Cooperative fluorescence effects for dipole-dipole interacting systems with experimentally relevant level configurations

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The mutual dipole-dipole interaction of atoms in a trap can affect their fluorescence. Extremely large effects were reported for double jumps between different intensity periods in experiments with two and three Ba^+ ions for distances in the range of about ten wave lengths of the strong transition while no effects were observed for Hg^+ at 15 wavelengths. In this theoretical paper we study this question for configurations with three and four levels, which model those of Hg^+ and Ba^+, respectively. For two systems in the Hg^+ configuration we find cooperative effects of up to 30% for distances around one or two wavelengths, about 5% around ten wavelengths, and, for larger distances in agreement with experiments, practically none. This is similar for two *V* systems. However, for two four-level configurations, which model two Ba^+ ions, cooperative effects are practically absent, and this latter result is at odds with the experimental findings for Ba^+.

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I. INTRODUCTION

The dipole-dipole interaction is ubiquitous in physics and, for example, responsible for the ever present van der Waals force. It is also important for envisaged quantum computers based on atoms or ions in traps. Considerable interest in the literature has also been aroused by its cooperative effects on the radiative behavior of atoms [1]. In an as yet unexplained experiment [3,4] with two and three Ba^+ ions, which exhibit macroscopic light and dark periods, a large fraction of double and triple jumps was reported, i.e., jumps by two or three intensity steps within a short resolution time. This fraction was orders of magnitudes larger than for independent ions. The quantitative explanation of such a large cooperative effect for distances of the order of ten wavelengths of the strong transition has been found difficult [5–10]. Experiments with other ions showed no observable cooperative effects [11,12], in particular, none were seen for Hg^+ for a distance of about 15 wavelengths [13]. More recently, an unexpected high number of simultaneous quantum jumps in a linear chain of trapped Ca^+ ions were reported [14], while no such effects were found in another experiment [15] using the same ion species and a similar setup.

Systems with macroscopic light and dark periods can provide a sensitive test for cooperative effects of the dipole-dipole interaction. These periods can occur for multilevel systems where the electron is essentially shelved for some time in a meta-stable state without photon emission [16]. For two *V* systems with macroscopic light and dark periods the effect of the dipole-dipole interaction was investigated numerically in Ref. [17] and analytically in Ref. [2] and shown to be up to 30% in the double-jump rate compared to independent systems. Monitoring the dipole-dipole interaction of two *V* systems via quantum jumps of individual atoms was investigated in Ref. [18]. The experimental systems of Refs. [3,13,19] are, however, not in the *V* configuration so that the results of Ref. [17] do not directly apply.

The experiment of Ref. [13] used two Hg^+ ions, with the relevant three levels as in Fig. 1, which we call a *D* configuration. From a theoretical point of view two such systems were studied in Ref. [20] for the special case $\lambda_1, \lambda_3 \ll r \ll \lambda_2$, where r is the distance and the wavelengths refer to the respective transitions of Fig. 1, and for this case no cooperative effects were found. The general case will be treated explicitly further below. The cooperative effects found here are of similar magnitude as for *V* systems, and for distances of the above range the result of Ref. [20] is confirmed. Our results are also in agreement with the experimentally observed absence of cooperative effects for distances of about 15 wavelengths [13].

The levels of Ba^+ used in the experiment of Refs. [3,19] are depicted in Fig. 2(a). The ground state $6^2S_{1/2}$ and the two upper states $6^2P_{1/2}$ and $5^2D_{5/2}$ constitute a strongly driven fluorescing Λ system, which provides the light periods. Only when the system is in the ground state can the weak incoherent driving of the $6\,^2S_{1/2}$-$6^2P_{3/2}$ transition populate the metastable $5^2D_{5/2}$ state, with ensuing dark period. Therefore the details of the two upper states of the Λ system play no significant role for the transition to a dark period, and therefore these two states are replaced here by an *effective* single level. This leads to the four-level configuration of Fig. 2(b). The present paper is, to our knowledge, the first to theoretically investigate possible cooperative effects for two such four-level systems. Surprisingly, these effects turn out to be much smaller than for two *V* systems for distances $r \gg \lambda_3$, and this shows that cooperative effects sensitively depend on how the metastable level is populated. Our results for two four-level

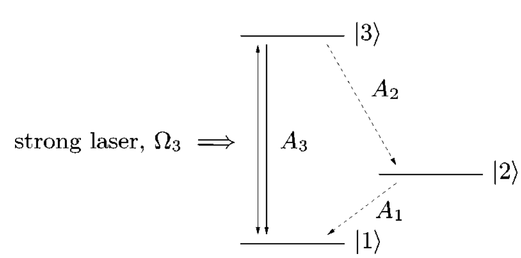

FIG. 1. Three-level system in *D* configuration with fast transitions (solid lines) and slow transitions (dashed lines).

