Coherence with incoherent light: A new type of quantum beats for a single atom

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Even with incoherent driving, coherence effects may show up in the spontaneous emissions of an atom. We derive a general master equation and predict that for both the three-level V and Λ systems the intensity correlation function $g^{(2)}(\tau)$ may exhibit pronounced oscillations with frequency $\delta\omega$ if the level separation $\hbar\delta\omega$ is small. We obtain a marked difference in the beat amplitude for a single Λ system as compared to a beam or gas. If $\delta\omega$ is much smaller than the natural linewidth we find dark and light periods in the spontaneous emission.

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I. INTRODUCTION

If a three-level systems with two upper levels and a common ground state, a so-called V system, with small separation $\hbar\delta\omega$ of the upper levels, is initially excited by a sharp laser pulse or by a passage through a foil, one observes an exponential decay with oscillations superimposed. These are the familiar quantum beats [1], and they are due to a coherence of the upper levels. For a Λ system, which has transitions from one upper to two lower levels, they do not occur [2]. For laser-driven systems a similar coherence is responsible for dark resonances [3], and it may even lead to macroscopic dark periods [4] without a metastable state, in contrast to the dark periods of the Dehmelt system [5]. For incoherent driving, on the other hand, one often assumes that atomic coherence plays no role [6]. It was shown in Ref. [7] in the case of large level separations for a Λ system or ladder system that finite laser bandwidths tend to destroy population trapping and dark resonances, unless the two lasers are cross correlated.

In this paper we study systems continuously driven by incoherent light and their spontaneous photon emission. It is predicted that for both the V and Λ system the intensity correlation function $g^{(2)}(\tau)$ will exhibit pronounced oscillations with frequency $\delta\omega$ if the level separation $\hbar\delta\omega$ between the two upper and lower levels, respectively, is assumed small, but still larger than the natural linewidth. These oscillations are new quantum beats which arise, as the familiar ones for V systems, from coherence of neighboring atomic states, a coherence not destroyed despite the incoherent pumping. The beats should be observable for a single system in a trap. For the Λ system there is a marked difference in the beat amplitude for a beam and a single system: the amplitude is much larger for the latter. For $\delta\omega$ much smaller than the natural linewidth we predict dark periods for a single V or Λ system if the dipole transition moments are parallel. We propose to investigate the beats of frequency $\delta\omega$ for their possible use in a frequency standard.

II. DESCRIPTION OF ENSEMBLE VERSUS SINGLE SYSTEM

Ensemble master equation. We first consider quite generally an ensemble of N-level atoms interacting with the quantized radiation field $\mathbf{E}$ and with an external field $\mathbf{E}_e$ which is linearly polarized, broadband, stochastic, stationary, and of zero mean. We write $\mathbf{E}_e = E_e(t)\mathbf{E}_0$ and denote its spectral energy density by $W(\omega)$ [8]. The standard Hamiltonian in dipole form is given by [9]

$$H = H_A^0 + H_R^0 + e\mathbf{D}\cdot(\mathbf{E}_e + \mathbf{E}),$$

where $H_A^0 = \sum_i \hbar\,\omega_i \mid i\rangle\langle i \mid$ is the Hamiltonian for the atom, H_R^0 that for the radiation field, and where $\mathbf{D} = \sum_{i,j} \mathbf{D}_{ij} \mid i\rangle\langle j \mid$, with $e\mathbf{D}_{ij} = e\langle i \mid \mathbf{X} \mid j\rangle$ the electric dipole moment for the i-j transition. Different atoms will see different realizations of E_e. Therefore the density matrix, $\rho_{\rm tot}$, for atoms plus radiation fields includes an average $\langle\,\rangle_e$ over the external field, so that $\rho_{\rm tot} = \langle\rho^e_{\rm tot}\rangle_e$ where $\rho^e_{\rm tot}$ refers to a particular realization of E_e.

