_\alpha-\omega_k)\boldsymbol{\mu}_{l\beta}\cdot\boldsymbol{\mu}_{\alpha k} .$$

Introduction of the slowly varying fields

$$\begin{aligned}&\mathbf{A}_{ij}^+(t)=\mathbf{A}_f^+(t)e^{-i(\omega_i-\omega_j)t},\quad i<j\\&\mathbf{A}_{ji}^-(t)=\mathbf{A}_f^-(t)e^{i(\omega_i-\omega_j)t},\quad i<j\\&\overline{\Lambda}_{ij}^N(t)=\Lambda_{ij}^N(t)e^{-i(\omega_i-\omega_j)t} ,\\&\mathbf{V}_{ij}(t)=\mathbf{V}(t)e^{-i(\omega_i-\omega_j)t},\quad i<j\end{aligned} \tag{2.23}$$

(where ω_i have been ordered so that $i<j \leftrightarrow \omega_i<\omega_j$) enables one to transform the Heisenberg equations (2.21) into the form

$$\begin{aligned}\dot{\overline{\Lambda}}{}^{N}_{kl}(t)=&-\tfrac{1}{2}\sum_{i<k} a_{ki}\overline{\Lambda}{}^{N}_{kl}(t)-\frac{i}{2}\sum_{i<k} b_{ki}\overline{\Lambda}{}^{N}_{kl}(t)+\frac{1}{2\hbar}\sum_{i<k}(\omega_i-\omega_k)\boldsymbol{\mu}_{ik}\cdot\mathbf{A}^{-}_{ki}(t)\overline{\Lambda}{}^{N}_{il}\\&-\frac{1}{2\hbar}\sum_{i>l}(\omega_l-\omega_i)\boldsymbol{\mu}_{li}\cdot\mathbf{A}^{-}_{il}(t)\overline{\Lambda}{}^{N}_{ki}+\tfrac{1}{2}\delta_{kl}\sum_{i>k}a_{ik}\overline{\Lambda}{}^{(N-1)}_{ii}(t)(1-\delta_{N,0})\\&+\frac{i}{2}\delta_{kl}\sum_{i>k}b_{ki}\overline{\Lambda}{}^{(N-1)}_{ii}(t)(1-\delta_{N,0})+\mathrm{H.c.}(k\leftrightarrow l)\ ,\end{aligned} \tag{2.24}$$

where

$$\begin{aligned}a_{li}&=a\boldsymbol{\mu}_{li}\cdot\boldsymbol{\mu}_{il}(\omega_l-\omega_i)^3\ ,\\ b_{il}&=b\boldsymbol{\mu}_{li}\cdot\boldsymbol{\mu}_{il}(\omega_i-\omega_l)^2\ .\end{aligned} \tag{2.25}$$

Effects due to degeneracy have been neglected. Also neglected are oscillations at optical frequencies. This is generally referred to as the rotating-wave approximation. The procedure which leads to (2.24) is also called the two-time-scales approximation. Corrections to quantities calculated in the RWA are of the order

$$\mu^2E^2/\overline{\omega}^2\hbar^2\ , \tag{2.26}$$

or

$$(a_{ki}/\overline{\omega})^2\ ,$$

where E is the externally imposed electric field, μ a typical transition matrix element, and $\overline{\omega}$ the smallest optical transition frequency. These terms are the ratio of Rabi flopping frequency to the optical frequency and the ratio of inverse lifetime to the optical frequency.

Corrections to (2.16) which are proportional to $\partial J/\partial t$ and are therefore non-Markovian should appear in a more complete evaluation of this model system (2.12).[19] In the two-timing approximation used here these corrections are again down by the factor (2.26) and they are therefore neglected.

The Λ^N_{ij} describe every observable for an atom interacting with the EM field. In particular, (2.18) yields an expression for the EM field scattered in a direction different from the laser:

$$\mathbf{E}(\mathbf{r},t)=\frac{1}{8\pi\epsilon_0c^2}\sum_{i,j}(\omega_i-\omega_j)^2\left[\frac{\boldsymbol{\mu}_{ij}}{r}-\frac{(\boldsymbol{\mu}_{ij}\cdot\mathbf{r})\mathbf{r}}{r^3}\right]\times\Lambda_{ij}\left[t-\frac{r}{c}\right]\quad\text{where } t\geq\frac{r}{c}\ . \tag{2.27}$$

From various moments of $\mathbf{E}$ one can form the intensity (photon number) as well as all quantities measured by a photodetector. The various frequency components of $\mathbf{E}$ correspond to the various photons emitted by the multilevel atom.

III. CLOSED EQUATIONS FOR THE REDUCED ATOMIC DENSITY MATRIX: THE INCOHERENT OPTICAL BLOCH EQUATIONS VERSUS THE COHERENT NEXT PHOTON DYNAMICS

The atom plus scattered field [in an imposed coherent state $\mathbf{V}(\mathbf{x},0)$] is described by the general density matrix which at $t=0$ is given by

$$\rho(0)=\sum_{i,j,\{n\},\{n'\}}|i,\{n\}\rangle\rho_{ij\{n\}\{n'\}}(0)\langle\{n'\},j|\ . \tag{3.1}$$

A reduced density matrix ρ_{ij} can be used to describe the product of atomic state amplitudes subject to some specified condition for the scattered EM field. There are two situations in which the equations of motion for ρ_{ij} are closed:

$$\rho_{ij}=\sum_{\{n\}}\rho_{ij\{n\}\{n\}} \tag{3.2}$$

and

$$\rho_{ij}=\rho_{ij00}\equiv\rho^{(0)}_{ij}\ . \tag{3.3}$$

In case (3.2) ρ_{ij} includes a sum over all possible numbers of scattered photons. The equations of motion for ρ_{ij} given by (3.2) are the so-called optical Bloch equations. As all EM states are mixed together these equations are incoherent. A property of this incoherence is that the inequality

$$\sum_j\rho_{ij}\rho_{jk}\neq f(t)\rho_{ik} \tag{3.4}$$

applies for all choices of the function $f(t)$. In case (3.3) the atomic quantum dynamics is restricted by the condition that there are no outgoing (scattered) photons ($N=0$), other than the coherent laser state. For this quantum radiationless situation there does exist a function f such that

$$\sum_j\rho^{(0)}_{ij}\rho^{(0)}_{jk}=f(t)\rho^{(0)}_{ik}\ , \tag{3.5}$$

so that coherence is maintained.

The time development of the reduced density matrices can be found in terms of the total density matrix or the operators $\Lambda^{\{n\}}_{ij}$ which obey

$$\begin{aligned}\rho(t)&=e^{-iHt/\hbar}\rho(0)e^{iHt/\hbar}\ ,\\ \Lambda^{\{n\}}_{ij}(t)&=e^{iHt/\hbar}\Lambda^{\{n\}}_{ij}(0)e^{-iHt/\hbar}\ .\end{aligned} \tag{3.6}$$

The relationship of Λ_{ij} to ρ follows from the statistical average:

$$\begin{aligned}\langle\Lambda^{\{n\}}_{ij}(t)\rangle_{\rho(0)}&\equiv\mathrm{Tr}\rho(0)\Lambda^{\{n\}}_{ij}(t)\\&=\mathrm{Tr}\rho(t)\Lambda^{\{n\}}_{ij}(0)\\&=\langle\{n\},j|\rho(t)|i,\{n\}\rangle\ .\end{aligned} \tag{3.7}$$

Thus $\langle \Lambda_{ij}^{\{n\}}(t) \rangle_{\rho(0)}$ is the matrix element of the density matrix $\rho(t)$ [evolving from an initial condition $\rho(0)$] between the states $\langle \{n\},j|$ and $|i,\{n\}\rangle$. The equations of motion for ρ_{ij} therefore follow from (2.14). The trace in (3.7) is over $\{n_k\}$ for the fixed coherent state.

The reduced density matrix corresponding to (3.2) can be written as

$$\begin{aligned}\rho_{ij}(t) &= \sum_{\{n\}} \langle \Lambda_{ji}^{\{n\}}(t) \rangle_{\rho(0)} \\ &= \langle \Lambda_{ji}(t) \rangle_{\rho(0)} \\ &= \sum_{\{n\}} \langle \{n\},i|\rho(t)|j,\{n\}\rangle \ ,\end{aligned} \tag{3.8}$$

whereas the reduced density matrix described by (3.3) is

$$\rho_{ij}^{(0)}(t) = \langle 0,i|\rho(t)|j,0\rangle = \langle \Lambda_{ji}^{(0)}(t) \rangle_{\rho(0)} \ . \tag{3.9}$$

The radiationless reduced density matrix (3.9) is related to the incoherent matrix (3.8) by a projection

$$\mathcal{P} \equiv \sum_i |i,0\rangle\langle 0,i| \ , \tag{3.10}$$

so that

$$\rho_{ij}^{(0)}(t) = \mathrm{Tr}\mathcal{P}\Lambda_{ji}(0)\rho(t) \ . \tag{3.11}$$

The closed equations which describe the dynamics of null emission follow from (3.9) and (2.22) with $N=0$:

$$\frac{d}{dt}\rho_{kl}^{(0)} = \frac{i}{\hbar} \sum_j (H_{lj}^* \rho_{kj}^{(0)} - H_{kj}\rho_{jl}^{(0)}) \ . \tag{3.12}$$

The form of (3.12) immediately implies the factorization property mentioned in Sec. I (from here on we drop the subscript 0 from $C_{i,0}$):

$$\rho_{kl}^{(0)} = C_k C_l^* \ , \tag{3.13}$$

where now

$$i\hbar \frac{dC_k}{dt} = \sum_j H_{kj} C_j \ . \tag{3.14}$$

In terms of the Rabi flopping frequencies and spontaneous decay coefficients,

$$\Omega_{kj}(t) = \frac{i}{2\hbar}(\omega_k - \omega_j)\boldsymbol{\mu}_{jk}^* \cdot (\mathbf{V} + \mathbf{V}^*) \ , \tag{3.15}$$

$$\beta_{kj} = a \sum_{\alpha<j} (\omega_k - \omega_\alpha)(\omega_j - \omega_\alpha)^2 \boldsymbol{\mu}_{\alpha k} \cdot \boldsymbol{\mu}_{j\alpha} \ , \tag{3.16}$$

the Hamiltonian matrix elements are

$$H_{kj} = \hbar\omega_k \delta_{kj} - \frac{i}{2}\hbar\beta_{kj} - \hbar\Omega_{kj}(t) \ , \tag{3.17}$$

where the Lamb shift has been absorbed into ω_k. The terms in β_{kj} for $k \neq j$ are Onsager cross terms arising from spontaneous decay. They become especially important in the case of degeneracy. The optical Bloch equations for the time development of ρ_{ij} given by (3.2) are obtained by summing the expectation of (2.22) over all N to obtain the closed equation

$$\begin{aligned}\frac{d}{dt}\rho_{kl} &= \frac{i}{\hbar} \sum_j (H_{lj}^* \rho_{kj} - H_{kj}\rho_{jl}) \\ &\quad + \sum_{\beta,\alpha>k} (L_{kl\alpha\beta}\rho_{\beta\alpha} + L_{lk\alpha\beta}^* \rho_{\alpha\beta}) \ .\end{aligned} \tag{3.18}$$

Since all photon occupations are mixed (3.18) does not allow the decomposition (3.13) and the ρ_{kl} determined by (3.18) obey the inequality (3.4). In fact, for an n-level system (3.18) yields n^2-1 independent linear equations, whereas (3.12) involves only n such equations. When $N=0$ the function $f(t)$ in (3.5) is found to be $f = \sum_j |C_j(t)|^2$. This expression depends on the initial condition.

