Macroscopic dark periods without a metastable state

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The well-known three-level system of Dehmelt [Bull. Am. Phys. Soc. **20**, 60 (1975)] exhibits macroscopic dark periods because one of the upper levels is metastable. In this paper we show that one can also have the same phenomenon for a three-level-atom system with *both* upper states rapidly decaying to the ground state, provided (a) their level separation is very small and (b) their dipole moments for the ground-state transition are parallel. For suitably chosen laser parameters we also exhibit an exact nonradiating solution of the complete time development for atom plus radiation field. This leads to the surprising effect that the atom, after emission of a number of photons, will stop to fluoresce although still irradiated by the laser. The mean number of photons emitted before this switching off of the atom is calculated, as well as the mean duration of the dark periods for the general case.

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I. INTRODUCTION

Macroscopic dark periods of a single fluorescent atom were predicted by Dehmelt [1] for a system with two excited states, one rapidly decaying and the other metastable. Driving such a system by two lasers one intuitively expects frequent transitions from the ground state to the nonmetastable excited state with the subsequent emission of a spontaneous photon (light period). Once in a while there will be a transition to the metastable state, where the electron will stay for an extended period, and there will be no photons ("dark period," "electron shelving"). These ideas have been analyzed semiclassically by the telegraph process [2] as well as quantum mechanically [3,4]. Macroscopic dark periods were indeed found experimentally for single atoms in a Paul trap [5], confirming a spectacular quantum effect.

In this paper we present an alternative mechanism for dark periods which is not based on the existence of a metastable state but rather on a quantum coherence and interference phenomenon similar to the one responsible for a type of nonabsorption resonances studied by Orriols [6] and Cardimona, Raymer, and Stroud [7], for some population trappings [8] and lasing without inversion [9, 10]. We deal with a V system, a three-level atom consisting of two excited states optically coupled to a common ground state.

The basic feature is the existence of a coherent superposition of atomic states which does not couple to the quantized radiation field nor to the laser. For two degenerate states this was already pointed out by Dicke [11], and the corresponding superposition is invariant in time, except for an overall phase. For a Λ system, which consists of two nondegenerate ground states optically coupled to a common excited state, this is also true, provided one makes the rotating-wave approximation (RWA). Now, however, due to the nondegeneracy, the purely atomic part of the time development operator will create a time-dependent relative phase, and the superposition is no longer invariant.

For a V system as in Fig. 1, with a single laser, one can obtain a superposition of $|2\rangle$ and $|3\rangle$ which does not couple to the electromagnetic field if their dipole moments are parallel. Again one needs the RWA, and the purely atomic time development will create a time-dependent relative phase. The existence of such a superposition is the deeper reason for the vanishing of spontaneous emission in these systems when driven by a laser of a particular frequency as found in Refs. [6, 7]. Cardimona [12] applied similar ideas to a V system interacting with a single quantized mode in a cavity, constructed an invariant state of the combined system for a special mode frequency and exhibited nonrevivals of atomic excitations.

We study a single V system as in Fig. 1 with parallel or nearly parallel transition dipole moments $e\mathbf{D}_{21}$ and $e\mathbf{D}_{31}$ and very small level separation $\delta\omega$. This is similar to the system of Ref. [7], except that our $\delta\omega$ will be much smaller. The possible existence of dark periods may now be understood in analogy to the Dehmelt system, and the following qualitative remarks will be substantiated by a quantitative analysis. There exists a superposition, denoted by $|a\rangle$, of $|2\rangle$ and $|3\rangle$ which does not couple to the electromagnetic field. The noncoupling may be seen as a quantum interference phenomenon since the spontaneous transition probability to the lower level is

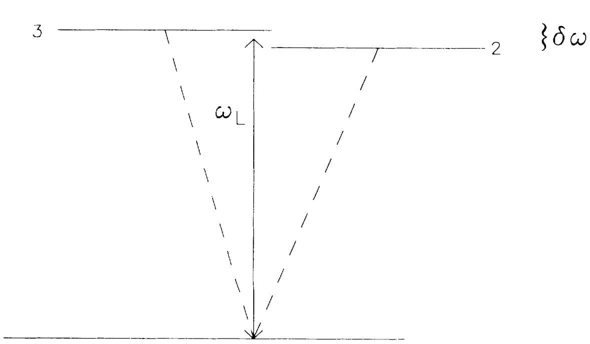

FIG. 1. V system with energy separation $\hbar\ \delta\omega$ of the excited levels; ω_L denotes the laser frequency.

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obtained by squaring the sum of two probability amplitudes, and coherence can lead to interference terms. This superposition $| a\rangle$ may be compared to the metastable state of the Dehmelt system, and the orthogonal superposition, denoted by $| s\rangle$, to the rapidly decaying state. After a photon emission one starts out from the ground state, and the laser will build up an $| s\rangle$ component. This will generally lead to a photon emission and back to the ground state so that the cycle can start all over again. In case of degeneracy, $\delta\omega = 0$, one would effectively have a two-level system, with the state $| a\rangle$ completely stable and invariant in time. However, for $\delta\omega \neq 0$, the purely atomic time development mixes $| a\rangle$ and $| s\rangle$, and this the faster the bigger $\delta\omega$ is. It will turn out that there may be a small probability for a delayed photon emission, and during this time the purely atomic time development may partially change the $| s\rangle$ component to an $| a\rangle$ component while the laser cooperates to decrease the remaining $| s\rangle$ component even further. In this way one may reach a state with a large $| a\rangle$ component and thus with a low transition probability to the ground state. This is the beginning of a dark period.