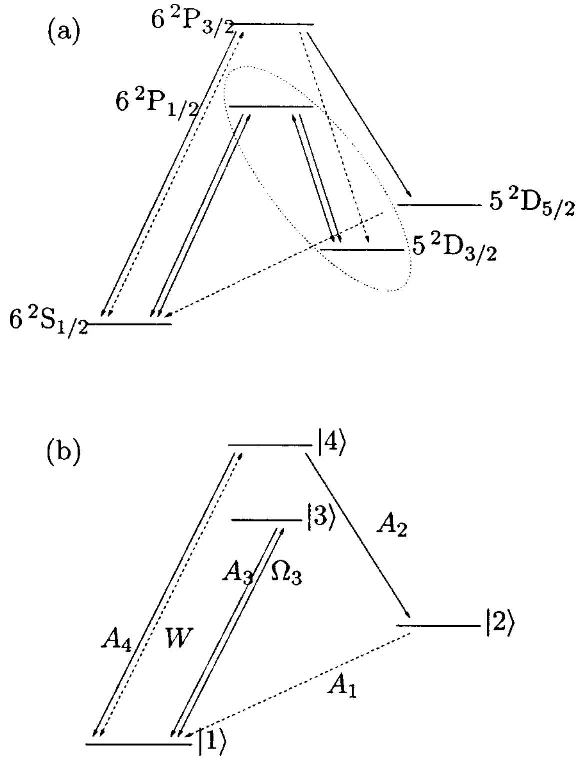

FIG. 2. (a) Relevant level scheme of Ba^+ [3,4]. For circled levels see text. (b) Effective four-level system for Ba^+. Strong coherent driving of the $|1\rangle$–$|3\rangle$ transition by a laser, weak incoherent driving of the $|1\rangle$–$|4\rangle$ transition by a lamp, weak decay of level $|2\rangle$.

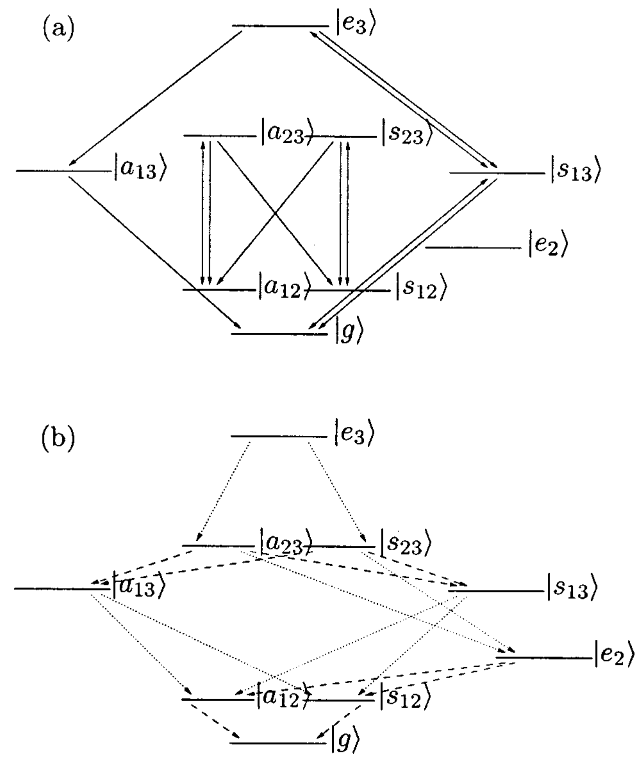

FIG. 3. Level configuration of two D systems in the Dicke basis. (a) Slow transitions omitted. (b) Transitions with rate $A_2 \pm \mathrm{Re}\, C_2$ (dotted arrows) and transitions with rate $A_1 \pm \mathrm{Re}\, C_1$ (dashed arrows). Line shifts due to detuning and to $\mathrm{Im}\, C_i$ are omitted.

configurations are at odds with the experimental findings of Ref. [3] on the magnitude of double jump rates for two Ba^+ ions [21].

The methods presented in this paper can be carried over to describe the Ca^+ experiments of Refs. [14,15] although this would of course require the use of a different level system.

The plan of the paper is as follows. In Sec. II we treat two dipole-dipole interacting D systems and explicitly calculate the transition rates between the various light and dark periods as well as the double-jump rate. This is done by means of Bloch equations. In Sec. III the method is carried over to two four-level systems of Fig. 2(b) and the transition rates are calculated. In the Appendix a direct quantum jump approach [22] is outlined for two D systems.

II. TWO DIPOLE-INTERACTING *D* SYSTEMS

The D configuration, as displayed in Fig. 1, is a model of the level system of Hg^+ used in the experiments of Refs. [13,23]. The transition $|1\rangle$–$|3\rangle$ is driven by a strong laser. Level $|3\rangle$ can also decay via a slow transition to the metastable level $|2\rangle$. For simplicity all transitions are treated as dipole transitions.