The optical Bloch equations provide for the conservation of probability of the atomic levels or

$$\frac{d}{dt} \sum_i \rho_{ii}(t) = 0 \ . \tag{3.19}$$

Proof of (3.19) follows from the identity

$$\sum_{\alpha \le j} L_{\alpha\alpha jk} = -\tfrac{1}{2}\beta_{kj} \ . \tag{3.20}$$

For the $N=0$ equations the probability of the atom remaining radiationless in an excited state goes to zero as $t \to \infty$. This fact is described by the inequality

$$\begin{aligned}\frac{d}{dt} \sum_i \rho_{ii}^{(0)}(t) &= \frac{d}{dt} \sum_i |C_i^2(t)| \\ &= -\tfrac{1}{2} \sum_{k,j} (\beta_{kj} + \beta_{jk}^*) C_k^*(t) C_j(t) < 0 \ .\end{aligned} \tag{3.21}$$

For the Bloch equations the probability ρ_{ii} that the atom is in the ith level (independent of the state of the EM field) eventually settles down to a steady-state value for steady illumination.

For the null emission dynamics (3.12), $C_i(t)$ is the amplitude, where the atom will be in state i at time t and where no photons will have been emitted between the initial time (say, $t=0$) and t. For the initial state we take

$$\sum_i |C_i^2(0)| = 1 \ . \tag{3.22}$$

The probability that the next photon is emitted between t and $t+dt$ is then

$$\begin{aligned}D(t)dt &= -\frac{d}{dt} \sum_i |C_i^2(t)| dt \\ &= -\tfrac{1}{2} \sum_{k,j} (\beta_{kj} + \beta_{jk}^*) C_k^*(t) C_j(t) dt \ ,\end{aligned} \tag{3.23}$$

where $C_i(t)$ is determined via the effective (but non-Hermitian) Hamiltonian equation (3.14).

For a three-level system with $\mu_{12}=0$ the imposition of the RWA and neglect of degeneracy yields (1.34)–(1.36) and (1.26)–(1.31) from (3.14) and (3.18), respectively. In this regard one sets $\beta_{kk} = \beta_k$ and

$$\Omega_{k0}^+(t) = \Omega_{k0}\exp[-i(\omega_k - \omega_0 + \Delta_k)t] \ .$$

The key experimental observable is the scattered light.

The next photon equations yield, according to (3.23), the probability of periods of darkness. The Bloch equations yield the expected rate of photon emission which is proportional to the expected intensity of fluorescence. When degeneracy is neglected the average rate of photon emission by a given transition $k \rightarrow j$ is

$$\langle \dot{N}_{kj} \rangle = \beta_k^{(j)} \rho_{kk}(t) , \tag{3.24}$$

which is the probability that the atom is in the kth level times the probability per second of spontaneous decay for the transition in question [$\beta_k^{(j)}$ is the contribution to β_{kk} in (3.16) from the term $\alpha = j$]. By mixing together all photon occupation numbers the optical Bloch equations (3.18) yield the probability of observing *a* photon, whereas the null emission dynamics (3.14) yield the quantum amplitudes for the *next* photon.

IV. MULTIPLE PHOTON MEASUREMENTS

A measurement which depends upon the state of the scattered EM field at a succession of time is proportional to the n-point Green's function

$$\langle \mathbf{E}_{i_1 j_1}(t_1) \cdots \mathbf{E}_{i_n j_n}(t_n) \rangle_{\rho(0)} . \tag{4.1}$$

According to (2.27) the E_{ij} are proportional to the Λ_{ij}. Thus we can set

$$\mathbf{E} = \sum_{i,j} \mathbf{E}_{ij} . \tag{4.2}$$

For $i < j$ the frequency is positive. The contribution to E^+ therefore takes the form

$$E^+ = \sum_{i<j} E_{ij} . \tag{4.3}$$

The correlation function to observe *a* photon from the $j \rightarrow i$ transition within dt_1 about t_1 as well as *a* photon from the $l \rightarrow k$ transition within dt_2 about t_2 is proportional to the product of intensities from these transitions, or

$$\begin{aligned} I_{jilk}(t_1,t_2)dt_1 dt_2 &= \langle \Lambda_{ji}(t_1)\Lambda_{lk}(t_2)\Lambda_{kl}(t_2)\Lambda_{ij}(t_1) \rangle_{\rho(0)} dt_1 dt_2 \\ &= \mathrm{Tr}[\Lambda_{ij}(0)\rho(t_1)\Lambda_{ji}(0)\Lambda_{lk}(t_2-t_1)\Lambda_{kl}(t_2-t_1)]dt_1 dt_2 \\ &= \langle \Lambda_{jj}(t_1) \rangle_{\rho(0)} \langle i | \Lambda_{ll}(t_2-t_1) | i \rangle dt_1 dt_2 . \end{aligned} \tag{4.4}$$

As before the trace is calculated for fixed coherent state $\mathbf{V}(\mathbf{x},0)$. In writing (4.4) it has been assumed that $t_2 > t_1$, $j > i$, and $l > k$. The factorization apparent in (4.4) describes the atomic reset that accompanies the photon observation process.[9,16] According to (4.4) the probability of observing a $j \rightarrow i$ photon (at t_1) and a $l \rightarrow k$ photon (at t_2) emitted from the same atom is the product of (a) the probability of being in state j at time t_1, and (b) the probability of being in state l at time t_2 given that it was in state i at time t_1. The transition of the atom from $j \rightarrow i$ at time t_1 resets the atom to state i at this time. For a two-level system this means that the atom is in the ground state after the emission of a photon and the past history is destroyed at least as regards the model system considered here.

Equation (4.4) is a special case of the more general Markovian factorization property of the multiple time scales correlations. Consider again the general multiple time scale correlation [proportional to (4.1)]

$$\begin{aligned} \langle \Lambda_{i_1 j_1}(t_1) \cdots \Lambda_{i_n j_n}(t_n) \rangle_{\rho(0)} &= \mathrm{Tr}[\Lambda_{i_n j_n}(t_n - t_1)\rho(t_1)\Lambda_{i_1 j_1}(0)\Lambda_{i_2 j_2}(t_2 - t_1) \cdots \Lambda_{i_{n-1} j_{n-1}}(t_{n-1} - t_1)] \\ &= \mathrm{Tr}[\hat{\rho}(t_n - t_1, t_1)]\mathrm{Tr}[\tilde{\rho}(t_n - t_1, t_1)\Lambda_{i_2 j_2}(t_2 - t_1) \cdots \Lambda_{i_{n-1} j_{n-1}}(t_{n-1} - t_1)] , \end{aligned} \tag{4.5}$$

where

$$\hat{\rho}(t_n - t_1, t_1) = \Lambda_{i_n j_n}(t_n - t_1)\rho(t_1)\Lambda_{i_1 j_1}(0) \tag{4.6}$$

and

$$\tilde{\rho} = \hat{\rho} / \mathrm{Tr}\hat{\rho} \tag{4.7}$$

is the normalized "initial condition" from which $\langle E_2 \cdots E_{n-1} \rangle$ develops.

Correlations such as (4.5) describe multiphoton observations in the presence of a filter. Let $F(\omega)$ characterize the relative extent to which the filter passes photons of frequency ω. The filtered spectral intensity is

$$\langle E_F^2(\omega) \rangle_t = \langle E^2(\omega,t) \rangle F(\omega) = \int \langle E^-(t)E^+(t+\tau) \rangle e^{i\omega\tau} d\tau F(\omega) . \tag{4.8}$$

The total rate at which filtered energy is radiated at time t is then given by

$$\epsilon_0 c \int \int \langle E_F^2(\omega) \rangle_t d\omega \, \hat{k} \cdot d\mathbf{s} = \epsilon_0 c \int \int \langle \mathbf{E}^-(t,\mathbf{x})\mathbf{E}^+(t+\tau,\mathbf{x}) \rangle F(\tau) d\tau \, \hat{k} \cdot d\mathbf{s} , \tag{4.9}$$

where

$$F(t) = \int F(\omega) e^{i\omega t} d\omega \tag{4.10}$$

and $d\mathbf{s}$ is an element of surface through which the radiation is flowing in the outward direction $\hat{k}$. So, in the presence of filters the probability of seeing a photon from the $j\to i$ transition within dt_1 of t_1 and a photon from the $l\to k$ transition within dt_2 of t_2 is proportional to

$$I_{jilk}(t_1,t_2)=\int\int\langle\Lambda_{ji}(t_1)\Lambda_{lk}(t_2)\Lambda_{kl}(t_2')\Lambda_{ij}(t_1')\rangle_{\rho(0)}F_1(t_1-t_1')F_2(t_2-t_2')dt_1'dt_2' \ , \tag{4.11}$$

where F_1,F_2 are the filtering functions applied to the separate transitions. The initial state from which the $l\to k$ transition develops is determined from (4.6) and (4.7) as

$$\tilde{\rho}_{F_1}(t_1)=\hat{\rho}_{F_1}(t_1)/\mathrm{Tr}\hat{\rho}_{F_1}(t_1) \ , \tag{4.12}$$

where

$$\hat{\rho}_{F_1}(t_1)=\int\Lambda_{ij}(t'-t_1)\rho(t_1)\Lambda_{ji}(0)F_1(t_1'-t_1)dt_1' \tag{4.13}$$

so that

$$I_{jilk}(t_1,t_2)=\mathrm{Tr}\hat{\rho}_{F_1}(t_1)\mathrm{Tr}\int\tilde{\rho}_{F_1}(t_1)\Lambda_{lk}(t_2-t_1)\Lambda_{kl}(t_2'-t_1)F_2(t_2'-t_2)dt_2' \tag{4.14}$$

$$=\sum_{\alpha}\mathrm{Tr}\left[\int\Lambda_{j\alpha}(t_1)\Lambda_{ij}(t_1')\rho(0)F_1(t_1'-t_1)dt_1'\right]\int\langle i|\Lambda_{lk}(t_2-t_1)\Lambda_{kl}(t_2'-t_1)|\alpha\rangle F_2(t_2'-t_2)dt_2' \ . \tag{4.15}$$