The quantitative analysis will show that indeed macroscopic dark and light periods exist if $\delta\omega$ is much smaller than the Einstein coefficients of level $| 2\rangle$ and $| 3\rangle$ as well as the Rabi frequencies. If $\delta\omega$ is larger, of the order of the Rabi frequencies, then one may either have long dark periods alternating with extremely short light periods of a few photons only—and thus difficult to observe—with an overall very low average fluorescence intensity, or the dark periods become so short as to merge with the waiting time between photons.

For a special laser tuning—in case of equal and parallel dipole moments halfway between the upper levels—we find that after the emission of a number of photons the atomic fluorescence is switched off, and the atom remains dark. The average number of photons emitted before the switching off depends on the parameters of atom and laser. The ensuing infinitely long dark period is related to the nonabsorption resonance found in Refs. [6,7]. We explicitly determine the corresponding nonradiating state of the *complete* system consisting of atom and quantized radiation field with an external classical laser field. For a single mode in a cavity interacting with such a V system a similar state was found in Ref. [12].

For the study of light and dark periods in Sec. II we use the probability $P_0(t)$ of finding no photon until time t provided a photon was emitted at $t = 0$. The importance of this probability for the study of dark periods was first stressed and applied to the Dehmelt system by Cohen-Tannoudji and Dalibard [3]. The form of $P_0(t)$ needed here is, however, different from the one used in Ref. [3] for the Dehmelt system. The general form of $P_0(t)$ has been determined by Porrati and Putterman [13]; an alternative and independent derivation can be found in Ref. [14]. For a V system with two closely spaced upper levels there arise important additional generalized damping constants which were absent in the form of $P_0(t)$ used in Ref. [3]. Related damping terms also arise in the Bloch equation for this system [7, 16]. In Sec. III light and dark periods are determined analytically.

II. DETERMINATION OF EMISSION BEHAVIOR

We consider a V system as in Fig. 1 with energy levels 2 and 3 close together but not degenerate. A single laser with its frequency ω_L somewhere in the vicinity of 2 and 3 is treated classically. The Hamiltonian in dipole form [15] for the atom interacting with the radiation field in RWA is given by

$$\begin{aligned} H/\hbar = &\sum_{i=2}^{3} \omega_i \,| i\rangle\langle i \,| \;+\; \sum_{\mathbf{k},\lambda} \omega_{\mathbf{k}\lambda} a^\dagger_{\mathbf{k}\lambda} a_{\mathbf{k}\lambda} \\ &+\sum_{i=2}^{3} \tfrac{1}{2}\,\Omega_i \left\{ e^{-i\omega_1 t} \,| i\rangle\langle 1 \,| \;+\; \text{H.c.}\right\} \\ &+i \sum_{i=2}^{3} \sum_{\mathbf{k},\lambda} g_{i\mathbf{k}\lambda} \left\{ a_{\mathbf{k}\lambda} \,| i\rangle\langle 1 \,| \;-\; \text{H.c.}\right\} . \end{aligned} \tag{1}$$

Because of the closeness of the upper levels the laser couples to both of them. Here

$$g_{i\mathbf{k}\lambda} = e\, \mathbf{D}_{i1} \cdot \epsilon_{\mathbf{k}\lambda}\, (\omega_{\mathbf{k}}/2\epsilon_0 \hbar V)^{1/2} \tag{2}$$

with $e\mathbf{D}_{i1} = e\langle i \mid \mathbf{X} \mid 1\rangle$ the dipole moment for the transition from the excited level i to the ground state. Ω_i is the Rabi frequency corresponding to the same transition.

$$\Omega_i = \hbar^{-1}\, e\, \mathbf{E}_L \cdot \mathbf{D}_{i1} . \tag{3}$$

V is a quantization volume, later taken to infinity. By parity conservation there is no dipole transition between $| 3\rangle$ and $| 2\rangle$.

We assume that right after an emission, at $t = 0$ say, the atom is in its ground state $| 1\rangle$. Then the stochastic behavior of the next emission is governed by $P_0(t)$, the probability of finding no photon before t [3]. We can apply the results of Refs. [13, 14] which in our situation, with a single laser coupled to $| 2\rangle$ and $| 3\rangle$, can be rewritten slightly to yield [17]

$$P_0(t) = \| \exp\{-iH_R t\} \,| 1\rangle \|^2 \tag{4}$$

where, in matrix form with respect to the atomic basis $| 1\rangle, | 2\rangle, | 3\rangle$, the nonHermitian reduced Hamiltonian H_R in the atomic space is given by

$$H_R = \begin{pmatrix} 0 & \Omega_2/2 & \Omega_3/2 \\ \Omega_2/2 & \omega_3 - \omega_L - i\Gamma_{22} & -i\Gamma_{23} \\ \Omega_3/2 & -i\Gamma_{32} & \omega_2 - \omega_L - i\Gamma_{33} \end{pmatrix} \tag{5}$$

where

$$\Gamma_{ij} = e^2\, \mathbf{D}_{i1} \cdot \mathbf{D}_{j1}\, \omega_j^3 \,/ 6\pi\epsilon_0 \hbar c^3 \tag{6}$$

so that, in particular,

$$\Gamma_{22} = \tfrac{1}{2} A_2, \quad \Gamma_{33} = \tfrac{1}{2} A_3, \tag{7}$$

where the A_i's are the Einstein coefficients.