The reduced atomic density operator is $\rho(t) = {\rm tr}_R\,\rho_{\rm tot}(t)$ where ${\rm tr}_R$ is the partial trace over the states of the radiation field. To obtain a differential equation for ρ we go to the interaction picture with respect to the atomic Hamiltonian and calculate $\Delta\rho_I \equiv \rho_I(t+\Delta t) - \rho_I(t)$ in perturbation theory. In second order $\mathbf{E}_e$ contributes

$$- e^2\,\hbar^{-2}\left\langle \int_t^{t+\Delta t} dt' \int_t^{t'} dt''[\mathbf{D}(t')\cdot\mathbf{E}_e(t'),[\mathbf{D}(t'')\cdot\mathbf{E}_e(t''),\ \rho_I^e(t)]]\right\rangle_e \tag{1}$$

and $\mathbf{E}$ a similar term in which $\langle\,\rangle_e$ and $\rho_I^e(t)$ are replaced by ${\rm tr}_R$ and $\rho_I^{\rm tot}$, respectively. We denote by τ_e the correlation time of $\mathbf{E}_e$ and assume $\tau_e \ll \Delta t$. Then $\mathbf{E}_e(t')$ and $\mathbf{E}_e(t'')$ are, in good approximation, independent of $\rho_I^e(t)$. Hence the averaging can be performed for $\rho_I^e(t)$ separately, giving ρ_I. Insertion of $\mathbf{D}(t)$ gives

$$-e^2\,\hbar^{-2}\sum_{i,j,k,l}(\mathbf{D}_{ij}\cdot\mathbf{E}_0)\,(\mathbf{D}_{kl}\cdot\mathbf{E}_0)\,[\,|\,i\rangle\langle j\,|,[|\,k\rangle\langle l\,|,\rho_I]\,]\times\int_t^{t+\Delta t}dt'\;e^{i(\omega_{ij}+\omega_{kl})t'}\int_t^{t'}dt''\;e^{-i\omega_{kl}(t'-t'')}\langle E_e(t'-t'')E_e(0)\rangle_e, \tag{2}$$

where $\omega_{ij}\equiv\omega_i-\omega_j$. Setting $\tau=t'-t''$ the range for the inner integral becomes $0\le\tau\le t'-t$. Since the correlation function of E_e is assumed to vanish rapidly for $\tau>\tau_e$ and since $\Delta t\gg\tau_e$ one may extend the τ integral to ∞. Then the real part of this integral becomes $\pi W(\omega_{kl})/2\varepsilon_0$ [8]. The imaginary part can be shown to be negligible if W is sufficiently broad and slowly varying around the transition frequencies. The remaining t' integral is practically $\Delta t\exp\{i(\omega_{ij}+\omega_{kl})t\}$ if $(\omega_{ij}+\omega_{kl})\Delta t$ is small compared to 1 [10]. We use the abbreviation $W_{ijkl}\equiv(\mathbf{D}_{ij}\cdot\mathbf{E}_0)(\mathbf{D}_{kl}\cdot\mathbf{E}_0)\;W(\omega_{kl})\pi e^2/2\epsilon_0\hbar^2$. Back in the Schrödinger picture Eq. (1) then becomes

$$-\sum_{i,j,k,l}W_{ijkl}[\,|\,i\rangle\langle j\,|,[|\,k\rangle\langle l\,|,\rho]\,].$$

By the same argument the first-order contribution vanishes. The contribution from the radiation field is the same as in the optical Bloch equations without lasers [11]. It leads to damping terms involving quantities of the form

$$\Gamma_{ijkl}\;\equiv\;\mathbf{D}_{ij}\cdot\mathbf{D}_{kl}\,|\,\omega_{kl}\,|^3\,e^2/6\pi\varepsilon_0\hbar c^3 \tag{3}$$

due to spontaneous emissions [12]. With $\Gamma\equiv\sum_{i,k>j}\Gamma_{ijjk}\,|\,i\rangle\langle k\,|$ one then obtains from Eqs. (1) to (3) for the atomic density operator in the Schrödinger picture

$$\begin{aligned}\dot\rho=&-i\left\{\left(H_A^0-i\,\Gamma\right)\rho-\rho\left(H_A^0-i\Gamma\right)^*\right\}\\&-\sum_{i,j,k,l}W_{jikl}\;[\,|\,i\rangle\langle j\,|,[|\,k\rangle\langle l\,|,\rho]\,]\\&+\sum_{\substack{i,j,k,l\\ i>j\\ k>l}}(\Gamma_{jikl}+\Gamma_{klij})\,|\,j\rangle\langle i\,|\,\rho\,|\,k\rangle\langle l\,|,\end{aligned} \tag{4}$$

where a difference quotient has been replaced by a differential quotient [13]. The Γ terms in Eq. (4) arise from the quantized radiation field, the W terms from the incoherent classical driving field. For large level separations the off-diagonal terms will give rapidly oscillating contributions, as seen in the interaction picture, and can therefore be neglected, leading to effective rate equations. For smaller level separation this is in general not so.