In the presence of broadband detection $F_i(\tau)=\delta(\tau)$ and Eq. (4.4) is recovered. Furthermore,

$$\langle i|\Lambda_{ll}(t_2-t_1)|i\rangle$$

is zero for $t_2=t_1$ and $i\neq l$. Thus the rate of emission of the second photon is suppressed for short times (antibunching) unless the lower level of the first emission i coincides with the upper level l of the second emission (a cascade). Whereas broadband detection resets the atom to the lower level, measurements which discriminate the frequency yield reset atomic density matrices where $\alpha\neq i$. Thus the reset atom has a nonzero probability to be in an excited level.

The expected rate of outflow of energy from the $j\to i$ transition is

$$Q_{ji}(t)\equiv\int\epsilon_0c\langle\mathbf{E}_{ji}(\mathbf{x},t)\mathbf{E}_{ij}(\mathbf{x},t)\rangle_{\rho(0)}\hat{k}\cdot d\mathbf{s} \ . \tag{4.16}$$

In view of (2.27) and (3.16), (4.16) becomes

$$Q_{jj}=\beta_{jj}\hbar(\omega_j-\omega_i)\mathrm{Tr}[\Lambda_{ji}(t)\Lambda_{ij}(t)\rho(0)]$$
$$=\beta_{jj}\hbar(\omega_j-\omega_i)\langle\Lambda_{jj}(t)\rangle_{\rho(0)} \ . \tag{4.17}$$

Thus $\rho_{jj}(t)$ which solves the optical Bloch equations (3.18) determines the average rate of emission of photons [viz., (3.24)].

The unfiltered spectral intensity, which is the energy per second radiated between ω and $\omega+d\omega$ for the $j\to i$ transition, is

$$q_{ji}(\omega)=\epsilon_0c\int\hat{k}\cdot d\mathbf{s}\int\langle\mathbf{E}_{ji}(\mathbf{x},t)\cdot\mathbf{E}_{ij}(\mathbf{x},t+\tau)\rangle_{\rho(0)}e^{i\omega\tau}d\tau \ , \tag{4.18}$$

which is determined by the correlation

$$I_{ji}(t,t+\tau)=\mathrm{Tr}[\rho(0)\Lambda_{ji}(t)\Lambda_{ij}(t+\tau)] \ . \tag{4.19}$$

The steady-state value of this correlation is obtained by taking the $t\to\infty$ state as the initial condition for ρ. In this case (4.19) becomes

$$\lim_{t\to\infty}I_{ji}(t,t+\tau)=\mathrm{Tr}\hat{\rho}\,\mathrm{Tr}\tilde{\rho}(0)\Lambda_{ij}(\tau) \ , \tag{4.20}$$

where

$$\tilde{\rho}(0)=\hat{\rho}/\mathrm{Tr}\hat{\rho} \ ,$$
$$\hat{\rho}=\rho(\infty)\Lambda_{ji}(0) \ , \tag{4.21}$$
$$\rho(\infty)=\sum_{i,j}\rho_{ij}(\infty)\Lambda_{ij}(0) \ , \tag{4.22}$$

where $\rho_{ij}(\infty)=\langle\Lambda_{ji}(\infty)\rangle$ are the steady-state solutions to the optical Bloch equations (3.18). In the steady state the two-time correlation of the electric field is therefore proportional to

$$\lim_{t\to\infty}I_{ji}(t,t+\tau)=\mathrm{Tr}\left[\sum_{\alpha}\rho_{\alpha j}(\infty)\Lambda_{\alpha i}(0)\Lambda_{ij}(\tau)\right]$$
$$=\rho_{ij}(\infty)\rho_{ji}(\tau) \ , \tag{4.23}$$

where $\rho_{ji}(\tau)$ is the solution of (3.18) subject to the initial conditions $\rho_{ii}(0)=1$, $\rho_{ji}(0)=\rho_{jj}(\infty)/\rho_{ij}(\infty)$, and $\rho_{\alpha\beta}(0)=0$ for all other components. The matrix elements are subject to the usual normalization

$$\sum_{\alpha}\rho_{\alpha\alpha}=1 \ . \tag{4.24}$$

So, the spectral intensity of the $j\to i$ transition is proportional to the Fourier transform of the solution, $\rho_{ji}(\tau)$, of the optical Bloch equations evolving from the initial conditions (4.21) and (4.22). For $\tau>0$ the $\rho_{ji}(\tau)$ in (4.23) may be calculated from the optical Bloch equations as written in (3.18). However, for $\tau<0$ the $\rho_{ji}(\tau)$ must be calculated from the advanced optical Bloch equations where each β_{kj} is changed to $-\beta_{kj}^*$.

The physical density matrix ρ_{ij} [given by (3.2)] and the intensities $I_{jilk}(t_1,t_2)$ must have the property of positivity. For example, $\rho_{ij}=\rho_{ji}^*$ and $\rho_{ij}v_iv_j^*>0$ for all vectors v_i. This positivity requirement, however, does not apply to initial conditions such as (4.13) that determine the development of conditional correlations such as (4.14). Nevertheless, these correlations obey the Bloch equations in each variable t_i [with the external field in the coherent state $\mathbf{V}(\mathbf{x},t)$].

To evaluate the filtered intensities requires $F(t)$ for the given filter. A typical filter will pass light that is within a bandwidth γ of some frequency ω_F. In the time representation this filter has an $F(t)$ given by

$$F(t)=\gamma \cos\omega_F t e^{-\gamma|t|} , \tag{4.25}$$

so that

$$F(\omega)=\int_{-\infty}^{\infty} F(t)e^{i\omega t}dt = \frac{\gamma^2}{\gamma^2+(\omega+\omega_F)^2}+\frac{\gamma^2}{\gamma^2+(\omega-\omega_F)^2} .$$

A broadband filter has $F(t)=\delta(t)$, so that $F(\omega)=1$ everywhere. The narrow-band filter (4.25) has $F(\omega)=1$ only for the range of values $|\omega-\omega_F| \ll \gamma$.

V. CORRELATIONS IN PHOTON EMISSION FROM A TWO-LEVEL ATOM

For a two-level system driven by an external field, (3.14)–(3.17) yield the equation of motion for the amplitude, $C_i(t)$, where the atom is in state i and where also no photons have been emitted between some initial state and time t:

$$i\hbar\frac{dC_1}{dt}=\left[\hbar\omega_1-\frac{i}{2}\hbar\beta_1\right]C_1-\hbar\Omega_{10}(t)C_0 , \tag{5.1}$$

$$i\hbar\frac{dC_0}{dt}=\hbar\omega_0 C_0-\hbar\Omega^*_{10}(t)C_1 . \tag{5.2}$$

The rate at which the probability to be in the upper level decays is β_1, which is therefore the same as the Einstein A coefficient. Substantial oscillations in $|C_i|$ occur only when the external field is tuned sufficiently close to the atomic transition frequency, $\omega_1-\omega_0$. Thus we transform (5.1) and (5.2) to the slow variables

$$\bar{C}_i(t)=C_i(t)e^{i\omega_i t} , \tag{5.3}$$

$$\bar{\Omega}_i(t)=\Omega^+_{i0}(t)e^{+i(\omega_i-\omega_0)t} , \tag{5.4}$$

and obtain in the RWA

$$\frac{d\bar{C}_1}{dt}=-\tfrac{1}{2}\beta_1\bar{C}_1+i\bar{\Omega}_1(t)\bar{C}_0 , \tag{5.5}$$

$$\frac{d\bar{C}_0}{dt}=i\bar{\Omega}^*_1(t)\bar{C}_1 . \tag{5.6}$$

These equations describe situations where the rate of change in intensity of the external field is slow compared to $\omega_1-\omega_0$. A case of particular interest occurs when the laser frequency ω_{L_i} is sinusoidally detuned from a given transition(s) by an amount Δ_i, or

$$\omega_{L_i}=\omega_i-\omega_0+\Delta_i . \tag{5.7}$$

In this case equations with time-independent coefficients can be obtained by setting

$$\Omega^+_{i0}(t)=\Omega_{i,0}e^{-i(\omega_i-\omega_0+\Delta_i)t} , \tag{5.8}$$

$$\bar{c}_i=C_i e^{i(\omega_i+\Delta_i)t}, \quad i\neq 0 \tag{5.9}$$

$$\bar{c}_0=C_0^{i\omega_0 t} , \tag{5.10}$$

to yield

$$\frac{d\bar{c}_1}{dt}=(i\Delta-\tfrac{1}{2}\beta)\bar{c}_1+i\Omega\bar{c}_0 , \tag{5.11}$$

$$\frac{d\bar{c}_0}{dt}=i\Omega^*\bar{c}_1 . \tag{5.12}$$

The subscript 1 has now been dropped from β, Δ, Ω, and $\Omega\equiv\Omega_{i,0}=\Omega^+_{i0}(0)$.