In the following we will investigate two dipole-interacting D systems a fixed distance r apart and calculate the transition rates between the three types of fluorescence periods. This will be done in this section by means of the Bloch equations. In the Appendix the efficient quantum jump approach will be applied to two D systems, not only confirming the Bloch equation result but also giving higher-order terms. For simplicity the laser direction will be taken as perpendicular to the line joining the two systems. Rabi frequency and Einstein coefficients satisfy

$$A_3, \Omega_3 \gg A_1, A_2. \tag{1}$$

It is convenient to use a Dicke basis,

$$\begin{aligned} |g\rangle &= |1\rangle|1\rangle, \quad |e_2\rangle = |2\rangle|2\rangle, \quad |e_3\rangle = |3\rangle|3\rangle, \\ |s_{ij}\rangle &= (|i\rangle|j\rangle + |j\rangle|i\rangle)/\sqrt{2}, \\ |a_{ij}\rangle &= (|i\rangle|j\rangle - |j\rangle|i\rangle)/\sqrt{2}\mathrm{i}. \end{aligned} \tag{2}$$

In Fig. 3(a) the level configuration for two D systems is displayed in this basis, with the slow decays neglected, while in Fig. 3(b) only the slow decays are shown. From Fig. 3(a) it is seen that without slow decays (i.e., for $A_1 = A_2 = 0$) the configuration decouples into three independent subspaces, denoted by $\mathcal{S}_0, \mathcal{S}_1, \mathcal{S}_2$, with

$$\begin{aligned} \mathcal{S}_0 &= \{|e_2\rangle\}, \\ \mathcal{S}_1 &= \{|s_{12}\rangle, |a_{12}\rangle, |s_{23}\rangle, |a_{23}\rangle\}, \\ \mathcal{S}_2 &= \{|g\rangle, |s_{13}\rangle, |a_{13}\rangle, |e_3\rangle\}. \end{aligned} \tag{3}$$

By means of the conditional Hamiltonian $H_{\rm cond}$ and the reset operation $\mathcal{R}$ the Bloch equations can be written in the form [24]

$$\dot{\rho}=-\frac{\mathrm{i}}{\hbar}[H_{\rm cond}\rho-\rho H_{\rm cond}^{\dagger}]+\mathcal{R}(\rho). \tag{4}$$

For two D systems one finds by the same method as in Refs. [2,25],

$$\begin{aligned}H_{\rm cond}=&\frac{\hbar}{2\mathrm{i}}\Bigg\{A_1\Bigg[2|e_2\rangle\langle e_2|+\sum_{j=1}^{2}|s_{jj+1}\rangle\langle s_{jj+1}|\\&+|a_{jj+1}\rangle\langle a_{jj+1}|\Bigg]+(A_2+A_3+2\mathrm{i}\Delta_3)\Bigg[2|e_3\rangle\langle e_3|\\&+\sum_{j=1}^{2}|s_{j3}\rangle\langle s_{j3}|+|a_{j3}\rangle\langle a_{j3}|\Bigg]+\sum_{j=1}^{2}C_j[|s_{jj+1}\rangle\langle s_{jj+1}|\\&-|a_{jj+1}\rangle\langle a_{jj+1}|]+C_3[|s_{13}\rangle\langle s_{13}|-|a_{13}\rangle\langle a_{13}|]\Bigg\}\\&+\frac{\hbar}{2}\Omega_3\{\sqrt{2}(|g\rangle\langle s_{13}|+|s_{13}\rangle\langle e_3|)+|s_{12}\rangle\langle s_{23}|-|a_{12}\rangle\\&\times\langle a_{23}|+\mathrm{H.c.}\},\end{aligned} \tag{5}$$

where Δ_3 is the detuning of the laser. The complex dipole coupling constants C_j depend in an oscillatory way on the distance r,

$$\begin{aligned}C_j=&\frac{3A_j}{2}\mathrm{e}^{\mathrm{i}k_jr}\Bigg[\frac{1}{\mathrm{i}k_jr}(1-\cos^2\theta_j)\\&+\left(\frac{1}{(k_jr)^2}-\frac{1}{\mathrm{i}(k_jr)^3}\right)(1-3\cos^2\theta_j)\Bigg],\end{aligned} \tag{6}$$

where $k_j=2\pi/\lambda_j$ and θ_j is the angle between the corresponding dipole moment and the line connecting the systems. For maximal effect we take $\theta_j=\pi/2$ in the following. The real part of C_j leads to changes of the decay constants and the imaginary part to a level shift in the Dicke basis, as seen from the expression for $H_{\rm cond}$. The reset operation can be written in the form

$$\begin{aligned}\mathcal{R}(\rho)=\sum_{j=1}^{3}&[(A_j+\mathrm{Re}\,C_j)R_+^{(j)}\rho R_+^{(j)\dagger}\\&+(A_j-\mathrm{Re}C_j)R_-^{(j)}\rho R_-^{(j)\dagger}],\end{aligned} \tag{7}$$

where

$$R_+^{(1)}=|g\rangle\langle s_{12}|+|s_{12}\rangle\langle e_2|+\frac{1}{\sqrt{2}}(|s_{13}\rangle\langle s_{23}|+|a_{13}\rangle\langle a_{23}|),$$

$$R_-^{(1)}=|g\rangle\langle a_{12}|+|a_{12}\rangle\langle e_2|+\frac{1}{\sqrt{2}}(|a_{13}\rangle\langle s_{23}|-|s_{13}\rangle\langle a_{23}|),$$