Without the laser the state $| 0_{\text{ph}}\rangle \,| 2\rangle$ is not an eigenstate of H in Eq. (1), due to the interaction with the radiation field, and hence the total energy is not sharp. The time development therefore may mix $| 2\rangle$ and $| 3\rangle$

by emission and subsequent absorption of virtual photons; this is easily checked in second-order perturbation theory. This leads to the generalized damping terms Γ_{ij} whose explicit form is well known (cf. Refs. [7, 12, 13, 16]). These terms also appear in Ref. [9].

We note that the off-diagonal terms $-i\Gamma_{23}$ and $-i\Gamma_{32}$ which will be crucial for the following were absent in the corresponding expressions of Cohen-Tannoudji and Dalibard [3] for two widely separated upper levels and two lasers [18] .

We denote by λ_i the eigenvalues of H_R and by $|\,\mathbf{u}_i\rangle$ and $|\,\mathbf{v}_i\rangle$ the left and right eigenvectors, normalized to

$$\langle \mathbf{u}_i \mid \mathbf{v}_j\rangle = \delta_{ij} \ . \tag{8}$$

Then

$$\exp(-iH_Rt) = \sum_i e^{-i\lambda_i t} \ |\,\mathbf{v}_i\rangle\langle \mathbf{u}_i\,| \tag{9}$$

and

$$P_0(t) = \left\| \sum_i e^{-i\lambda_i t} \ \langle \mathbf{u}_i \mid 1\rangle \mid \mathbf{v}_i\rangle \right\|^2 . \tag{10}$$

If one of the λ_i's, λ_1 say, has an imaginary part which is very small compared to the other ones this may lead to an extended dark phase if the corresponding coefficient $|\langle \mathbf{u}_1 \mid 1\rangle|^2$ is nonzero. This is so because then there is a nonzero probability to reach the region where $P_0(t)$ changes very little in time. Then the probability density for the emission of a photon at time t, which is $-P_0'(t)$, is very small.

It may happen, however, that the coefficient $|\langle \mathbf{u}_1 \mid 1\rangle|^2$ is not very small. If it were $\frac{1}{10}$, say, this would mean on the average the emission of only 10 photons before reaching an extended dark period. Thus the light period would be very short, too short to be measured experimentally. If, on the other hand, the coefficient $|\langle \mathbf{u}_1 \mid 1\rangle|^2$ were extremely small one would have very many emissions before the occurrence of a dark period, and the light periods might become so long that a dark period would never be observed in practice. It is thus not only the existence of a very small imaginary part of an eigenvalue that leads to a dark period, but also the associated coefficient $|\langle \mathbf{u}_1 \mid 1\rangle|^2$ has to be small, though not vanishingly small.

The mean duration of a dark period is easily calculated as follows [4, 13, 14]. Let T_0 be a time such that $(2\,\mathrm{Im}\lambda_{2,3})^{-1} \ll T_0 \ll (2\,\mathrm{Im}\lambda_1)^{-1}$. Then $P_0(T_0) \cong |\langle \mathbf{u}_1 \mid 1\rangle|^2$ is the probability for the occurrence of a dark period after the emission of a photon. The probability density for the emission of the first photon after $t=0$ is $w_1(t) = -P_0'\,(t)$. If the first photon is emitted later than T_0 we call it a dark period, and the average time for this is

$$\begin{aligned} T_D &= \int_{T_0}^{\infty} dt\ t\ w_1(t)/P_0(T_0) \\ &= T_0 + \int_{T_0}^{\infty} dt\ P_0(t)/P_0(T_0) \end{aligned} \tag{11}$$

by partial integration. For $t \gg T_0$ all exponentials in $P_0(t)$ can be neglected except the one with λ_1, and one obtains

$$T_D = T_0 + 1/2\ \mathrm{Im}\ \lambda_1 \approx \frac{1}{2\ \mathrm{Im}\lambda_1}. \tag{12}$$

If successive photons are less than T_0 apart we speak of a light period. If $\bar{\tau}$ is the mean time between such emissions, then the mean duration of a light period is

$$T_L = \bar{\tau}/P_0(T_0) \tag{13}$$

since $1/P_0(T_0)$ is the mean number of such consecutive emissions. A precise expression is

$$\bar{\tau} = \int_0^{T_0} dt\ t\ w_1(t)\ /\ [1 - P_0(T_0)], \tag{14}$$

which will be evaluated in the next section.

III. MACROSCOPIC DARK PERIODS

In applications there is no need to determine the left and right eigenvectors of H_R. Instead of Eq. (9) one can use, with $\lambda_{j+3} \equiv \lambda_j$,

$$\exp\{-iH_Rt\} = \sum_{i=1}^{3} \frac{(H_R - \lambda_{i+1})(H_R - \lambda_{i+2})}{(\lambda_i - \lambda_{i+1})(\lambda_i - \lambda_{i+2})}\ e^{-i\lambda_i t}. \tag{15}$$

Equation (15) is verified by applying it to the eigenvectors of H_R. Hence a knowledge of the eigenvalues λ_i of H_R is sufficient. These can be determined in closed form by Cardano's formula, but it is more instructive to use approximate expressions.