The general solution to these equations is determined by the eigenvalues

$$\lambda_\pm=-\tfrac{1}{2}(\tfrac{1}{2}\beta-i\Delta)\pm\tfrac{1}{2}(\tfrac{1}{4}\beta^2-\Delta^2-4|\Omega|^2-i\beta\Delta)^{1/2} . \tag{5.13}$$

The solution for the probability amplitudes which starts out from $\bar{c}_0(0)=1$ is

$$\bar{c}_0=[1/(\lambda_+-\lambda_-)](\lambda_+e^{\lambda_- t}-\lambda_-e^{\lambda_+ t}) , \tag{5.14}$$

$$\bar{c}_1=[i\Omega/(\lambda_+-\lambda_-)](e^{\lambda_+ t}-e^{\lambda_- t}) . \tag{5.15}$$

According to (4.4) the observation of a scattered photon resets the atom to the ground state. Denote the reset time by $t=0$. After this reset the probability that the *next* photon is emitted between t and $t+dt$ is

$$\beta|\bar{c}^{\,2}_1(t)|dt , \tag{5.16}$$

so that the average time between photons is

$$\int_0^\infty \beta|\bar{c}_1(t)^2|t\, dt=\langle t_f\rangle , \tag{5.17}$$

which is proportional to the inverse of the intensity of fluorescence. By direct integration

$$\langle t_f\rangle=\frac{\beta^2+8|\Omega|^2+4\Delta^2}{4\beta|\Omega|^2} . \tag{5.18}$$

This equation describes the variation in fluorescent intensity as a function of Ω and Δ. For example, the ratio of intensity at a detuning Δ to that at resonant excitation ($\Delta=0$) is

$$\frac{I(\Delta)}{I(0)}=\frac{\beta^2+8|\Omega|^2}{\beta^2+8|\Omega|^2+4\Delta^2} . \tag{5.19}$$

The intensity of external illumination is proportional to $|\Omega|^2$. From (5.18) the ratio of fluorescent intensities at two different values ($\Omega_{\rm I},\Omega_{\rm II}$) of Ω, is, for resonant excitation,

$$\frac{I(\Omega_{\rm I})}{I(\Omega_{\rm II})}=\frac{|\Omega_{\rm I}|^2}{|\Omega_{\rm II}|^2}\frac{(\beta^2+8|\Omega_{\rm II}|^2)}{(\beta^2+8|\Omega_{\rm I}|^2)}, \quad \Delta=0 . \tag{5.20}$$

For $|\Omega_{\rm I}|$ and $|\Omega_{\rm II}|$ much less than β the scattered intensities are in the ratio of the exciting intensities.

For resonant excitation

$$\bar{c}_1(t)=\frac{i}{x}e^{-\beta t/4}\sin(|\Omega|xt) , \tag{5.21}$$

where in the underdamped case

$$x^2 \equiv \left[1-\frac{\beta^2}{16|\Omega|^2}\right] < 1 \ . \tag{5.22}$$

The time which the atom takes to build up a significant probability (~ 1) to be in the excited state is $\sim 1/x|\Omega|$. The probability of the next photon being emitted between t and $t+dt$ is, for short t,

$$\beta|\Omega|^2 t^2 dt \ . \tag{5.23}$$

Integrating (5.23) from 0 up till time t yields the number of next photons to be emitted between 0 and t,

$$\tfrac{1}{3}\beta|\Omega|^2 t^3 \ . \tag{5.24}$$

The fact that successive emissions from an atom are repelled is referred to as antibunching. Observation of the dependence of this phenomenon on the third power of t is beyond current experimental capabilities.

For strong detuning $\Delta \gg \beta, |\Omega|$ the eigenvalues are approximated by

$$\lambda_+ \cong -\tfrac{1}{2}\beta\left[1-\frac{|\Omega|^2}{\Delta^2}\right]+i\Delta\left[1+\frac{|\Omega|^2}{\Delta^2}\right] , \tag{5.25}$$

$$\lambda_- \cong -\tfrac{1}{2}\beta\left[\frac{|\Omega|^2}{\Delta^2}\right]+i\Delta\left[\frac{|\Omega|^2}{\Delta^2}\right] . \tag{5.26}$$

In this case there are two well-separated time scales which determine the relaxation of the atom. The probability of being in the excited state t seconds after the emission of the previous photon is to leading order

$$|\bar{c}_1(t)|^2 \cong \frac{|\Omega|^2}{\Delta^2}(e^{-\beta t}+e^{-\beta|\Omega|^2 t/\Delta^2}-2e^{-\beta t/2}\cos\Delta t) \ . \tag{5.27}$$

For very short times ($\Delta t \ll 1$) the probability rate of the next photon emission again follows (5.23). But for times such that $\Delta t \sim 1$, the probability for the case of strong detuning only builds up to the small value $|\Omega|^2/\Delta^2$. There are two time scales that determine the emission of next photons for strong detuning. These are a fast-time scale of order $1/\beta$ and a long-time scale of order $\Delta^2/\beta|\Omega|^2$. The ratio of next photons coming out after a long time to next photons coming out on the fast-time scale is $\Delta^2/|\Omega|^2 \gg 1$. So, if no emission is observed for a time $\gg 1/\beta$ but $\ll \Delta^2/|\Omega|^2\beta$ it can be concluded that the next photon will be of the slow variety. During the time between photons the probability that the atom is in the ground state is $1-|\Omega|^2/\Delta^2$, which is very close to unity. In view of the inefficiency of photodetection the observation of the null time between photons for a two-level atom is impractical. However, competing transitions make such observations possible in a three- or higher-level atom.

The optical Bloch equations for a two-level atom follow from (3.18). In the case of sinusoidal detuning (5.7) they become

$$\frac{d\bar{\rho}_{11}}{dt} = -\beta\bar{\rho}_{11}+i\Omega\bar{\rho}_{01}-i\Omega^*\bar{\rho}_{10} \ , \tag{5.28}$$

$$\frac{d\bar{\rho}_{01}}{dt} = -(i\Delta+\tfrac{1}{2}\beta)\bar{\rho}_{01}+i\Omega^*(\bar{\rho}_{11}-\bar{\rho}_{00}) \ , \tag{5.29}$$

$$\frac{d\bar{\rho}_{10}}{dt} = (i\Delta-\tfrac{1}{2}\beta)\bar{\rho}_{10}-i\Omega(\bar{\rho}_{11}-\bar{\rho}_{00}) \ . \tag{5.30}$$

In general, $\rho_{00}+\rho_{11}\equiv p$ is a constant of the motion; so as regards the physical density matrix it is therefore possible to impose

$$\rho_{00}+\rho_{11}=1 \ .$$

The slow-time scale has been introduced via

$$\begin{aligned} &\bar{\rho}_{ii}=\rho_{ii} \ , \\ &\bar{\rho}_{10}=\rho_{10}e^{i(\omega_1-\omega_0+\Delta)t} \ , \\ &\bar{\rho}_{01}=\rho_{01}e^{-i(\omega_1-\omega_0+\Delta)t} \ . \end{aligned} \tag{5.31}$$

In the steady state these equations yield

$$\bar{\rho}_{11}(\infty)=p|\Omega|^2/(\Delta^2+\tfrac{1}{4}\beta^2+2|\Omega|^2) \ , \tag{5.32}$$

$$\bar{\rho}_{01}(\infty)=\frac{-i\Omega^*(\tfrac{1}{2}\beta-i\Delta)}{\Delta^2+\tfrac{1}{4}\beta^2+2|\Omega|^2}p \ , \tag{5.33}$$

$$\bar{\rho}_{10}(\infty)=\bar{\rho}_{01}(\infty)^* \ .$$

Comparing (5.32) with (5.18) yields

$$\beta\bar{\rho}_{11}(\infty)=\langle t_f\rangle^{-1} \ . \tag{5.34}$$

Equation (5.34) relates a quantity determined by the optical Bloch equations to the first moment of the distribution function for next photons.

Equations (5.28)–(5.30) determine three eigenvalues that characterize the approach to the steady state. In order to evaluate these eigenvalues ($\lambda_1,\lambda_2,\lambda_3$) introduce the deviation from the steady state:

$$\delta\bar{\rho}_{ij}(t)=\bar{\rho}_{ij}(t)-\bar{\rho}_{ij}(\infty)\sim\rho'_{ij}e^{\lambda t} \ . \tag{5.35}$$

Substituting (5.35) into (5.28)–(5.30) yields

$$(\lambda+\beta)\rho'_{11}=i\Omega\rho'_{01}-i\Omega^*\rho'_{10} \ , \tag{5.36}$$

$$(\lambda+\tfrac{1}{2}\beta+i\Delta)\rho'_{01}=2i\Omega^*\rho'_{11} \ , \tag{5.37}$$

$$(\lambda+\tfrac{1}{2}\beta-i\Delta)\rho'_{10}=-2i\Omega\rho'_{11} \ . \tag{5.38}$$

The consistency condition for these equations is

$$(\lambda+\beta)(\lambda+\tfrac{1}{2}\beta+i\Delta)(\lambda+\tfrac{1}{2}\beta-i\Delta)+4|\Omega|^2(\lambda+\tfrac{1}{2}\beta)=0 \ . \tag{5.39}$$

For resonant tuning, $\Delta=0$, the roots of (5.39) are

$$\begin{aligned} &\lambda_1=-\tfrac{1}{2}\beta \ , \\ &\lambda_{2,3}=-\frac{3\beta}{4}\pm\frac{i}{2}(16|\Omega|^2-\tfrac{1}{4}\beta^2)^{1/2} \ , \end{aligned} \tag{5.40}$$

where if $\Delta \gg |\Omega|,\beta$ the roots are

$$\lambda_1=-\beta\left[1-\frac{2|\Omega|^2}{\Delta^2}\right] , \tag{5.41}$$

$$\lambda_2 = -\tfrac{1}{2}\beta - i\Delta\left[1+\frac{2|\Omega|^2}{\Delta^2}-\frac{2|\Omega|^4}{\Delta^4}\right], \tag{5.42}$$

$$\lambda_3 = -\tfrac{1}{2}\beta + i\Delta\left[1+\frac{2|\Omega|^2}{\Delta^2}-\frac{2|\Omega|^4}{\Delta^4}\right]. \tag{5.43}$$

In this limit the eigenvalues for the optical Bloch equations are qualitatively different from the eigenvalues for the next photon equation (5.25) and (5.26). In particular, (5.41)–(5.43) do not have a long-time scale where $\mathrm{Re}(-\lambda) \ll \beta$.