Single atom. The photon statistics for a single system can be determined by the technique of repeated *gedanken* measurements [14–16]. This method gives the no-photon probability [18, 19] $P_0(t;\rho_0)$, where ρ_0 is an initial state, and the reset matrix [16] to which the atom has to be reset after a photon detection. An analysis similar to that in Ref. [14] then shows that the no-photon probability can be written as $P_0(t)=\mathrm{tr}\rho^{(0)}(t)$ where the density matrix $\rho^{(0)}/\mathrm{tr}\rho^{(0)}$ describes a single atom between spontaneous emission and ρ^0 satisfies Eq. (4) without the last sum. With the notation $W\equiv\sum_{i,j,k}W_{ijjk}\,|\,i\rangle\langle k\,|$ and $H_{\rm eff}\equiv H_A^0-i(\Gamma+W)$ one can write

$$\begin{aligned}\dot\rho^{(0)}=&-i\left(H_{\rm eff}\rho^{(0)}-\rho^{(0)}H_{\rm eff}^*\right)\\&+\sum_{i,j,k,l}(W_{ijkl}+W_{klij})\,|\,i\rangle\langle j\,|\,\rho^{(0)}\,|\,k\rangle\langle l\,|\,.\end{aligned} \tag{5}$$

Since a single atom interacts with a particular realization of the external field $\mathbf{E}_e$ there should, in principle, be no averaging over $\mathbf{E}_e$. However, one can show that for most of the relevant time-averaged statistical quantities, such as the photon correlation functions for a single atom, one may use an ensemble average over the external field, due to ergodicity. This averaging leads from a pure-state or wave-function description as in Refs. [14, 15] to a density matrix.

III. APPLICATIONS: QUANTUM BEATS AND DARK PERIODS

V system. Here one has $\mathbf{D}_{21}$, $\mathbf{D}_{31}\neq0$, while $\mathbf{D}_{32}=\mathbf{0}$ due to parity conservation. One can choose both as real. Then Eq. (4) becomes, with the abbreviations $W_{ij}\equiv W_{i11j},\Gamma_{ij}\equiv\Gamma_{i11j}$, and $\delta\omega\equiv\omega_3-\omega_2$,

$$\begin{aligned}\dot\rho_{22}=&\;2\,W_{22}\rho_{11}-2(W_{22}+\Gamma_{22})\rho_{22}\\&-(W_{23}+\Gamma_{23})(\rho_{23}+\rho_{32}),\\ \dot\rho_{33}=&\;2\,W_{33}\rho_{11}-2(W_{33}+\Gamma_{33})\rho_{33}\\&-(W_{32}+\Gamma_{32})(\rho_{23}+\rho_{32}),\\ \dot\rho_{23}=&\;(W_{23}+W_{32})\rho_{11}-(W_{32}+\Gamma_{32})\rho_{22}\\&-(W_{23}+\Gamma_{23})\rho_{33}\\&-(i\delta\omega-W_{22}-W_{33}-\Gamma_{22}-\Gamma_{33})\rho_{23}\;.\end{aligned} \tag{6}$$

Furthermore, $\rho_{11}=1-\rho_{11}-\rho_{22}$ and $\rho_{32}=\bar\rho_{23}$ [20]. Recall that $2\Gamma_{ii}$ is an Einstein coefficient and $2W_{ii}$ a pumping rate. With the column vector $\boldsymbol{\rho}=(\rho_{11},\rho_{22},\rho_{33},\rho_{23},\rho_{32})^t$, where t denotes transpose, Eq. (6) can be written in the form $d\boldsymbol{\rho}/dt=\underline{M}\,\boldsymbol{\rho}$ where the 5×5 matrix $\underline{M}$ is read off Eq. (6).