We now make the assumption

$$\mathbf{D}_{21} \parallel \mathbf{D}_{31} \ . \tag{16}$$

From Eqs. (2), (3), (6), and (16) one then obtains

$$g_{2\mathbf{k}\lambda}/g_{3\mathbf{k}\lambda} = \Omega_2/\Omega_3, \tag{17}$$

$$\Gamma_{23}\ \Gamma_{32} = \Gamma_{22}\ \Gamma_{33}, \tag{18}$$

$$\Omega_2^2/\Gamma_{22} = \Omega_3^2/\Gamma_{33}. \tag{19}$$

We introduce a weighted mean frequency ω_M by

$$\omega_M = \frac{\Gamma_{33}\ \omega_2 + \Gamma_{22}\ \omega_3}{\Gamma_{22} + \Gamma_{33}} = \frac{\Omega_3^2\ \omega_2 + \Omega_2^2\ \omega_3}{\Omega_2^2 + \Omega_3^2}. \tag{20}$$

Note that for $\Gamma_{22} = \Gamma_{33}$ one has $\omega_M = (\omega_2 + \omega_3)/2$. We define the detuning Δ and the mean Rabi frequency Ω by

$$\Delta = \omega_L - \omega_M, \tag{21}$$

$$2\Omega^2 = \Omega_2^2 + \Omega_3^2. \tag{22}$$

We now make the additional assumptions

$$\delta\omega \equiv \omega_3 - \omega_2 \ll \Omega_i,\ \Gamma_{ii}, \tag{23}$$

$$|\,\Delta\,|\,\delta\omega \ll \Omega_2\Omega_3\ ,\ \Gamma_{22}\Gamma_{33}\ . \tag{24}$$

The root λ_1 of the characteristic polynomial for H_R with smallest imaginary part can now be obtained by Newton's method by starting from $\lambda_1^0 = -\Delta$, which is a good zeroth approximation for small $\delta\omega$. One finds

$$\lambda_1 \cong -\Delta + \frac{\Delta^2\,\delta\omega^2\,\Gamma_{22}\Gamma_{33}/(\Gamma_{22}+\Gamma_{33})^2}{\Omega^2/2 + i\,\Delta(\Gamma_{22}+\Gamma_{33})}. \tag{25}$$

This gives

$$\mathrm{Im}\,\lambda_1 \cong \frac{\Delta^2\,\delta\omega^2\,\Gamma_{22}\Gamma_{33}/(\Gamma_{22}+\Gamma_{33})}{\Omega^4/4 + \Delta^2(\Gamma_{22}+\Gamma_{33})^2}. \tag{26}$$

The imaginary parts of λ_2 and λ_3 are of the order of Γ_{22} and Γ_{33}. They are thus much larger than Im λ_1, and one has two widely different time scales in $P_0(t)$, which is a prerequisite for dark periods. From Eq. (12) we get for the mean duration of a dark period

$$T_D = \frac{\Omega^4/4 + \Delta^2(\Gamma_{22}+\Gamma_{33})^2}{2\,\Delta^2\,\delta\omega^2\,(\Gamma_{22}+\Gamma_{33})}. \tag{27}$$

As explained in Sec. II, the coefficient of $\exp\{-2\,\mathrm{Im}\lambda_1\,t\}$ in $P_0(t)$ gives the inverse of the mean number of emissions in a light period. Using Eqs. (15) and (25) as well as

$$\lambda_2\,\lambda_3 = \det H_R/\lambda_1, \tag{28}$$

$$\lambda_2 + \lambda_3 = \mathrm{Tr}\,H_R - \lambda_1, \tag{29}$$

one obtains after some algebra for this coefficient

$$p \equiv |\langle \mathbf{u}_1 \mid 1\rangle|^2 = \frac{\delta\omega^2\,\Omega_2^2\,\Omega_3^2/2\,\Omega^2}{\Omega^4 + 4\,\Delta^2(\Gamma_{22}+\Gamma_{33})^2}. \tag{30}$$

The coefficient $p \cong P_0(T_0)$ must be sufficiently small for having enough consecutive emissions, and thus observable light periods. One can estimate their length by Eq. (13). To evaluate the exact expression Eq. (14) for $\bar{\tau}$, the mean time between two photons in a light period, one has to determine λ_2 and λ_3. A lengthy calculation gives

$$\bar{\tau} \cong \frac{1}{\Gamma_{22}+\Gamma_{33}}\;\frac{\Omega^2 + (\Gamma_{22}+\Gamma_{33})^2 + \Delta^2}{\Omega^2} \tag{31}$$

and from this one determines $T_L = \bar{\tau}/p$. As a typical case we now discuss

$$\Omega_i = 5\,\Gamma\,,\quad \Gamma_{ij} \equiv \Gamma\,. \tag{32}$$

Then one obtains

$$T_D = \frac{625/4}{(\delta\omega/\Gamma)^2(\Delta/\Gamma)^2}\,\Gamma^{-1} + \frac{4}{(\delta\omega/\Gamma)^2}\,\Gamma^{-1}, \tag{33}$$

$$p = \frac{50(\delta\omega/\Gamma)^2}{2500 + 64(\Delta/\Gamma)^2}, \tag{34}$$

$$p \cong \frac{1}{50}(\delta\omega/\Gamma)^2 \qquad \text{for } \Delta \leq \Gamma, \tag{35}$$

$$\bar{\tau} = [58 + 2(\Delta/\Gamma)^2]\,10^{-2}\,\Gamma^{-1}. \tag{36}$$

For $\Delta \leq \Gamma$ one sees that p, $\bar{\tau}$ and thus also $T_L = \bar{\tau}/p$ are relatively insensitive to the detuning Δ. Since the number of photons in a light period is given by $1/p$ it follows from Eq. (35) that the level splitting $\delta\omega$ has to be much smaller than Γ for this number to be observable.