The steady-state spectral intensity of the fluorescent emission from the two-level atom is obtained by taking the Fourier transform of $\rho_{10}(\tau)$ subject to the initial conditions [viz., (4.23)]

$$\begin{aligned} &\rho_{10}(0)=\rho_{11}(\infty)/\rho_{01}(\infty)\ ,\\ &\rho_{00}(0)=1\ ,\\ &\rho_{11}(0)=0=\rho_{01}(0)\ . \end{aligned} \tag{5.44}$$

That is, one must solve the Bloch equations (5.28)–(5.30) for the $\rho_{10}(\tau)$ that evolves from the initial state (5.44). The steady-state "∞" values are given by (5.32) and (5.33) with $p=1$; in particular,

$$\frac{\bar{\rho}_{11}(\infty)}{\bar{\rho}_{01}(\infty)} = \frac{i\Omega}{\frac{1}{2}\beta - i\Delta}\ . \tag{5.45}$$

The solution to (5.28)–(5.30) for $\rho_{10}(\tau)$ takes the form

$$\rho_{10}(\tau) = \bar{\rho}_{10}(\infty)e^{-i\omega_L\tau} + \sum_{\alpha=1}^{3} d_\alpha e^{(\lambda_\alpha - i\omega_L)\tau}\ , \tag{5.46}$$

where the d_α are determined by the initial conditions ($\tau=0$) given by (5.44). Use of (5.46) in (4.18) and (4.19) yields a steady-state spectral intensity $q_{ji}(\omega)$ that is proportional to

$$\begin{aligned} \langle \Lambda_{10}^2(\omega)\rangle &\equiv \int T\gamma[\rho(0)\Lambda_{10}(t)\Lambda_{01}(t+\tau)]e^{i\omega\tau}d\tau \\ &= \bar{\rho}_{01}(\infty)\int \rho_{10}(\tau)e^{i\omega\tau}d\tau \\ &= \bar{\rho}_{10}(\infty)\bar{\rho}_{01}(\infty)2\pi\delta(\omega-\omega_L) - \left[\sum_{\alpha=1}^{3}\frac{d_\alpha\bar{\rho}_{01}(\infty)}{i(\omega-\omega_L)+\lambda_\alpha} + \text{c.c.}\right]. \end{aligned} \tag{5.47}$$

For the case of complete saturation ($\Omega \gg \beta, \Delta$), (5.47) becomes[9]

$$\langle \Lambda_{10}^2(\omega)\rangle = \left[\frac{\frac{1}{4}\beta^2+\Delta^2}{4|\Omega|^2}\right]2\pi\delta(\omega-\omega_L) + \frac{\beta/4}{(\omega-\omega_L)^2+\beta^2/4} + \frac{3\beta/16}{(\omega-\omega_L-2|\Omega|)^2+\frac{9}{16}\beta^2} + \frac{3\beta/16}{(\omega-\omega_L+2|\Omega|)^2+\frac{9}{16}\beta^2}\ . \tag{5.48}$$

For the case of strong detuning in a strong field $\Delta \gg |\Omega| \gg \beta$, (5.47) become[9]

$$\begin{aligned} \langle \Lambda_{10}^2(\omega)\rangle = {}& \frac{2\pi|\Omega|^2}{\Delta^2}\delta(\omega-\omega_L) + \frac{8\beta(|\Omega|/\Delta)^6}{(\omega-\omega_L)^2+\beta^2} \\ &+ \frac{\beta(|\Omega|/\Delta)^4}{(\omega-\omega_L-\Delta)^2+\beta^2/4} \\ &+ \frac{\beta(|\Omega|/\Delta)^4}{(\omega-\omega_L+\Delta)^2+\beta^2/4}\ . \end{aligned} \tag{5.49}$$

For a two-level system with a characteristic frequency $\omega_1-\omega_0$, the external drive leads to the appearance of sidebands described in various limits by (5.48) and (5.49). At this leading order of the perturbation expansion there are three peaks in the spectrum. For large detuning Δ these peaks can be well separated and the measurement of a photon as well as an associated peak or frequency is possible. In this case the observation of a photon is not accompanied by a reset of the atom to its ground state. Rather, the imposition of the filter causes the system to set itself into a state described by a density matrix of an incoherent state. Succeeding photons will evolve from this state, and correlations in the time ordering of the frequencies of successive photons appear. In particular, it is more probable to observe a higher-frequency photon ($\omega_L+\Delta$) followed by a lower-frequency photon ($\omega_L-\Delta$) than the reverse (for $\Delta>0$). Therefore a measurement of the frequency autocorrelation of scattered photons provides an arrow to time. As the widths of the sideband peaks is β the filters used for these measurements will have widths $\delta\omega$ such that

$$\beta \ll \delta\omega \ll \Delta\ . \tag{5.50}$$

The time between photons t_2-t_1 must satisfy

$$t_2-t_1 \gg 1/\delta\omega \tag{5.51}$$

so as not to conflict with the frequency resolution of the filter.

The joint rate for recording photons at t_1 and t_2, in the presence of filters, is given by (4.15). For the two-level system it yields

$$I_{1010}(t_1,t_2) = \sum_i \int I_{1i}(\tau)F_1(\tau)\int \langle 0|\Lambda_{10}(t_2-t_1)\Lambda_{01}(t_2'-t_1)|i\rangle F_2(t_2'-t_2)dt_2'd\tau\ , \tag{5.52}$$

where, following (4.19),

$$\bar{I}_{1i}(\tau)=\mathrm{Tr}[\rho(0)\Lambda_{1i}(0)\bar{\Lambda}_{01}(\tau)]=I_{1i}(\tau)e^{i\omega_L(t_i'-t_1)} , \tag{5.53}$$

and we will have in mind that $\rho(0)$ is the steady-state density matrix so that

$$\bar{I}_{10}(t)=z^2+4z^6e^{\lambda_1 t}+z^4e^{\lambda_2 t}+z^4e^{\lambda_3 t} , \tag{5.54}$$

$$\bar{I}_{11}(t)=\frac{\Omega}{\Delta}(1-2z^2)z^2[-1+2z^2+2z^2e^{\lambda_1 t}+(1-3z^2)e^{\lambda_2 t}-z^2e^{\lambda_3 t}] , \tag{5.55}$$

where $z^2\equiv|\Omega|^2/\Delta^2 \ll 1$ and these results apply for $t>0$. For $t<0$ the corresponding equations are obtained by changing β to $-\beta$ in (5.41)–(5.43). In (5.54) and (5.55) λ_1, λ_2, λ_3 correspond to the sidebands at ω_L, $\omega_L+\Delta$, $\omega_L-\Delta$, respectively. The terms which are independent of time correspond to elastic scattering at ω_L.

When a broadband filter is used $[F_1(\tau)=\delta(\tau)]$ the frequency is not measured, so that a subsequent photon is emitted from an atom starting off in the ground state $[I_{11}(0)=0]$. In this case

$$I_{1010}(t_1,t_2)=2\pi z^2\int\langle 0|\Lambda_{10}(t_2-t_1)\Lambda_{01}(t_2'-t_1)|0\rangle F_2(t_2'-t_2)dt_2' . \tag{5.56}$$

For a filter F_1 satisfying (5.50) the two-time intensity correlation for observing the first photon near $\omega_L+\Delta$ is

$$I^{(2)}_{1010}(t_1,t_2)=2\pi z^4\int\langle 0|\Lambda_{10}(t_2-t_1)\Lambda_{01}(t_2'-t_1)|0\rangle F_2(t_2'-t_2)dt_2'$$
$$+2\pi(\Omega/\Delta)z^2\int\langle 0|\Lambda_{10}(t_2-t_1)\Lambda_{01}(t_2'-t_1)|1\rangle F_2(t_2'-t_2)dt_2' . \tag{5.57}$$

When the first photon is observed in sideband 3 (near $\omega_L-\Delta$) the two-point correlation is

$$I^{(3)}_{1010}(t_1,t_2)=2\pi z^4\int\langle 0|\Lambda_{10}(t_2-t_1)\Lambda_{01}(t_2'-t_1)|0\rangle F_2(t_2'-t_2)dt_2'$$
$$-2\pi(\Omega/\Delta)z^4\int\langle 0|\Lambda_{10}(t_2-t_1)\Lambda_{01}(t_2'-t_1)|1\rangle F_2(t_2'-t_2)dt_2' . \tag{5.58}$$

Comparison of (5.57) and (5.58) shows that the probability for the second photon being emitted from the $1\to 0$ transition is diminished by a factor of z^2 if the first photon was observed near $\omega_L-\Delta$.

The complete calculation of (5.56)–(5.58) requires an evaluation of the amplitudes

$$\langle 0|\Lambda_{10}(t_2-t_1)\Lambda_{01}(t_2'-t_1)|\alpha\rangle\equiv\bar{f}_\alpha\exp[-i\omega_L(t_2'-t_2)] ,$$

where

$$\langle 0|\bar{\Lambda}_{10}(t_2-t_1)\bar{\Lambda}_{01}(t_2'-t_1)|\alpha\rangle\equiv\bar{f}_\alpha . \tag{5.59}$$

These amplitudes can be expressed in the form

$$\mathrm{Tr}[\rho(0)\bar{\Lambda}_{10}(t_2-t_1)\bar{\Lambda}_{01}(t_2'-t_1)]=\mathrm{Tr}[\bar{\rho}(t_2-t_1)\bar{\Lambda}_{10}(0)\bar{\Lambda}_{01}(\tau_2)]$$
$$=\bar{\rho}_{01}(t_2-t_1)\langle 0|\bar{\Lambda}_{01}(\tau_2)|0\rangle+\bar{\rho}_{11}(t_2-t_1)\langle 0|\bar{\Lambda}_{01}(\tau_2)|1\rangle , \tag{5.60}$$

where $\tau_2\equiv t_2'-t_2$ and for $\alpha=0$ in (5.59), $\rho(0)=\Lambda_{00}(0)$, whereas for $\alpha=1$ in (5.59), $\rho(0)=\Lambda_{10}(0)$. The Bloch equations yield $\bar{\rho}_{ij}(t_2-t_1)$.