$$R_+^{(2)}=|e_2\rangle\langle s_{23}|+|s_{23}\rangle\langle e_3|+\frac{1}{\sqrt{2}}(|s_{12}\rangle\langle s_{13}|+|a_{12}\rangle\langle a_{13}|),$$

$$R_-^{(2)}=|e_2\rangle\langle a_{23}|+|a_{23}\rangle\langle e_3|+\frac{1}{\sqrt{2}}(|s_{12}\rangle\langle a_{13}|-|a_{12}\rangle\langle s_{13}|),$$

$$R_+^{(3)}=|g\rangle\langle s_{13}|+|s_{13}\rangle\langle e_3|+\frac{1}{\sqrt{2}}(|s_{12}\rangle\langle s_{23}|-|a_{12}\rangle\langle a_{23}|),$$

$$R_-^{(3)}=|g\rangle\langle a_{13}|+|a_{13}\rangle\langle e_3|+\frac{1}{\sqrt{2}}(|s_{12}\rangle\langle a_{23}|+|a_{12}\rangle\langle s_{23}|).$$

To determine the transition rates we write Eq. (4) in a Liouvillean form as

$$\dot{\rho}=\mathcal{L}\rho=\{\mathcal{L}_0(A_3,C_3,\Omega_3,\Delta_3)+\mathcal{L}_1(A_1,A_2,C_1,C_2)\}\rho, \tag{8}$$

where the superoperator $\mathcal{L}_1(A_1,A_2,C_1,C_2)$ is a perturbation depending on the small parameters, and we employ the following important property of the time development. Starting with an initial state in one of the subspaces $\mathcal{S}_i$, the system will rapidly—on a time scale of Ω_3^{-1} and A_3^{-1}—approach one of the quasistationary states $\rho_{{\rm ss},i}$. Thereafter—for times much larger than Ω_3^{-1} and A_3^{-1}, but much smaller than A_1^{-1} and A_2^{-1}—small populations in the other subspaces will build up until eventually, on a time scale of A_1^{-1} and A_2^{-1}, the true stationary state is approached. Hence we consider a time Δt with

$$A_3^{-1},\Omega_3^{-1}\ll\Delta t\ll A_1^{-1},A_2^{-1} \tag{9}$$

and calculate $\rho(t_0+\Delta t)$ for initial $\rho(t_0)=\rho_{{\rm ss},i}$. The quasi-invariant states are easily calculated from $\mathcal{L}_0\rho_{{\rm ss},i}=0$ as

$$\rho_{\rm ss,0}=|e_2\rangle\langle e_2|, \tag{10}$$

$$\begin{aligned}\rho_{\rm ss,1}=&\frac{1}{2}\frac{A_3^2+\Omega_3^2+4\Delta_3^2}{A_3^2+2\Omega_3^2+4\Delta_3^2}(|s_{12}\rangle\langle s_{12}|+|a_{12}\rangle\langle a_{12}|)\\&+\frac{1}{2}\frac{\Omega_3^2}{A_3^2+2\Omega_3^2+4\Delta_3^2}(|s_{23}\rangle\langle s_{23}|+|a_{23}\rangle\langle a_{23}|)\\&+\Bigg\{\frac{1}{2}\frac{(\mathrm{i}A_3-2\Delta_3)\Omega_3}{A_3^2+2\Omega_3^2+4\Delta_3^2}(|s_{12}\rangle\langle s_{23}|-|a_{12}\rangle\langle a_{23}|)\\&+\mathrm{H.c.}\Bigg\},\end{aligned} \tag{11}$$

$$\begin{aligned}\rho_{\rm ss,2}=&\frac{1}{N}[\{N-\Omega_3^2(2A_3^2+3\Omega_3^2+8\Delta_3^2)\}|g\rangle\langle g|\\&+\Omega_3^2(2A_3^2+\Omega_3^2+8\Delta_3^2)|s_{13}\rangle\langle s_{13}|\\&+\Omega_3^4\{|e_3\rangle\langle e_3|+|a_{13}\rangle\langle a_{13}|\}\end{aligned}$$

$$+\{\Omega_3(\mathrm{i}A_3-2\Delta_3)(\sqrt{2}(A_3^2+\Omega_3^2+4\Delta_3^2+(A_3-2\mathrm{i}\Delta_3)C_3)|g\rangle\langle s_{13}|+\Omega_3(\mathrm{i}A_3-2\Delta_3+\mathrm{i}C_3)\times|g\rangle\langle e_3|+\sqrt{2}\Omega_3^2|s_{13}\rangle\langle e_3|)+\mathrm{H.c.}\}], \tag{12}$$

where

$$N=(A_3^2+2\Omega_3^2+4\Delta_3^2)^2+(A_3^2+4\Delta_3^2)(|C_3|^2+2A_3\,\mathrm{Re}\,C_3+4\Delta_3\,\mathrm{Im}\,C_3).$$