On the other hand, the dark period can be made very long by choosing the detuning Δ small. For $\bar{\nu}$, the overall mean photon rate including the dark periods, one has [19]

$$\begin{aligned}\bar{\nu} &= p^{-1}\,/\,(T_L + T_D)\\ &= 1/(\bar{\tau} + p\,T_D)\,.\end{aligned} \tag{37}$$

Insertion of Eqs. (30) and (28) yields

$$\begin{aligned}I &\sim \bar{\nu}\\ &= 4\Delta^2\frac{\Gamma_{22}+\Gamma_{33}}{\Omega^2 + 4\Delta^2[1 + \{(\Gamma_{22}+\Gamma_{33})^2 + \Delta^2\}/\Omega^2]}.\end{aligned} \tag{38}$$

This expression is valid for $\delta\omega \ll \Omega_i$, Γ. For $\Delta^2 \ll \Omega_2^2 + \Omega_3^2$ it can be approximated by

$$\bar{\nu} \cong 4\,\Delta^2(\Gamma_{22}+\Gamma_{33})/\Omega^2\,. \tag{39}$$

One sees that the fluorescence rate *drops* with increasing laser intensity. This is due to the faster growth of T_D compared to T_L with increasing Rabi frequency. For Δ approaching Ω, however, the intensity increases with Ω. This is displayed in Fig. 2. In Ref. [7] one has $\delta\omega \gg \Gamma$, and there $\bar{\nu}$ increases with the laser intensity.

For the values

$$\Omega_i = 5\,\Gamma\,,\quad \Gamma = 10^8\ \mathrm{s}^{-1}, \tag{40}$$

$$\delta\omega = 10^6\ \mathrm{s}^{-1}\,,\quad \Delta = 2\cdot 10^7\ \mathrm{s}^{-1}, \tag{41}$$

one obtains

$$T_D = 390.6\ \ \mathrm{ms}, \tag{42}$$

$$T_L = 2.9\ \ \mathrm{ms}, \tag{43}$$

and for the number of photons in a light period

$$p^{-1} = 5 \times 10^5\,. \tag{44}$$

In Fig. 3 we have plotted T_L and T_D over the detuning Δ. The dotted curve shows T_D for parallel transition dipole moments.

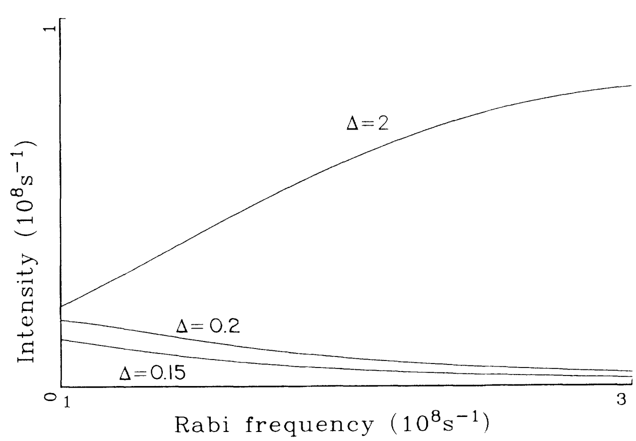

FIG. 2. Fluorescence intensity vs Rabi frequency for three different detunings Δ, with $\Omega_i = \Gamma_{ij} = 10^8\ \mathrm{s}^{-1}$ and $\delta\omega = 0.01\Gamma_{ij}$. For $\Delta = 0.15\Gamma_{ij}$ and $\Delta = 0.2\Gamma_{ij}$ the fluorescence intensity drops with increasing Rabi frequency, while it grows for $\Delta = 2\,\Gamma_{ij}$.

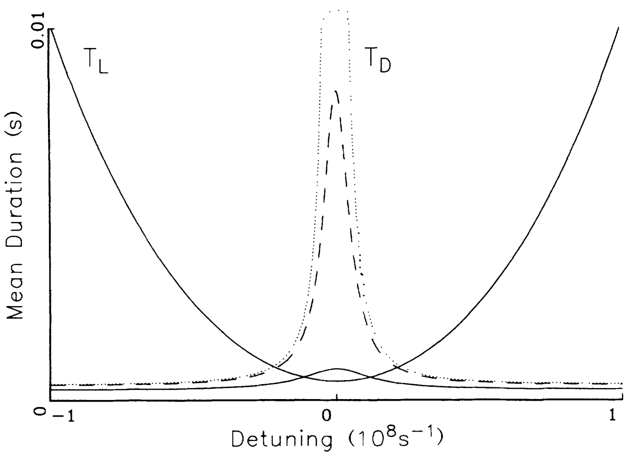

FIG. 3. Mean duration T_D and T_L of dark and light periods. T_D is plotted for three different angles ϕ between the transition dipole moments (dotted line: $\phi = 0$, dashed line: $\phi = 6 \times 10^{-7}$ rad, solid line: $\phi = 6 \times 10^{-6}$ rad). T_L does not depend appreciably on ϕ.

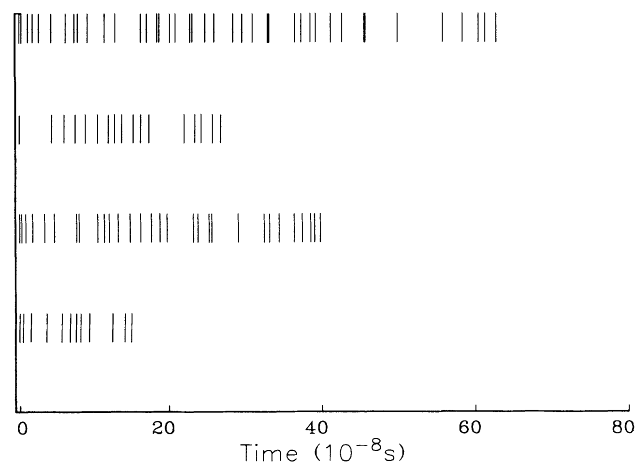

FIG. 4. Simulation of atomic emission behavior for $\Delta = 0$, $\Omega_i = \Gamma_{ij} = 10^8$ s^{-1} and $\delta\omega = 0.3\Gamma_{ij}$. After a finite number of emissions the atom stops to fluoresce. Here the mean number of photons emitted is 23.2. Four runs are plotted, with each vertical line representing a photon emission.