When $\alpha=0$, $\rho_{ij}(0)=\delta_{i0}\delta_{j0}$ and

$$\bar{\rho}_{01}(t_2-t_1)=\frac{\Omega^*}{\Delta}[-1+2z^2+2z^2e^{\lambda_1(t_2-t_1)}+(1-3z^2)e^{\lambda_2(t_2-t_1)}-z^2e^{\lambda_3(t_2-t_1)}] , \tag{5.61}$$

$$\rho_{11}(t_2-t_1)=z^2+z^2e^{\lambda_1(t_2-t_1)}-z^2e^{\lambda_2(t_2-t_1)}-z^2e^{\lambda_3(t_2-t_1)} . \tag{5.62}$$

For the case where α is equal to unity $\rho_{ij}(0)=\delta_{i,1}\delta_{j0}$ and the time development of the density matrix yields

$$\bar{\rho}_{01}(t_2-t_1)=\frac{\Omega^*}{\Omega}[(2z^2-8z^4)e^{\lambda_1(t_2-t_1)}-(z^2-4z^4)e^{\lambda_2(t_2-t_1)}-(z^2-4z^4)e^{\lambda_3(t_2-t_1)}] , \tag{5.63}$$

$$\rho_{11}(t_2-t_1)=\frac{\Delta}{\Omega}[(z^2-4z^4)e^{\lambda_1(t_2-t_1)}+z^4e^{\lambda_2(t_2-t_1)}-(z^2-3z^4)e^{\lambda_3(t_2-t_1)}] . \tag{5.64}$$

When τ_2 is greater than zero one similarly finds

$$\langle 0|\bar{\Lambda}_{01}(\tau_2)|0\rangle=\frac{\Omega}{\Delta}[-1+2z^2+2z^2e^{\lambda_1\tau_2}-z^2e^{\lambda_2\tau_2}+(1-3z^2)e^{\lambda_3\tau_2}] , \tag{5.65}$$

$$\langle 0|\bar{\Lambda}_{01}(\tau_2)|1\rangle=(2z^2-8z^4)e^{\lambda_1\tau_2}+z^4e^{\lambda_2\tau_2}+(1-2z^2+7z^4)e^{\lambda_3\tau_2} . \tag{5.66}$$

The finite separation Δ between filters of the first and second photons leads to the appearance of terms in I_{1010} that involve $\cos[\Delta(t_2-t_1)]$ and its harmonics. These terms appear in $\bar{\rho}_{ij}(t_2-t_1)$ through the contributions proportional to $\exp[\lambda_2(t_2-t_1)]$ and $\exp[\lambda_3(t_2-t_1)]$. Mathematically, these oscillatory terms also appear as a result of the requirement that the integrations over t_1',t_2' can no longer run from $-\infty$ to $+\infty$. As measurements are meaningful only on time scales large compared to $1/\Delta$ [see (5.50) and (5.50)] we avoid these algebraic difficulties (arising from the incomplete convolutions contributing to I_{1010}) by averaging over times (t_2-t_1) long compared to $1/\Delta$ but short compared to $1/\beta$. Then, as regards the rate of high-frequency photons followed by low-frequency photons one finds from (5.57), (5.60), and (5.64),

$$I_{1010}^{(2,3)}(t_1,t_2)=4\pi^2\{z^4\exp[-\beta(t_2-t_1)]+z^6\} \ . \tag{5.67}$$

For times comparable to $1/\beta$ the term involving z^4 dominates but for very long times there of course remains a steady-state contribution. In deriving (5.67) the averaging over the Δ^{-1} time scale was effected by retaining the contribution involving $\exp[\lambda_1(t_2-t_1)]$. Similarly the rate of low-frequency photons followed by high-frequency photons is

$$I_{1010}^{(3,2)}=4\pi^2\{z^6+z^{10}\exp[-\beta(t_2-t_1)]\} \ . \tag{5.68}$$

Not only is (5.68) substantially smaller than (5.67) but for time scales t_2-t_1, satisfying

$$1/\Delta<t_2-t_1\sim 1/\beta \ ,$$

it fails to exhibit the clear photon bunching characteristic of the filtered measurement described by (5.67).

VI. INTERMITTENCY IN FLUORESCENCE IN A MULTILEVEL ATOM; WAVE-FUNCTION COLLAPSE DUE TO MEASUREMENTS WITH A NULL RESULT

For a two-level atom the periods between photons are difficult to observe in a sensible fashion because of the inherent experimental inefficiency of the photodetectors. However, in the V arrangement (Fig. 1) proposed by Dehmelt the fluorescence from the strong transition can be used as a meter for determining the period between photons when the long-time scale of the forbidden transition dominates the motion. Since the strong transition $0\leftrightarrow1$ can spew out over 10^8 photons/sec a turnoff of this signal can be a very sensitive (even classical) amplifier for dark periods longer than about $10/\beta_1$.

Use of the strong fluorescence as a meter for determining when the system is in a state of null emission makes the $C_i(t)$ directly observable. For the case of a three-level atom Eqs. (3.14) and (3.17) again yield the time development of the probability amplitude $C_i(t)$ that the atom will be in state i and that no photons will have been emitted between some reference time and t. For the V system in the presence of the external field $\mathbf{E}(t)$ one is led to equations of motion:

$$i\hbar\frac{dC_2}{dt}=\left[\hbar\omega_2-\frac{i}{2}\hbar\beta_2\right]C_2-\hbar\Omega_{20}(t)C_0-\frac{i}{2}\hbar\beta_{21}C_1 \ , \tag{6.1}$$

$$i\hbar\frac{dC_1}{dt}=\left[\hbar\omega_1-\frac{i}{2}\hbar\beta_1\right]C_1-\hbar\Omega_{10}(t)C_0-\frac{i}{2}\hbar\beta_{12}C_2 \ , \tag{6.2}$$

$$i\hbar\frac{dC_0}{dt}=\hbar\omega_0C_0-\hbar\Omega_{10}^*(t)C_1-\hbar\Omega_{20}^*(t)C_2 \ . \tag{6.3}$$

Except for the Onsager-type cross terms these equations are identical to those presented by Cohen-Tannoudji and Dalibard.[14] The decay coefficients β_1 and β_2 are shorthand for β_{11} and β_{22}, and $\mu_{12}=0$.

If the slowly varying envelope functions are introduced,

$$\begin{aligned}\bar{\Omega}_i(t)&=\Omega_{i0}^+(t)e^{i(\omega_i-\omega_0)t} \ ,\\ \bar{C}_i(t)&=C_i(t)e^{i\omega_i t} \ ,\end{aligned} \tag{6.4}$$

and the electric field is assumed to have components approximately tuned to the $0\leftrightarrow1$ as well as $0\leftrightarrow2$ transitions then (6.1)–(6.3) become (in the RWA)

$$\frac{d\bar{C}_2}{dt}=-\tfrac{1}{2}\beta_2\bar{C}_2+i\bar{\Omega}_2(t)\bar{C}_0 \ , \tag{6.5}$$

$$\frac{d\bar{C}_1}{dt}=-\tfrac{1}{2}\beta_1\bar{C}_1+i\bar{\Omega}_1(t)\bar{C}_0 \ , \tag{6.6}$$

$$\frac{d\bar{C}_0}{dt}=+i\bar{\Omega}_1^*(t)\bar{C}_1+i\bar{\Omega}_2^*(t)\bar{C}_2 \ . \tag{6.7}$$

Equations (6.5)–(6.7) describe the response for quite general imposed external fields. The $\bar{\Omega}_i(t)$ should be thought of as the envelope which modulates the resonant component of $\mathbf{E}(t)$ for the $i\leftrightarrow0$ transition. If the laser fields illuminating the two transitions are sinusoidally detuned so that (5.7) and (5.8) apply then the equations for $\bar{c}_i(t)$, defined by (5.9), are given by (1.34)–(1.36). We note that the quantum Onsager cross terms involving β_{21} and β_{12} drop out in the RWA when degeneracy is absent.

The time development of the $\bar{c}_i$ is determined by three eigenvalues λ_α such that $\bar{c}_i$ is proportional to terms in $\exp\lambda_\alpha t$ or

$$\bar{c}^{\,i}=\sum_{j=0}^{2}c_{ij}\exp\lambda_j t \ . \tag{6.8}$$

The c_{ij} are determined by initial conditions and the λ_α are determined by the cubic equation

$$\begin{aligned}(\lambda-i\Delta_2+\tfrac{1}{2}\beta_2)[\lambda^2+(\tfrac{1}{2}\beta_1-i\Delta_1)\lambda+|\Omega_{1,0}|^2]&\\ =-|\Omega_{2,0}|^2(\lambda-i\Delta_1+\tfrac{1}{2}\beta_1) \ .&\end{aligned} \tag{6.9}$$

Consider first the case $\Delta_1=0$ which has all the requisite features of the telegraph, and which leads to the roots

$$\lambda_0=-\tfrac{1}{4}\beta_1+i|\Omega_{1,0}|x+\epsilon_0 \ , \tag{6.10}$$

$$\lambda_1=-\tfrac{1}{4}\beta_1-i|\Omega_{1,0}|x+\epsilon_1 \ , \tag{6.11}$$

$$\lambda_2 = -\tfrac{1}{2}\beta_2 + i\Delta_2 + \epsilon_2 \;, \tag{6.12}$$

where

$$x \equiv [1-(\beta_1^2/16|\Omega_{1,0}|^2)]^{1/2} \tag{6.13}$$

is the saturation parameter. There are three cases: (i) underdamped resonance $\beta_1^2/16|\Omega_{1,0}|^2 \ll 1$, (ii) critical resonance $|x| \ll 1$, and (iii) overdamped response $\beta_1^2/16|\Omega_{1,0}|^2 \gg 1$. The existence of the telegraph (or intermittent fluorescence) requires a wide separation of time scales or

$$\beta_2 \ll \beta_1 \;, \tag{6.14}$$

$$|\Omega_{2,0}| \ll |\Omega_{1,0}| \;, \tag{6.15}$$

$$|\mathrm{Re}\epsilon_2| \ll \beta_1 \;, \tag{6.16}$$

$$|\mathrm{Im}\epsilon_2| \ll |\Omega_{1,0}| \;. \tag{6.17}$$

The slow decay constant β_l of Sec. I is in this notation given by $-\mathrm{Re}\lambda_2$. For the underdamped case (i) the eigenvalues can be expanded in powers of the small parameter $|\Omega_{2,0}|^2$ so that

$$\epsilon_0 = \frac{|\Omega_{2,0}|^2(\frac{1}{4}\beta_1 + i|\Omega_{1,0}|x)}{(2i|\Omega_{1,0}|x)(\frac{1}{4}\beta_1 + i\Delta_2 - i|\Omega_{1,0}|x)} \;, \tag{6.18}$$

$$\epsilon_1 = \frac{-|\Omega_{2,0}|^2(\frac{1}{4}\beta_1 - i|\Omega_{1,0}|x)}{(2i|\Omega_{1,0}|x)(\frac{1}{4}\beta_1 + i\Delta_2 + i|\Omega_{1,0}|x)} \;, \tag{6.19}$$

$$\epsilon_2 = \frac{-|\Omega_{2,0}|^2(i\Delta_2 + \frac{1}{2}\beta_1)}{|\Omega_{1,0}|^2 - \Delta_2^2 + \frac{1}{2}i\Delta_2\beta_1} \;. \tag{6.20}$$