As in Ref. [2] one has in perturbation theory

$$\rho(t_0+\Delta t)=\rho_{\mathrm{ss},i}+\int_0^{\Delta t}\mathrm{d}\tau e^{\mathcal{L}_0\tau}\mathcal{L}_1\rho_{\mathrm{ss},i}+O((A_1,A_2,C_1,C_2)^2), \tag{13}$$

but, unlike Ref. [2], $\mathcal{L}_1\rho_{\mathrm{ss},i}$ is not a superposition of just the eigenstates for nonzero eigenvalues of $\mathcal{L}_0$ but also of the $\rho_{\mathrm{ss},j}$'s. We therefore decompose $\mathcal{L}_1\rho_{\mathrm{ss},i}$ into a superposition of all eigenstates (matrices) of $\mathcal{L}_0$,

$$\mathcal{L}_1\rho_{\mathrm{ss},i}=\sum_{j=0}^{2}\alpha_{ij}\rho_{\mathrm{ss},j}+\tilde{\rho}, \tag{14}$$

where $\tilde{\rho}$ contains the contributions from the eigenstates for nonzero eigenvalues of $\mathcal{L}_0$. For later use we note that these eigenvalues are of the order of A_3 and Ω_3. The coefficients α_{ij} can easily be determined by means of the reciprocal (or dual) eigenstates, where only those for eigenvalue 0 of $\mathcal{L}_0$ are needed. They are denoted by ρ_{ss}^i and are defined through

$$\mathrm{Tr}(\rho_{\mathrm{ss}}^{i\dagger}\rho_{\mathrm{ss},j})=\delta_{ij},\quad i,j=0,1,2, \tag{15}$$

$$\mathrm{Tr}(\rho_{\mathrm{ss}}^{i\dagger}\mathcal{L}_0A)=0\quad\text{for any matrix } A. \tag{16}$$

The latter means $\mathcal{L}_0^\dagger\rho_{\mathrm{ss}}^i=0$, with the adjoint $\mathcal{L}_0^\dagger$ defined with respect to a scalar product given by $\mathrm{Tr}(A^\dagger B)$. Then one has

$$\alpha_{ij}=\mathrm{Tr}(\rho_{\mathrm{ss}}^{j\dagger}\mathcal{L}_1\rho_{\mathrm{ss},i}). \tag{17}$$

The reciprocals ρ_{ss}^i are easily determined as follows. Since the Bloch equations conserve the trace one has

$$0=\mathrm{Tr}\,\dot{\rho}=\mathrm{Tr}\,\mathcal{L}_0\rho \tag{18}$$

for any ρ. Thus

$$0=\mathrm{Tr}(\mathbb{1}\mathcal{L}_0\rho)=\mathrm{Tr}((\mathcal{L}_0^\dagger\mathbb{1})\rho) \tag{19}$$

for any ρ and therefore $\mathcal{L}_0^\dagger\mathbb{1}=0$. Now $\mathbb{1}$ can be written as a sum of terms purely from $\mathcal{S}_0,\mathcal{S}_1$, and $\mathcal{S}_2$ and, since the subspaces are invariant under $\mathcal{L}_0$, these terms must be annihilated by $\mathcal{L}_0^\dagger$ individually. This yields

$$\rho_{\mathrm{ss}}^0=|e_2\rangle\langle e_2|,$$

$$\rho_{\mathrm{ss}}^1=|s_{12}\rangle\langle s_{12}|+|a_{12}\rangle\langle a_{12}|+|s_{23}\rangle\langle s_{23}|+|a_{23}\rangle\langle a_{23}|,$$

$$\rho_{\mathrm{ss}}^2=|g\rangle\langle g|+|s_{13}\rangle\langle s_{13}|+|a_{13}\rangle\langle a_{13}|+|e_3\rangle\langle e_3|,$$

since the sum of the right-hand sides indeed yields $\mathbb{1}$ and the normalization condition of Eq. (15) is fulfilled. From Eq. (17) one then obtains the α_{ij}. Inserting now $\mathcal{L}_1\rho_{\mathrm{ss},i}$ into Eq. (13) one obtains

$$\rho(t_0+\Delta t)=\rho_{\mathrm{ss},i}+\int_0^{\Delta t}\mathrm{d}\tau\left(\sum_{j=0}^{2}\alpha_{ij}\rho_{\mathrm{ss},j}+e^{\mathcal{L}_0\tau}\tilde{\rho}\right) \tag{20a}$$

$$=\rho_{\mathrm{ss},i}+\sum_{j=0}^{2}\alpha_{ij}\rho_{\mathrm{ss},j}\Delta t+(\epsilon-\mathcal{L}_0)^{-1}\tilde{\rho}, \tag{20b}$$

where for the $\tilde{\rho}$ term the upper integration limit can be extended to infinity since $\tilde{\rho}$ belongs to nonzero eigenvalues of $\mathcal{L}_0$ and is therefore rapidly damped. Now, $\mathcal{L}_0^{-1}$ is of the order of A_3^{-1} and Ω_3^{-1} on $\tilde{\rho}$, and thus the last term in Eq. (20b) is of the order of $\mathcal{L}_1/(A_3,\Omega_3)$, which is much smaller than $\alpha_{ij}\Delta t\sim\mathcal{L}_1\Delta t$, by Eq. (9). Therefore the last term in Eq. (20b) can be neglected, and this equation then reveals that the α_{ij}'s have the meaning of transition rates from subspace $\mathcal{S}_i$ to $\mathcal{S}_j$, i.e.,