Switching the atom off. From Eq. (30) it appears that for $\Delta \to 0$ the dark period becomes infinitely long. Indeed, for $\Delta = 0$ and $\mathbf{D}_{31} \parallel \mathbf{D}_{21}$, the reduced Hamiltonian H_R in Eq. (5) has an eigenvalue $\lambda_1 = 0$. The corresponding left and right eigenvectors in the atomic space coincide and are given by

$$|\, \mathbf{u}_1\rangle = |\, \mathbf{v}_1\rangle = \left\{(\Omega_2\, \Omega_3/\Omega^2)\,\delta\omega\, |\, 1\rangle + \Omega_3\, |\, 2\rangle - \Omega_2\, |\, 3\rangle\right\}/\|\cdots\|. \tag{45}$$

This state is constant under the reduced atomic time development given by H_R in the interaction picture [20]. The ground state $|\, 1\rangle$ has a nonvanishing $|\, \mathbf{u}_1\rangle$ component. Since H_R has an eigenvalue 0 for $\Delta = 0$ one sees that

$$e^{-iH_Rt/\hbar}\, |\, 1\rangle = \alpha_1\, |\, \mathbf{u}_1\rangle + \sum_2^3 \alpha_i e^{-i\lambda_i t}\, |\, \mathbf{v}_i\rangle$$

converges to $\alpha_1 \,|\, \mathbf{u}_1\rangle$ for large t. Hence $P_0(t)$ tends to a nonzero constant for $t \to \infty$, which is $|\langle \mathbf{u}_1 \mid 1\rangle|^2$ and gives the probability that after an emission no photon is emitted for all times. Thus

$$|\langle \mathbf{u}_1 \mid 1\rangle|^{-2} = (\delta\omega)^{-2}\, \Omega^2(\Gamma_{22} + \Gamma_{33})^2/2\, \Gamma_{22}\, \Gamma_{33} \tag{46}$$

is the average number of photons before this constant nonradiating state is reached. This means that after the emission of a number of photons the fluorescence is switched off completely. Figure 4 shows a simulation of this phenomenon. If the laser is switched off this state is no longer stable, and the atom will make a transition to the ground state. In the next section we discuss this in terms of the complete Hamiltonian, including the radiation field. This will allow an intuitive understanding of the appearance of dark periods.

IV. AN EXACT NONRADIATING SOLUTION OF THE COMPLETE TIME DEVELOPMENT

To better understand the appearance of a nonradiating state and of dark periods we return to the complete Hamiltonian (1) which includes the radiation field. We assume parallel dipole moments as well as $\omega_L = \omega_M$ and consider in analogy to Eq. (45), at $t = 0$, the no-photon state

$$|\, \psi(0)\rangle = |\, 0_{\rm ph}\rangle \left\{(\Omega_2\Omega_3/\Omega^2)\delta\omega\, |\, 1\rangle + \sqrt{2}\,\Omega\, |\, a\rangle\right\}/\|\cdots\|. \tag{47}$$

We claim that, for any $\delta\omega$, its time development under the complete Hamiltonian is given by

$$U_H(t,0)\, |\, \psi(0)\rangle = |\, 0_{\rm ph}\rangle \left\{(\Omega_2\Omega_3/\Omega^2)\,\delta\omega\, |\, 1\rangle + \sqrt{2}\,\Omega\, e^{-i\omega_L t}\, |\, a\rangle\right\}/\|\cdots\| \tag{48}$$

so that it remains a no-photon state for all times, and the laser will create no fluorescence with $|\, \psi(0)\rangle$ as initial state even though it pumps $|\, 1\rangle$ to $|\, s\rangle$ and vice versa.

To prove this we use an interaction picture. We set

$$H_L^0 = \hbar\omega_L\{|\, 2\rangle\langle 2\,| + |\, 3\rangle\langle 3\,|\} + H_F^0 \tag{49}$$

and go over to the interaction picture with respect to H_L^0. The time development is then given by

$$H_I^L(t) \;=\; e^{iH_L^0 t/\hbar}(H - H_L^0)\, e^{-iH_L^0 t/\hbar}\,. \tag{50}$$

We now express this in terms of the normed states

$$\begin{aligned} |\, s\rangle &\equiv \{\Omega_2 \,|\, 2\rangle \;+\; \Omega_3 \,|\, 3\rangle\}/\sqrt{2}\,\Omega, \\ |\, a\rangle &\equiv \{\Omega_3 \,|\, 2\rangle \;-\; \Omega_2 \,|\, 3\rangle\}/\sqrt{2}\,\Omega, \end{aligned} \tag{51}$$

where $2\Omega^2 = \Omega_2^2 + \Omega_3^2$. For parallel dipole moments, which is assumed in the following, these states are independent of the Rabi frequencies [21], and Eq. (50) can be written as

$$\begin{aligned} \hbar^{-1} H_I^L(t) = {} & (2\,\Omega^2)^{-1}(\omega_2\Omega_2^2 + \omega_3\,\Omega_3^2)\,|\, s\rangle\langle s\,| \;+\; (\omega_M - \omega_L)\,|\, a\rangle\langle a\,| - (\Omega_2\Omega_3/2\Omega^2)\delta\omega\{|\, s\rangle\langle a\,| \;+\; |\, a\rangle\langle s\,|\} \\ & + 2^{-1/2}\,\Omega\,\{|\, s\rangle\langle 1\,| \;+ \text{H.c.}\} + i\,2^{1/2}\,\Omega\,\sum_{\mathbf{k},\lambda}(g_{2\mathbf{k}\lambda}/\Omega_2)\left\{a_{\mathbf{k},\lambda}e^{-i(\omega-\omega_L)t}\,|\, s\rangle\langle 1\,| \;-\; \text{H.c.}\right\}, \end{aligned} \tag{52}$$

where ω_M is given by Eq. (20). The first three terms arise from the purely atomic Hamiltonian minus H_L^0.