We now discuss the case of small separation of the upper levels, $\delta\omega\equiv\omega_{32}\ll\omega_{31},\,\omega_{21}$ and first consider $\delta\omega\gg\Gamma,W$. In the matrix $\underline{M}$ the dominant term then is $\pm i\delta\omega$. $\underline{M}$ has 0 as an eigenvalue because $\mathrm{tr}\rho=1$, two further eigenvalues are real, and the other two are given to excellent accuracy by

$$\mu_{4,5}=\pm i\delta\omega-W_{22}-W_{33}-\Gamma_{22}-\Gamma_{33}\;. \tag{7}$$

Hence ρ contains terms which oscillate with frequency $\delta\omega$.

These oscillations show up in $g^{(2)}(\tau)$, the intensity correlation function, which is essentially the conditional probability density to find a spontaneous photon at time τ provided there was one at $t = 0$. From the general expression [21] one has

$$g^{(2)}(\tau) = \{ 2\Gamma_{22}\, \rho_{22}(\tau) + 2\Gamma_{33}\rho_{33}(\tau) + (\Gamma_{23} + \Gamma_{32})[\rho_{23}(\tau) + \rho_{32}(\tau)]\}/\bar{I}, \quad (8)$$

where $\bar{I}$ is the mean number of photons per second [22]. The initial condition is $\rho(0) = |1\rangle\langle 1|$ since after a photon detection a V system is in the ground state [16]. By Eqs. (7) and (8), $g^{(2)}(\tau)$ has an oscillatory component with frequency $\delta\omega$. The amplitude of these oscillations is proportional to $\delta\omega^{-1}$ if $\mathbf{D}_{21} \cdot \mathbf{D}_{31} \neq 0$ and proportional to $\delta\omega^{-2}$ otherwise, provided $\mathbf{E}_e$ is not perpendicular to $\mathbf{D}_{21}$ or $\mathbf{D}_{31}$. Figure 1 shows these oscillations.

The underlying reason for these oscillations is very similar to that for the usual quantum beats in the decay after a sudden excitation. As seen in second-order perturbation theory levels 2 and 3 are indirectly coupled via the transitions $2 \to 1 \to 3$ and $3 \to 1 \to 2$, mediated both by a virtual photon emission and reabsorption if $\mathbf{D}_{21} \cdot \mathbf{D}_{31} \neq 0$ and by $\mathbf{E}_e$. This leads to a coherence between the two upper levels.

We now discuss the case $\delta\omega \ll \Gamma, W$ and $\mathbf{D}_{21}$ parallel to $\mathbf{D}_{31}$. For the above $\rho^{(0)}$ we define $\boldsymbol{\rho}^{(0)}$ analogously to $\boldsymbol{\rho}$. The equation for $\rho^{(0)}$ can also be written in matrix form, $d\boldsymbol{\rho}^{(0)}/dt = \underline{M}^{(0)}\, \boldsymbol{\rho}^{(0)}$. In the present case $\underline{M}^{(0)}$ can be shown to have an eigenvalue with real part much closer to 0 than those of the other eigenvalues. This leads to a slowly decreasing part in the no-photon probability $P_0(t) = \mathrm{tr}\rho^{(0)}(t)$, and this in turn leads to dark periods

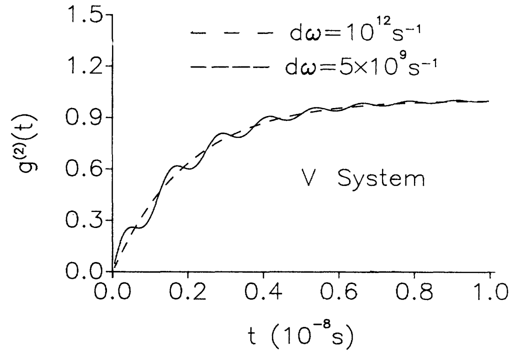

FIG. 1. The intensity correlation function $g^{(2)}(\tau)$ plotted for two different level separations $\delta\omega$, $\Gamma_{ij} = 10^8\ \mathrm{s}^{-1}$ and $W_{ij} = 10^8\ \mathrm{s}^{-1}$. For $\delta\omega = 5 \times 10^9\ \mathrm{s}^{-1}$ there are pronounced oscillations.

in the fluorescent light from such a single V system illuminated by incoherent light. By methods explained in Ref. [4] we find for their respective mean duration, in terms of $q \equiv D_{21}/D_{31}$,

$$T_D = \delta\omega^{-2}(W_{33} + \Gamma_{33})(1 + q^2)^3/2\, q^2, \quad (9)$$

$$T_L/T_D = (2\, W_{33} + \Gamma_{33})/W_{33}. \quad (10)$$

These expressions can be rewritten in a form symmetric in 2 and 3. For increasing pumping rate, T_D and T_L increase and T_L/T_D approaches 2. For decreasing $\delta\omega$ the light and dark periods become longer and longer.