In view of (6.20), which applies for all x, the inequalities (6.16) and (6.17) imply that

$$\frac{\frac{1}{2}|\Omega_{2,0}|^2|\Omega_{1,0}|^2}{(|\Omega_{1,0}|^2-\Delta_2^2)^2 + \frac{1}{4}\Delta_2^2\beta_1^2} \ll 1 \;, \tag{6.21}$$

$$\frac{|\Omega_{2,0}|^2\Delta_2\,||\Omega_{1,0}|^2 - \Delta_2^2 - \frac{1}{4}\beta_1^2|}{(|\Omega_{1,0}|^2-\Delta_2^2)^2 + \frac{1}{4}\Delta_2^2\beta_1^2} \ll |\Omega_{1,0}| \;. \tag{6.22}$$

In view of the Markovian nature of the photon emission (which applies in the RWA) the atom is reset to the ground state after each photon is fluoresced. Thus the solution to (1.34)–(1.36), subject to the initial condition $\bar{c}_0 = 1$, $\bar{c}_1 = \bar{c}_2 = 0$, describes the probability of dark periods between photons. For this case the long-time probability to be in the forbidden level is determined by

$$c_{22} = \tfrac{1}{2}\Omega_{2,0}\frac{(i\beta_1 - 2\Delta_2)}{|\Omega_{1,0}|^2 - \Delta_2^2 + \frac{1}{2}i\beta_1\Delta_2} = -\frac{\epsilon_2}{i\Omega_{2,0}^*} \;. \tag{6.23}$$

There is also a long-time probability amplitude to be in the strong or ground states given by

$$c_{12} = -2\frac{\epsilon_2^2\Omega_{1,0}}{|\Omega_{2,0}|^2}\frac{1}{(i\beta_1 - 2\Delta_2)} \;, \tag{6.24}$$

$$c_{02} = \epsilon_2^2/|\Omega_{2,0}|^2 \;. \tag{6.25}$$

The short-time probabilities to be in the strongly excited state and ground state are to lowest order in $|\Omega_{2,0}|$

$$c_{10} = -c_{11} \;, \tag{6.26}$$

$$c_{11} = -\frac{\Omega_{1,0}}{2|\Omega_{1,0}|x} \;, \tag{6.27}$$

$$c_{0,1} = \frac{-\frac{1}{4}\beta_1 + i|\Omega_{1,0}|x}{2i|\Omega_{1,0}|x} \;, \tag{6.28}$$

$$c_{00} = \frac{\frac{1}{4}\beta_1 + i|\Omega_{1,0}|x}{2i|\Omega_{1,0}|x} \;. \tag{6.29}$$

And the short-time contribution to the forbidden level is

$$c_{20} = \frac{\Omega_{2,0}}{2|\Omega_{1,0}|x}\frac{\frac{1}{4}\beta_1 + i|\Omega_{1,0}|x}{[-\frac{1}{4}\beta_1 + i(|\Omega_{1,0}|x - \Delta_2)]} \;, \tag{6.30}$$

$$c_{21} = \frac{\Omega_{2,0}}{2|\Omega_{1,0}|x}\frac{(\frac{1}{4}\beta_1 - i|\Omega_{1,0}|x)}{(\frac{1}{4}\beta_1 + i|\Omega_{1,0}|x + \Delta_2)} \;. \tag{6.31}$$

The probability that after a reset there will be a dark period of length greater than t, when $t \gg 1/\beta_1$, is

$$W(t) = (|c_{02}|^2 + |c_{12}|^2 + |c_{22}|^2)\exp(\lambda_2 + \lambda_2^*)t \;. \tag{6.32}$$

And the probability that a dark period of some length will occur is $W(t=0)$. This function is peaked at $\Delta_2 = |\Omega_{1,0}|$ (with a width β_1). For such tuning the inequality (6.16) requires

$$|\Omega_{2,0}|/\beta_1 \ll 1 \;, \tag{6.33}$$

and in this case

$$\epsilon_2 = -2|\Omega_{2,0}|^2/\beta_1 + i|\Omega_{2,0}|^2/|\Omega_{1,0}| \;.$$

For resonantly tuned coupling to level 2 so that $\Delta_2 = \Omega_1$ the long-time (dark) probability to be in the strong level is down from that to be in the forbidden level by the factor

$$\frac{|c_{12}|^2}{|c_{22}|^2} = \frac{4|\Omega_{2,0}|^2}{\beta_1^2} \;. \tag{6.34}$$

Regardless of the length of the dark period there is still a finite probability that the atom is shelved in the strong level $|1\rangle$ as opposed to forbidden level $|2\rangle$. In fact, the probability that a dark period will end with the emission of a strong transition photon $(1\to 0)$ compared to ending with a $2\to 0$ photon is

$$\frac{\beta_1|c_{12}|^2}{\beta_2|c_{22}|^2} = \frac{4|\Omega_{2,0}|^2}{\beta_1\beta_2} \;. \tag{6.35}$$

This quantity is not necessarily small. When this quantity is large the overwhelming probability is that a dark period ends with the emission of a $1\to 0$ photon. This effect can be understood in terms of the currents (2.17) which flow in the "atom" during the radiationless, dark period between photons.

The evaluation of the atomic currents at some time t requires the wave function of the atom during a dark period. The $C_i(t)$ are the amplitude for the atom to be in state i with no photons scattered between 0 and t, but if the system is actually observed to be dark between 0 and t then the wave function at time t is

$$|\Psi\rangle=\frac{1}{\left[\sum_j |C_j^2(t)|\right]^{1/2}}\sum_i C_i(t)|i,0\rangle \ . \tag{6.36}$$

If there actually is no fluorescence then the amplitude to be in state i is

$$C_i(t)\Big/\left[\sum_j |C_j^2(t)|\right]^{1/2} .$$

The factor $[\sum_j |C_j^2(t)|]^{1/2}$ normalizes $|\Psi\rangle$ in view of the wave-function collapse brought about by the null measurement: that is the observation of a period of darkness. The observation of no scattered photons for a time t since the last reset ($t=0$) changes $|\Psi\rangle$ as follows:

$$|\Psi(t)\rangle \to \frac{\mathcal{P}|\Psi(t)\rangle}{\langle\Psi(t)|\mathcal{P}|\Psi(t)\rangle} , \tag{6.37}$$

where $\mathcal{P}$ is the projection given by (3.10).

The observation of darkness for all times since the last reset leaves the atom in a fully determined state. In general, the observation of darkness in the interval of time between t' and t (where $t'<t$) is a non-Markovian measurement. Although such a measurement changes the state (or density matrix) of the system it still leaves the system in a final state that is determined by its past history (i.e., its state at time t'). This non-Markovian response should be contrasted with the process of photon observation which always involves a Markovian reset.

From (2.17) the expected value of the current t seconds into a dark period is given by

$$\mathbf{J}_0(t)=\frac{1}{2i}\sum_{i,j}(\omega_i-\omega_j)\boldsymbol{\mu}_{ij}C_j(t)C_i^*(t)\Big/\sum_\alpha |C_\alpha^2(t)| . \tag{6.38}$$

For the three-level system under consideration we find

$$\mathbf{J}_0(t)=\left[1\Big/\sum_\alpha |\bar{c}_\alpha^{\,2}(t)|\right]\left[\frac{1}{2i}(\omega_1-\omega_0)\boldsymbol{\mu}_{10}e^{-i(\omega_0-\omega_1)t}\bar{c}_0(t)\bar{c}_1^{\,*}(t)+\frac{1}{2i}(\omega_2-\omega_0)\boldsymbol{\mu}_{20}e^{-i(\omega_0-\omega_2-\Delta_2)t}\bar{c}_0(t)\bar{c}_2^{\,*}(t)+\text{c.c.}\right] . \tag{6.39}$$

The quantum radiationless current has contributions from the $0\leftrightarrow 1$ as well as $0\leftrightarrow 2$ transitions. At long times $|\bar{c}_2|^2 \gg |\bar{c}_1|^2$ (if darkness is verified) but the ratio of the squares of the respective contributions to J_0 is given by (6.35) when $\Delta_2=\Omega_1$. The fact that the long-time current for the strong transition can equal that due to the forbidden transition accounts for the possibility that dark periods can end with a $1\to 0$ emission.