From Eq. (52) one sees that for parallel dipole moments only $|\, s\rangle$ couples to the laser and radiation field, while the purely atomic part transforms $|\, s\rangle$ into $|\, a\rangle$ and vice versa. For any t we find

$$\begin{aligned} H_I^L(t)\,|\,\psi(0)\rangle &= 2^{-1/2}\,\Omega(\Omega_2\Omega_3/\Omega^2)\delta\omega\,|\, s\rangle \\ &\quad -2^{1/2}\,\Omega(\Omega_2\Omega_3/2\Omega^2)\delta\omega\,|\, s\rangle \qquad (53)\\ &= 0, \qquad (54) \end{aligned}$$

where the first term originates from the $|\, 1\rangle$ component of $|\,\psi(0)\rangle$ which is pumped by the laser to a multiple of $|\, s\rangle$. The second term comes from the $|\, a\rangle$ component of $|\,\psi(0)\rangle$ which is transformed by the purely atomic part to a multiple of $|\, s\rangle$, too. It is crucial that for $\omega_L = \omega_M$ there is no $|\, a\rangle\langle a\,|$ part in H_I^L. The photon operators give no contribution. Since Eq. (53) holds for all t, $|\,\psi_I(t)\rangle$ is constant in the interaction picture, by the Dyson series, and this proves Eq. (48).

The existence of a stable state for $\Delta = 0$ which does not radiate although its ground-state component is constantly pumped up to the $|\, s\rangle$ state is thus due to a cancellation of the laser pumping by the action of the purely atomic part of the Hamiltonian on $|\, a\rangle$.

If the laser power is changed the system will start to fluoresce again; in particular, if the laser is switched off after some time, the previously stable nonradiating state will acquire a photon part, and the atom will eventually make a transition to the ground state under emission of a photon.

For $\omega_L \neq \omega_M$, i.e., $\Delta \neq 0$, the term $(\omega_M - \omega_L)\,|\, a\rangle\langle a\,|$ in Eq. (52) contributes and makes the right-hand side of Eq. (53) nonzero and proportional to $|\, a\rangle$. By repeated action of H_I^L in the Dyson series this will then slowly create an $|\, s\rangle$ component which may lead to the emission of a photon.

If one starts with the atomic ground state and no photon, the laser pumping will create an $|\, s\rangle$ component which may lead to the emission of a photon. But if this emission is delayed the analysis of the preceding section shows that for small $\delta\omega$ the atomic state vector rapidly approaches the eigenvector belonging to the small eigenvalue λ_1 of the reduced Hamiltonian H_R. This eigenvector is close to the one for $\lambda_1 = 0$, i.e., to the vector in the curly brackets in Eq. (47) and thus close to a stable nonradiating state. If $\Delta = 0$ then also $\lambda_1 = 0$, and the atom approaches the stable, nonradiating state. If we assume $\delta\omega \ll \Omega, \Gamma$, then the stable state has a very small ground-state component. In case of degeneracy, $\delta\omega = 0$, it is easy to see that $|\, a\rangle$ is invariant, even without the rotating-wave approximation.

V. DISCUSSION

We have shown how quantum coherence effects of two very close upper levels of a V system may give rise to macroscopic dark periods provided the dipole moments of the upper levels with respect to the ground state are parallel. This last condition leads to $\Gamma_{23}\,\Gamma_{32} = \Gamma_{22}\,\Gamma_{33}$ in Eq. (5), and only this was needed. It is quite clear from our analysis that dark periods will persist if this equality holds only approximately. In case the dipole moments are not quite parallel the mean length of a dark period in Eq. (27) is modified and is given in zeroth order in $\delta\omega$ and Δ by

$$T_D \cong \Omega^2 \,/\left[\Omega_2^2\Gamma_{33} \;+\; \Omega_3^2\Gamma_{22} \;-\; 2\Omega_2\Omega_3\Gamma_{23}\right]. \tag{55}$$

In Fig. 3 we have plotted the duration of light and dark periods for different angles between the dipole moments. The light period is insensitive to the angle, but the dark period decreases rapidly with increasing angle between the dipole moments. An angle of 0.001 rad would still lead to appreciable dark periods. The nonradiating state for $\Delta = 0$, however, then no longer exists.

In our treatment the rotating-wave approximation has been used. Without it we do not expect any great change for the light and dark periods. However, the nonradiating state of Secs. III and IV will no longer be strictly stable on a very long time scale, and instead of an infinitely long dark period one would expect very long dark periods with some photons in between.

As mentioned in the Introduction, for a Λ system there also exists a state decoupled from the electromagnetic field, and one might expect similar dark periods. It turns out that for the Λ system the additional damping terms Γ_{23} and Γ_{32} do not appear in the reduced Hamiltonian H_R, and therefore the condition of parallel dipole moments is not necessary. Light and dark periods also exist for the Λ system, but the short light periods are probably too difficult to be observed individually. An analysis of these questions for the Λ system is planned to appear elsewhere.