Λ system. Here $\mathbf{D}_{31}$ and $\mathbf{D}_{32}$ are nonzero and can be chosen real, while $\mathbf{D}_{21} = \mathbf{0}$. With the abbreviation $\hat{W}_{ij} \equiv W_{i33j}, \hat{\Gamma}_{ij} \equiv \Gamma_{i33j}$, and $\delta\omega = \omega_2 - \omega_1$, Eq. (4) becomes

$$\begin{aligned} \dot{\rho}_{22} &= -2\, \hat{W}_{22}\, \rho_{22} + 2\left(\hat{W}_{22} + \hat{\Gamma}_{22}\right)\rho_{33} - \hat{W}_{12}(\rho_{21} + \rho_{12}), \\ \dot{\rho}_{33} &= 2\, \hat{W}_{11}\, \rho_{11} + 2\, \hat{W}_{22}\rho_{22} - 2\left(\hat{W}_{11} + \hat{W}_{22} + \hat{\Gamma}_{11} + \hat{\Gamma}_{22}\right)\rho_{33} + \left(\hat{W}_{12} + \hat{W}_{21}\right)(\rho_{21} + \rho_{12}), \\ \dot{\rho}_{12} &= \hat{W}_{12}\, \rho_{11} + \hat{W}_{21}\, \rho_{22} + \left(\hat{W}_{12} + \hat{W}_{21} + \hat{\Gamma}_{12} + \hat{\Gamma}_{21}\right)\rho_{33} + \left(i\delta\omega - \hat{W}_{11} - \hat{W}_{22}\right)\rho_{12}. \end{aligned} \quad (11)$$

In addition, $\rho_{21} = \bar{\rho}_{12}$ and $\mathrm{tr}\rho = 1$. Here $2\hat{\Gamma}_{ii}$ is the Einstein coefficient for the i-3 transition. We write Eq. (11) as in Eq. (6) in the form $d\boldsymbol{\rho}/dt = \underline{\hat{M}}\boldsymbol{\rho}$.

For $\delta\omega \gg \hat{W}, \hat{\Gamma}$ the matrix $\underline{\hat{M}}$ has two complex eigenvalues $\hat{\mu}_{4,5} = \pm i\delta\omega - (\hat{W}_{11} + \hat{W}_{22})/2$. The correlation function [21] is now $g^{(2)}(\tau) = \rho_{33}(\tau)/\rho_{33}(\infty)$ and may therefore exhibit oscillations with frequency $\delta\omega$. This in itself is interesting since, if excited by a short laser pulse, single Λ systems do not show quantum beats in their decay [2]. Even more interesting is the sensitive dependence of the oscillation amplitude on the initial condition $\rho(0)$. Starting from one of the two ground states the amplitude decreases, rather rapidly, as $1/\delta\omega^2$ while starting from a coherent superposition of the two ground states the amplitude decreases only as $1/\delta\omega$. Thus starting from a *thermal* mixture of the two ground states, as appropriate for a gas witout cooperative effects or a beam, the oscilla-

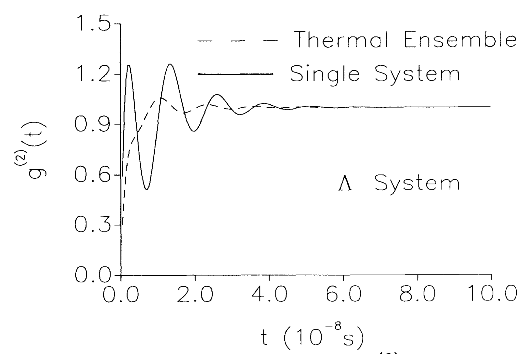

FIG. 2. The intensity correlation $g^{(2)}(\tau)$ for different initial states, the thermal mixture appropriate for a gas without cooperative effects or a beam (dotted curve) and the reset matrix for a single atom (solid curve), with $\Gamma_{ij} = 10^8\ \mathrm{s}^{-1}$, $W_{ij} = 10^8\ \mathrm{s}^{-1}$, $\delta\omega = 5 \times 10^8\ \mathrm{s}^{-1}$. In both cases $g^{(2)}(\tau)$ oscillates, but with much larger amplitude for the single system.