$$p_{ij}=\alpha_{ij}. \tag{21}$$

The transition rates are now obtained from Eqs. (21), (17), and (10)–(12) as

$$p_{01}=2A_1, \tag{22a}$$

$$p_{10}=\frac{A_2\Omega_3^2}{A_3^2+2\Omega_3^2+4\Delta_3^2}, \tag{22b}$$

$$p_{12}=A_1, \tag{22c}$$

and

$$p_{21}=2\frac{A_2\Omega_3^2[A_3^2+2\Omega_3^2+4\Delta_3^2]}{[A_3^2+2\Omega_3^2+4\Delta_3^2]^2+[A_3^2+4\Delta_3^2][|C_3|^2+2A_3\,\mathrm{Re}\,C_3+4\Delta_3\,\mathrm{Im}\,C_3]}$$

$$=\frac{2A_2\Omega_3^2}{A_3^2+2\Omega_3^2+4\Delta_3^2}\left[1-2\,\mathrm{Re}\,C_3\frac{A_3(A_3^2+4\Delta_3^2)}{(A_3^2+2\Omega_3^2+4\Delta_3^2)^2}-4\,\mathrm{Im}\,C_3\frac{\Delta_3(A_3^2+4\Delta_3^2)}{(A_3^2+2\Omega_3^2+4\Delta_3^2)^2}\right]+O(C_3^2). \tag{22d}$$

One sees that, for two D systems, only p_{21} depends on the dipole coupling constants C_3 in first order. This contrasts with two V systems, where both p_{21} and p_{12} depend on C_3. The physical reason for this is that transitions between bright periods for two D systems are due to decays and not due to the laser. Hence the transition rates should essentially be governed by the Einstein coefficients of these decays on one hand and by the population of the states in the initial subsystem on the other hand. Therefore the parameter C_3 enters only through the quasistationary state $\rho_{\mathrm{ss},i}$ of the initial subsystem. The absence of a linear C_1 and C_2 dependence can be understood from Fig. 3(b) as follows. For most slow transitions between two subspaces there are two channels with rates $A_j \pm \mathrm{Re}\, C_j$ so that $\mathrm{Re}\, C_j$ cancels. States with a single decay channel lie in $\mathcal{S}_1$ and, by symmetry, they appear in pairs with different sign of $\mathrm{Re}\, C_j$.

From the transition rates p_{ij} one obtains the double-jump rate n_{DJ} by the formula [2]

$$n_{\mathrm{DJ}} = 2\frac{p_{10}p_{01}p_{12}p_{21}}{p_{01}p_{21}+p_{21}p_{10}+p_{01}p_{12}}\Delta T_{\mathrm{DJ}}, \tag{23}$$

where ΔT_{DJ} is the defining small time interval for a double jump. Significant cooperative effects occur only as long as Ω_3 and Δ_3 are at least an order of magnitude smaller than A_3. When compared to noninteracting systems, the cooperative effects are up to 30% for distances between one and two wavelengths and 5% around ten wavelengths, similar as for two V systems. For longer distances they are practically absent and this is consistent with the experimental results of Ref. [13]. Figure 4 shows p_{21} and n_{DJ} versus the relative distance r/λ_3 for typical parameters, where λ_3 is the wavelength of the strong transition.

Our explicit results for arbitrary r confirm the large-distance result of Ref. [20], which argued that for $\lambda_1, \lambda_3 \ll r \ll \lambda_2$ cooperative effects are "suppressed by the rapid decay on the fast transition." In fact, we find in the Appendix that these effects are only to first order independent of the coupling parameter C_2; the second-order contributions in C_2 are, however, negligible for the experimental values of Ref. [13].

III. TWO DIPOLE-INTERACTING FOUR-LEVEL SYSTEMS AS A MODEL FOR TWO Ba^{+} IONS

The experiments of Refs. [3,4] used Ba^{+}. As explained in the introduction we model the relevant level scheme by the effective four-level configuration in Fig. 2(b). The $|1\rangle$–$|4\rangle$ transition is driven weakly and incoherently by a lamp, while the $|1\rangle$–$|3\rangle$ transition is driven coherently by a strong laser. This time the Bloch equation can be written as [26]

$$\begin{aligned}\dot{\rho} &= -\frac{\mathrm{i}}{\hbar}[H_{\mathrm{cond}}\rho - \rho H_{\mathrm{cond}}^{\dagger}] + \mathcal{R}_W(\rho) + \mathcal{R}(\rho)\\ &\equiv \{\mathcal{L}_0^{(0)}(A_2, A_3, A_4, \Omega_3, \Delta_3, C_3) + \mathcal{L}_0^{(1)}(C_2, C_4)\}\rho,\end{aligned} \tag{24}$$

where $\mathcal{R}_{\mathrm{W}}(\rho)$ describes the incoherent driving as in Ref. [26] and is given explicitly below. The Dicke states are defined in analogy to Eq. (2), and H_{cond} and $\mathcal{R}(\rho)$ can be calculated as in Refs. [2,25] as