In this paper we have discussed dark periods of a *single* atom. If instead of a single atom one has a gas of atoms each exhibiting longer and longer dark periods as the laser frequency approaches $\Delta = 0$, then the gas will absorb and rescatter less and less laser light, i.e., one will see reduced fluorescence and a nonabsorption resonance. In this way one can understand the nonabsorption resonances found in Refs. [6, 7] by dark periods of individual atoms.

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[1] H. G. Dehmelt, Bull. Am. Phys. Soc. **20**, 60 (1975).

[2] R. J. Cook and H. J. Kimble, Phys. Rev. Lett. **54**, 1023 (1985).

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

[4] A.Schenzle, R. G. DeVoe, and R. G. Brewer, Phys. Rev. A **33**, 2127 (1986); J. Javanainen, *ibid.* **33**, 2121 (1986); D. T. Pegg, R. Loudon, and P. L. Knight, *ibid.* **33**, 4085 (1986); G. Nienhuis, *ibid.* **35**, 4639 (1987); P. Zoller, M. Marte, and D. J. Walls, *ibid.* **35**, 198 (1987); M. Porrati and S. Puttermann, *ibid.* **36**, 925 (1987); S. Reynaud, J. Dalibard, and C. Cohen-Tannoudji, IEEE J. Quantum Electron. **24**, 1395 (1988).

[5] Th. Sauter, R. Blatt, W. Neuhauser, and P. E. Toschek, Opt. Commun. **60**, 287 (1986); W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986); J. C. Bergquist, W. M. Itano, R. G. Hulet, and D. J. Wineland, Phys. Scr. **T22**, 79 (1988).

[6] G. Orriols, Nuovo Cimento B **53**, 1 (1979); E. Arimondo and G. Orriols, Lett. Nuovo Cimento **17**, 333 (1976).

[7] D. A. Cardimona, M. G. Raymer, and C. R. Stroud Jr., J. Phys. B **15**, 55 (1982).

[8] H. R. Gray, R. M. Whitley, and C. R. Stroud Jr., Opt. Lett. **3**, 218 (1978); P. M. Radmore and P. L. Knight, J. Phys. B **15**, 561 (1982); S. Swain, *ibid.* **15**, 3405 (1982); H. I. Yoo and J. H. Eberly, Phys. Rep. **118**, 239 (1985).

[9] S.E. Harris, Phys. Rev. Lett. **62**, 1033 (1989); A. Imamoglu, Phys. Rev. A **40**, 2835 (1989).

[10] J. A. Bergou, M. Orszag, and M. O. Scully, Phys. Rev. A **38**, 754 (1988); M. O. Scully, S.-Y. Zhu, and A. Gavrielides, Phys. Rev. Lett. **62**, 2813 (1989).

[11] R. H. Dicke, Phys. Rev. **93**, 99 (1954).

[12] D. A. Cardimona, Phys. Rev. A **41**, 5016 (1990); D. A. Cardimona, M. P. Sharma, and M. A. Ortega, J. Phys. B **22**, 4029 (1989).

[13] M. Porrati and S. Putterman, Phys. Rev. A **39**, 3010 (1989).

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

[15] R. Loudon, *The Quantum Theory of Light*, 2nd ed. (Clarendon, Oxford, 1983).

[16] P. W. Milonni, Phys. Rep. C **25**, 1 (1976); L. D. Landau, Z. Phys. **45**, 430 (1927).

[17] Reference [13] has $P_0(t) = \|U_r(t,0) \mid 1\rangle\|^2$ where their U_r can be shown to equal $\exp\{-i\omega_L\ [\,|\,2\rangle\langle 2\,|\ +\ |\,3\rangle\langle 3\,|\,]\ t\}$ $\exp\{-iH_R t\}$. The first factor is unitary and drops out. Also, Ref. [13] uses as interaction $\mathbf{P}\cdot\mathbf{A}$ instead of the dipole form $\mathbf{D}\cdot\mathbf{E}$ as in (2). In Eq. (6) for Γ_{ij} one then has to replace ω_j^3 by $\omega_i\omega_j^2$. But this is of no consequence since for small level separation the expressions agree and for larger level separation the Γ_{ij} terms can be neglected, as explained below.

[18] These additional damping terms may indeed be omitted if the transition frequencies have a wide separation and if one drives the atom with two lasers. Then, in the interaction picture, they take the form $-i\Gamma_{23}\ \exp i(\omega_{L1}-\omega_{L2})t$ and do not contribute due to their rapid oscillations.

[19] This holds if one has pronounced light and dark periods, i.e., distinct time scales in $P_0(t)$ and $P_0(T_0) \ll 1$. An exact expresssion is $\bar{\nu}^{-1} = \int_0^\infty dt\, P_0(t)$.

[20] It is interesting to note that this state exhibits population inversion for small $\delta\omega$, with no additional microwave radiation between $|\,2\rangle$ and $|\,2\rangle$ needed, in contrast to a population inversion found by Scully *et al.* and Bergou *et al.* in Ref. [8]. In addition, this state has another noteworthy property. If one writes it in the form of a density matrix, $|\,\mathbf{u}_1\rangle\langle\mathbf{u}_1\,|$, then the density matrix is a stationary solution of the Bloch equations for the V system in the frame rotating with the field.

[21] Indeed, then one has from (22)

$$|\,s\rangle = \{\Gamma_{22}^{1/2}\ |\,2\rangle + \Gamma_{33}^{1/2}\ |\,3\rangle\}/\{\Gamma_{22}+\Gamma_{33}\}^{1/2}$$

and similarly for $|\,a\rangle$.