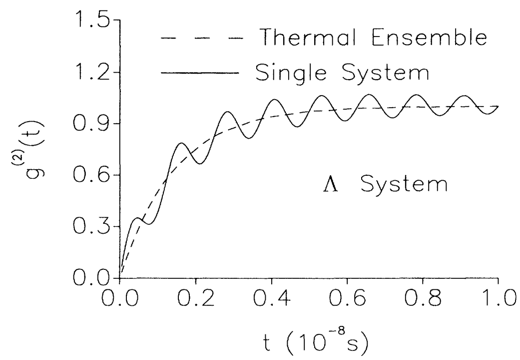

FIG. 3. The same situation as in Fig. 2, but with larger level separation, $\delta\omega = 5 \times 10^9$ s^{-1}. While the oscillations disappear in a thermal ensemble, they persist for a single system.

tions are very small and vanish rapidly for increasing $\delta\omega$. On the other hand, a single Λ system in a trap starts out, after each photon emission, from the reset matrix [16] $\sum_{i,j} \hat{\Gamma}_{ij} \mid i\rangle\langle j \mid /(\hat{\Gamma}_{22} + \hat{\Gamma}_{33})$; cf. in this context also Ref. [17].

Thus for a single Λ system the appropriate initial state is this reset matrix, and hence the correlation function of a single Λ system will show much more pronounced oscillations than that of a gas or beam. This is illustrated in Figs. 2 and 3.

For the case $\delta\omega \ll \Gamma, W$ and parallel transition dipole moments $\mathbf{D}_{31}, \mathbf{D}_{32}$, the Λ system also exhibits dark and light periods when illuminated by incoherent light. Again by the methods of Ref. [4] we find for their respective mean durations, in terms of $\hat{q} \equiv D_{32}/D_{31}$,

$$T_D = \delta\omega^{-2}\ \hat{W}_{33}\left(1 + \hat{q}^2\right)^3/2\hat{q}^2, \tag{12}$$

$$T_L/T_D = (2\ \hat{W}_{33} + \hat{\Gamma}_{33})/(\hat{W}_{33} + \hat{\Gamma}_{33}) \ . \tag{13}$$

A comparison with Eq. (9) shows that the dark and light periods are shorter for the Λ system.

IV. CONCLUSIONS

We have considered an N-level system illuminated by incoherent light and emitting spontaneous photons. A master equation was derived which becomes effectively the usual set of rate equations if the level separations are large, e.g., $\omega_i - \omega_j$ in the optical range. However, if two levels have only a small separation $\hbar\delta\omega$, then off-diagonal terms in the master equation may lead to coherence of atomic states. Specifically, we have studied the V and Λ systems for small $\delta\omega$ and have shown the existence of a new type of quantum beats with period $\delta\omega$ in their intensity correlation function. These may in principle be observed for a single system in a trap. We obtain a marked difference in the beat amplitude for a gas or atomic beam of Λ systems, assumed to be in a thermal mixture at the beginning of the irradiation, and a single Λ system. For the latter the oscillations are much more pronounced. We propose to investigate these oscillations for possible use in a frequency standard. If $\delta\omega$ is much smaller than the natural linewidths of the transitions then both a single V and Λ system may exhibit dark and light periods if the transition dipole moments are parallel. All these effects arise from atomic coherence surviving the incoherent illumination.

An experimental verification of the predicted effects should be possible with a single ion in a Paul trap along the lines of the recent experiment of Toschek and coworkers [23]. They measured the correlation function $g_2(\tau)$ for a single ion irradiated by narrow-band lasers. To adapt this to the present case one would only have to use broad-band illumination. With a similar setup it should also be possible to verify the predicted dark periods.