$$\begin{aligned}H_{\mathrm{cond}} = \frac{\hbar}{2\mathrm{i}}\Bigg\{ & A_1[2|e_2\rangle\langle e_2| + |s_{12}\rangle\langle s_{12}| + |a_{12}\rangle\langle a_{12}| + |s_{23}\rangle\langle s_{23}| + |a_{23}\rangle\langle a_{23}| + |s_{24}\rangle\langle s_{24}| + |a_{24}\rangle\langle a_{24}|] + (A_2 + A_4)\\ & \times\left[2|e_4\rangle\langle e_4| + \sum_{j=1}^{3}\{|s_{j4}\rangle\langle s_{j4}| + |a_{j4}\rangle\langle a_{j4}|\}\right] + (A_3 + 2\mathrm{i}\Delta_3)[2|e_3\rangle\langle e_3| + |s_{13}\rangle\langle s_{13}| + |a_{13}\rangle\langle a_{13}| + |s_{23}\rangle\langle s_{23}|\\ & + |a_{23}\rangle\langle a_{23}| + |s_{34}\rangle\langle s_{34}| + |a_{34}\rangle\langle a_{34}|] + W\Bigg[2|g\rangle\langle g| + 2|e_4\rangle\langle e_4| + \sum_{j=1}^{3}\{|s_{j4}\rangle\langle s_{j4}| + |a_{j4}\rangle\langle a_{j4}|\}\\ & + \sum_{j=2}^{4}\{|s_{1j}\rangle\langle s_{1j}| + |a_{1j}\rangle\langle a_{1j}|\}\Bigg]\Bigg\} + \frac{\hbar}{2\mathrm{i}}\{C_2(|s_{23}\rangle\langle s_{23}| - |a_{23}\rangle\langle a_{23}|) + C_3(|s_{13}\rangle\langle s_{13}| - |a_{13}\rangle\langle a_{13}|) + C_4(|s_{14}\rangle\langle s_{14}|\\ & - |a_{14}\rangle\langle a_{14}|)\} + \frac{\hbar}{2}\Omega_3\{\sqrt{2}(|g\rangle\langle s_{13}| + |s_{13}\rangle\langle e_3|) + |s_{12}\rangle\langle s_{23}| - |a_{12}\rangle\langle a_{23}| + |s_{14}\rangle\langle s_{34}| + |a_{14}\rangle\langle a_{34}| + \mathrm{H.c.}\},\end{aligned} \tag{25}$$

$$\mathcal{R}(\rho) = \sum_{j=1}^{4}[(A_j + \mathrm{Re}\, C_j)R_{+}^{(j)}\rho R_{+}^{(j)\dagger} + (A_j - \mathrm{Re}\, C_j)R_{-}^{(j)}\rho R_{-}^{(j)\dagger}], \tag{26}$$

with

$$R_+^{(1)} = |g\rangle\langle s_{12}| + |s_{12}\rangle\langle e_2| + \frac{1}{\sqrt{2}}(|s_{13}\rangle\langle s_{23}| + |a_{13}\rangle\langle a_{23}| + |s_{14}\rangle\langle s_{24}| + |a_{14}\rangle\langle a_{24}|),$$

$$R_-^{(1)} = |g\rangle\langle a_{12}| + |a_{12}\rangle\langle e_2| + \frac{1}{\sqrt{2}}(|a_{13}\rangle\langle s_{23}| - |s_{13}\rangle\langle a_{23}| + |a_{14}\rangle\langle s_{24}| - |s_{14}\rangle\langle a_{24}|),$$

$$R_+^{(2)} = |e_2\rangle\langle s_{24}| + |s_{24}\rangle\langle e_4| + \frac{1}{\sqrt{2}}(|s_{12}\rangle\langle s_{14}| + |a_{12}\rangle\langle a_{14}| + |s_{23}\rangle\langle s_{34}| - |a_{23}\rangle\langle a_{34}|),$$

$$R_-^{(2)} = |e_2\rangle\langle a_{24}| + |a_{24}\rangle\langle e_4| + \frac{1}{\sqrt{2}}(|s_{12}\rangle\langle a_{14}| - |a_{12}\rangle\langle s_{14}| + |s_{23}\rangle\langle a_{34}| + |a_{23}\rangle\langle s_{34}|),$$

$$R_+^{(3)} = |g\rangle\langle s_{13}| + |s_{13}\rangle\langle e_3| + \frac{1}{\sqrt{2}}(|s_{12}\rangle\langle s_{23}| - |a_{12}\rangle\langle a_{23}| + |s_{14}\rangle\langle s_{34}| + |a_{14}\rangle\langle a_{34}|),$$