After a reset, the ratio of the probabilities of initiating a dark period, for the cases $\Delta_2=0$ and $\Delta_2=|\Omega_{1,0}|$, is given by the small quantity

$$\frac{W(t=0,\ \Delta_2=0)}{W(t=0,\ \Delta_2=|\Omega_{1,0}|)}=\left[\frac{\beta_1^2}{4|\Omega_{1,0}|^2}\right]^2 . \tag{6.40}$$

The critical time T_c during which darkness must be observed so as to collapse the wave function into the forbidden level $|2\rangle$ is determined by

$$|\bar{c}_2^{\,2}(T_c)|=|\bar{c}_1^{\,2}(T_c)|+|\bar{c}_0^{\,2}(T_c)| \tag{6.41}$$

or

$$|c_{22}|^2=\exp(-\tfrac{1}{2}\beta_1 T_c) . \tag{6.42}$$

For $\Delta_1=0$, $\Delta_2=|\Omega_{1,0}|$, the critical time is found to be

$$T_c=-\frac{2}{\beta_1}\ln|c_{22}|^2=\frac{4}{\beta_1}\ln(\beta_1/|\Omega_{2,0}|) . \tag{6.43}$$

In the overdamped case ϵ_2, c_{22} still obey (6.20) and (6.23) but the strong transition eigenvalue λ_1 is greatly decreased:

$$\lambda_0=-\tfrac{1}{4}\beta_1(1+\bar{x})+\epsilon_0 , \tag{6.44}$$

$$\lambda_1=-\tfrac{1}{4}\beta_1(1-\bar{x})+\epsilon_1 , \tag{6.45}$$

where

$$\epsilon_0=-\tfrac{1}{2}|\Omega_{2,0}|^2\frac{(1-\bar{x})}{\bar{x}}\frac{1}{\tfrac{1}{4}\beta_1(1+\bar{x})+i\Delta_2} , \tag{6.46}$$

$$\epsilon_1=\tfrac{1}{2}|\Omega_{2,0}|^2\frac{(1+\bar{x})}{\bar{x}}\frac{1}{\tfrac{1}{4}\beta_1(1-\bar{x})+i\Delta_2} , \tag{6.47}$$

$$\bar{x}\equiv[1-(16|\Omega_{1,0}|^2/\beta_1^2)]^{1/2} . \tag{6.48}$$

For this overdamped case the maximum probability of a dark period is determined by

$$\Delta_2=0 . \tag{6.49}$$

In this case we determine

$$c_{22}=-\frac{1}{2}i\frac{\Omega_{2,0}\beta_1}{|\Omega_{1,0}|^2} , \tag{6.50}$$

$$\epsilon_2=-\frac{1}{2}\beta_1\frac{|\Omega_{2,0}|^2}{|\Omega_{1,0}|^2} . \tag{6.51}$$

The tuning width in Δ_2 for this maximum is rather sharp; it is given by

$$\Delta_2'=\beta_1\left[\sqrt{8}\frac{|\Omega_{1,0}|^2}{\beta_1^2}\right] \ll \beta_1 . \tag{6.52}$$

The existence of a solution to (6.9) with well-separated time scales requires

$$|c_{22}|^2 \ll 1 . \tag{6.53}$$

In the overdamped case this requirement can be realized by comparing ϵ_1 to $\beta_1(1-\bar{x})$ in the limit $\bar{x}\to 1$. For the overdamped case the collapse time is stretched out to

$$T_c=\frac{\beta_1^2}{2|\Omega_{1,0}|^2}\frac{1}{\beta_1}\ln\left[\frac{2|\Omega_{1,0}|^2}{\beta_1|\Omega_{2,0}|}\right] . \tag{6.54}$$

When the strong transition is driven off resonance so that

$$\Delta_1 \gg |\Omega_{1,0}| \gg \beta_1 , \tag{6.55}$$

then λ_0 and λ_1 are given to lowest order in $|\Omega_{2,0}|^2$ by (5.25) and (5.26), and ϵ_2, c_2 are determined by (6.23). The maximal contribution to c_{22} occurs for

$$\Delta_2 = \Delta_1 \frac{|\Omega_{1,0}|^2}{\Delta_1^2} \tag{6.56}$$

and in this case

$$c_{22} = \frac{\Omega_{2,0}\Delta_1^2}{\beta_1 |\Omega_{1,0}|^2} . \tag{6.57}$$

As before, (6.53) must apply. The collapse time is then given by

$$T_c = \frac{2\Delta_1^2}{|\Omega_{1,0}|^2} \frac{1}{\beta_1} \ln \left[\frac{|\Omega_{1,0}|^2 \beta_1}{\Delta_1^2 |\Omega_{2,0}|} \right] . \tag{6.58}$$

During bright periods the rate of fluorescence from the strong transition is $\beta_1 |\bar{c}_1(t)|^2$ evaluated for $t \ll T_c$. In the overdamped case and in the strong detuning case (6.55) the $1 \to 0$ intensity is down by a large factor which is proportional to the stretching out of the collapse time T_c. That is, the intensity of strong fluorescence is down by the factors $|\Omega_{1,0}|^2/\beta_1^2$ and $|\Omega_{1,0}|^2/\Delta_1^2$ in the cases corresponding to (6.54) and (6.58), respectively. In fact, the number of strong transition events which must be missed in order to collapse the atome into $|2\rangle$ is in all cases given by

$$n_D = -2 \log |c_{22}| . \tag{6.59}$$

The percentage of time that the system is dark p_D can be estimated using the average time between resets, $\langle t_f \rangle$, of the fast (strong) emissions. In particular,

$$p_D = (1-p_D)|c_{22}|^2 \langle t_D \rangle / \langle t_f \rangle , \tag{6.60}$$

where $\langle t_D \rangle$ is the average span of a single dark period. Equation (6.60) can be understood by noting that $(1-p_D)/\langle t_f \rangle$ is the average number of resets per second, and $|c_{22}|^2$ is the probability that a given reset leads to a dark period of length $\langle t_D \rangle$. Solving (6.60) for p_D yields

$$p_D = \frac{|c_{22}|^2 \langle t_D \rangle / \langle t_f \rangle}{1 + |c_{22}|^2 \langle t_D \rangle / \langle t_f \rangle} . \tag{6.61}$$

The average span of a dark period can be found by setting the atom in $|2\rangle$ at $t=0$; this yields $\langle t_D \rangle = 1/2|\mathrm{Re}\lambda_2|$. At saturation (6.61) then becomes (1.1) supplemented by (1.10).

In parallel with the sideband correlations in two-level resonance (Sec. V) the two-time correlations for the emission of $|1\rangle$-$|0\rangle$ and $|2\rangle$-$|0\rangle$ photons also depends upon the order in which they are recorded. From (4.4) we have in the steady state

$$I_{1020}(t_1,t_2) = \rho_{11}(\infty)\langle 0|\Lambda_{22}(t_2-t_1)|0\rangle , \tag{6.62}$$

$$I_{2010}(t_1,t_2) = \rho_{22}(\infty)\langle 0|\Lambda_{11}(t_2-t_1)|0\rangle . \tag{6.63}$$

If $\rho_{ij}(t_2-t_1)$ is the solution to the optical Bloch equations then

$$\rho_{ij}(t_2-t_1) = \rho_{ij}(\infty) + \sum_{\alpha=1}^{8} d_{ij}^{(\infty)} e^{\lambda_\alpha (t_2-t_1)} . \tag{6.64}$$

Imposition of the initial condition $\rho_{ij}(0) = \delta_{i0}\delta_{j0}$ leads to

$$\langle 0|\Lambda_{ii}(t_2-t_1)|0\rangle = \rho_{ii}(t_2-t_1) . \tag{6.65}$$

An estimate of the solution (6.64) indicates that (6.62) and (6.63) are quite different; the ratio is about $|\Omega_{2,0}|^2/\beta_1^2$ for short time. Thus the two-time correlations indicate an underlying irreversibility.

Note added. Since the submission of this paper there have appeared a number of contributions[20–22] that have a direct bearing on the issues discussed in this paper.

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[1]H. Dehmelt, in *Laser Spectroscopy V*, Vol. 3 of *Springer Series in Optical Sciences*, edited by A. R. W. Mckellar, T. Oka, and B. P. Stoicheff (Springer, Berlin, 1981); in *Advances in Laser Spectroscopy*, Vol. 95 of *NATO Advanced Study Institute, Series B: Physics*, edited by F. T. Arecchi, F. Strumia, and H. Walther (Plenum, New York, 1983).

[2]T. Erber and S. Putterman, Nature (London) **318**, 41 (1985).

[3]M. Porrati and S. Putterman, Phys. Rev. A **36**, 929 (1987).

[4]J. C. Bergquist, R. G. Hulet, M. W. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).

[5]W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986).

[6]Th. Sauter, W. Neuhauser, R. Blatt, and P. E. Toschek, Phys. Rev. Lett. **57**, 1696 (1968); Opt. Commun. **60**, 287 (1986).

[7]W. M. Itano, J. C. Bergquist, R. G. Hulet, and D. J. Wineland, Phys. Rev. Lett. **59**, 2732 (1987).

[8]T. Erber, P. Hammerling, M. Porrati, G. Hockney, and S. Putterman, Ann. Phys. (to be published).

[9]H. J. Kimble and L. Mandel, Phys. Rev. A **13**, 2123 (1976).

[10]B. R. Mollow, Phys. Rev. A **12**, 1919 (1975).

[11]L. Allen and J. M. Eberly, *Optical Resonance and Two Level Atoms* (Wiley-Interscience, New York, 1975); R. Loudon, *The*

Quantum Theory of Light, 2nd ed. (Clarendon, Oxford, 1983).

[12]F. T. Arecchi, A. Schenzle, R. G. De Voe, K. Jungmann, and R. G. Brewer, Phys. Rev. A **33**, 2124 (1986); J. Javanainen, *ibid.* **33**, 2121 (1986).

[13]H. J. Kimble, R. J. Cook, and A. L. Wells, Phys. Rev. A **34**, 3190 (1986); G. Nienhuis, *ibid.* **35**, 4639 (1987).

[14]C. Cohen-Tannoudji and J. Dalibard, Europhys. Lett. **1**, 441 (1986).

[15]A. Schenzle and R. G. Brewer, Phys. Rev. A **34**, 3127 (1986).

[16]P. Zoller, M. Marte, and D. F. Walls, Phys. Rev. A **35**, 198 (1987).

[17]H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977).

[18]A. Aspect, G. Roger, S. Reynaud, J. Dalibard, and C. Cohen-Tannoudji, Phys. Rev. Lett. **45**, 617 (1980); C. Cohen-Tannoudji and S. Reynaud, Philos. Trans. R. Soc. London, Ser. A **293**, 223 (1979); P. A. Apanasevich and S. Ja. Kilin, Phys. Lett. **62A**, 83 (1977); S. Reynaud, Ann. Phys. (Paris) **8**, 315 (1983).

[19]J. R. Ackerhalt, Phys. Rev. A **17**, 471 (1978).

[20]S. Reynaud, J. Dalibard, and C. Cohen-Tannoudji, IEEE J. Quantum. Electron. **24**, 1395 (1988).

[21]M. Ligare, Phys. Rev. A **37**, 3293 (1988).

[22]R. J. Cook, Phys. Scr. **T21**, 49 (1988).