ACKNOWLEDGMENT

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[1] Cf., e.g., S. Haroche, in *High Resolution Laser Spectroscopy*, edited by K. Shimoda (Springer, Berlin, 1976), p. 25, and references therein.
[2] R. M. Herman, H. Grotch, R. Kornblith, and J. H. Eberly, Phys. Rev. A **11**, 1389 (1975); W. W. Chow, M. O. Scully, and J. O. Stoner, Jr., *ibid.* **11**, 1380 (1975).
[3] D. A. Cardimona, M. G. Raymer, and C. R. Stroud, Jr., J. Phys. B **15**, 55 (1982); E. Arimondo and G. Orriols, Nuovo Cimento Lett. **17**, 333 (1976); G. Orriols, Nuovo Cimento B **53**, 1 (1979).
[4] G. C. Hegerfeldt and M. B. Plenio, Phys. Rev. A **46**, 373 (1992).
[5] H. G. Dehmelt, Bull. Am. Phys. Soc. **20**, 60 (1975).
[6] If this is justified one can use ordinary rate equations. An exception was discussed for a four-level system in the context of lasing without inversion by M. Fleischhauer, C. H. Keitel, M. O. Scully, and C. Su, Opt. Commun. **87**, 109 (1992).
[7] B. J. Dalton and P. L. Knight, J. Phys. B **15**, 3397 (1982).
[8] If $\langle\rangle_e$ denotes averaging over the external field then $\pi W(\omega)/\epsilon_0$ is the Fourier transform of $\langle E_e(t)E_e(0)\rangle_e$, by the Wiener-Khinchin theorem.
[9] R. Loudon, *The Quantum Theory of Light*, 2nd ed. (Clarendon, Oxford, 1983).
[10] If $(\omega_{ij} + \omega_{kl})\Delta t$ is large compared to 1 one obtains approximately 0. But one may still use the former expression since the exponential now oscillates rapidly and will contribute negligibly anyway. An alternative argument below avoids the introduction of Δt.
[11] P. W. Milonni, Phys. Rep. **25C**, 1 (1976); M. Porrati and S. Putterman, Phys. Rev. A **39**, 3010 (1989).
[12] Note that $2\Gamma_{lkkl}$ is the Einstein coefficient for the spontaneous k-l transition and that $2W_{lkkl}$ is a pumping rate.
[13] In an alternative derivation one integrates $\dot{\rho}^I_{\rm tot}(t) = -i\left[H_I, \rho^I_{\rm tot}(t)\right]/\hbar$ from 0 to t and reinserts the result for $\rho^I_{\rm tot}(t)$ on the right side. Performing the average

$\langle\,\rangle_e$ one can, in good approximation, pull out $\langle \rho_I^e(t') \rangle_e \equiv \rho_I(t')$ from the average. Then the analysis is similar as before, leading to Eq. (4).

[14] G. C. Hegerfeldt and T. S. Wilser, in *Proceedings of the Second International Wigner Symposium, Goslar, 1991*, edited by H. D. Doebner, W. Scherer, and F. Schroeck (World Scientific, Singapore, 1992).

[15] J. Dalibard, Y. Castin, and K. Mølmer, Phys. Rev. Lett. **68**, 580 (1992); H. J. Carmichael (unpublished).

[16] G. C. Hegerfeldt, Phys. Rev. A **47**, 449 (1993).

[17] G. S. Agarwal and S. S. Jha, Z. Phys. B **35**, 391 (1979).

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

[19] H. J. Carmichael, S. Singh, R. Vyas, and P. R. Price, Phys. Rev. A **39**, 1200 (1989).

[20] The related equation of C. Cohen-Tannoudji, in *Atomic Physics IV*, edited by G. zu Putlitz, E. Weber, and A. Winnacker (Plenum, New York, 1975), p. 589, differs from ours since they do not contain the important Γ_{23} and Γ_{32} terms.

[21] The general expression is $g^{(2)}(t) = \mathrm{tr}(\Gamma + \Gamma^*)\rho(\tau)/\bar{I}$; cf., e.g., Ref. [16].

[22] From Eq. (6), with $W \equiv 0$, the numerator of Eq. (8) is just the increase of ρ_{11} due to spontaneous emissions.

[23] M. Schubert, I. Siemers, R. Blatt, W. Neuhauser, and P. E. Toschek, Phys. Rev. Lett. **68**, 3016 (1992).