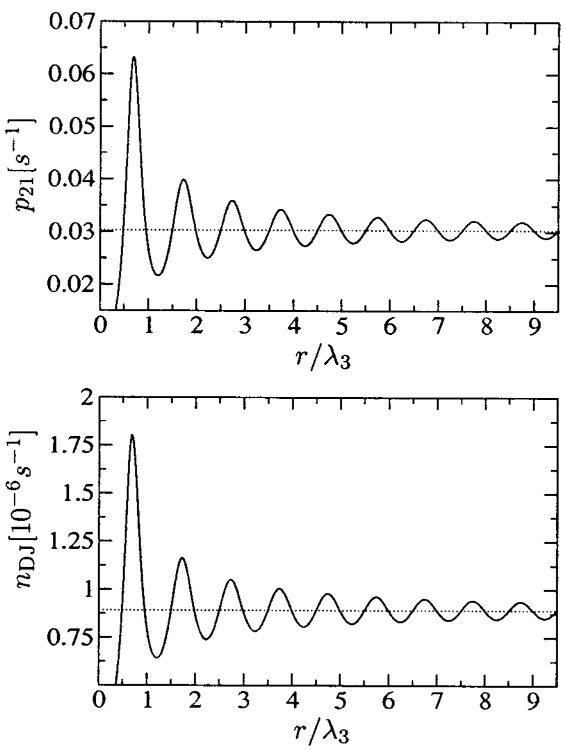

FIG. 4. Transition rate p_{21} and double-jump rate n_{DJ} for two dipole-interacting D systems. The dashed lines show the case of independent systems. Parameter values are $A_1 = 1\ s^{-1}$, $A_2 = 1\ s^{-1}$, $A_3 = 4\times 10^8\ s^{-1}$, $\Omega_3 = 5\cdot 10^7\ s^{-1}$, and $\Delta_3 = 0$.

$$R_-^{(3)} = |g\rangle\langle a_{13}| + |a_{13}\rangle\langle e_3| + \frac{1}{\sqrt{2}}(|s_{12}\rangle\langle a_{23}| + |a_{12}\rangle\langle s_{23}| + |s_{14}\rangle\langle a_{34}| - |a_{14}\rangle\langle s_{34}|),$$

$$R_+^{(4)} = |g\rangle\langle s_{14}| + |s_{14}\rangle\langle e_4| + \frac{1}{\sqrt{2}}(|s_{12}\rangle\langle s_{24}| - |a_{12}\rangle\langle a_{24}| + |s_{13}\rangle\langle s_{34}| - |a_{13}\rangle\langle a_{34}|),$$

$$R_-^{(4)} = |g\rangle\langle a_{14}| + |a_{14}\rangle\langle e_4| + \frac{1}{\sqrt{2}}(|s_{12}\rangle\langle a_{24}| + |a_{12}\rangle\langle s_{24}| + |s_{13}\rangle\langle a_{34}| + |a_{13}\rangle\langle s_{34}|).$$

The lamp term is obtained as in Ref. [26] as

$$\mathcal{R}_W(\rho) = W(R_{4+}\rho R_{4+}^\dagger + R_{4-}\rho R_{4-}^\dagger + R_{4+}^\dagger \rho R_{4+} + R_{4-}^\dagger \rho R_{4-}), \tag{27}$$

where W is the product of the spectral energy density of the lamp and the Einstein B coefficient of the $|1\rangle$–$|4\rangle$ transition.

Now the procedure is similar as for the D system. The Liouvillean $\mathcal{L}_0$ possesses three (quasi) stationary states $\rho_{\mathrm{ss},0}$, $\rho_{\mathrm{ss},1}$, and $\rho_{\mathrm{ss},2}$, which coincide with those for the D systems in Eqs. (10)–(12) and which are associated with the dark and the two bright periods. As before, one calculates $\rho(t_0+\Delta t)$ as in Eq. (13) and decomposes $\mathcal{L}_1\rho_{\mathrm{ss},i}$ as in Eq. (14). Now, however, the reciprocals ρ_{ss}^i are more difficult to determine since $|4\rangle$ can decay into $|1\rangle$ as well as $|2\rangle$. An exact solution of $\mathcal{L}_0^\dagger \rho_{\mathrm{ss}}^i = 0$ is rather elaborate. We therefore decompose

$$\mathcal{L}_0^\dagger = \mathcal{L}_0^{\dagger(0)}(A_2, A_3, A_4, \Omega_3, \Delta_3, C_3) + \mathcal{L}_0^{\dagger(1)}(C_2, C_4) \tag{28}$$

and, by Maple, have calculated ρ_{ss}^i to first order in perturbation theory with respect to C_2 and C_4, with the same constraint as in Eq. (15). The lengthy result will not be given here explicitly. The transition rates are again given by

$$p_{ij} = \mathrm{Tr}(\rho_{\mathrm{ss}}^{j\dagger}\mathcal{L}_1\rho_{\mathrm{ss},i}) \tag{29}$$

and one obtains for two dipole-interacting four-level systems of Fig. 2(b) to first order in C_2 and C_4,

$$p_{01} = 2A_1, \tag{30a}$$

$$p_{10} = \frac{A_2 W(A_3^2 + \Omega_3^2 + 4\Delta_3^2)}{(A_2 + A_4)[A_3^2 + 2\Omega_3^2 + 4\Delta_3^2]}, \tag{30b}$$

$$p_{12} = A_1, \tag{30c}$$

and

$$p_{21}=2A_2W\frac{(A_3^2+\Omega_3^2+4\Delta_3^2)(A_3^2+2\Omega_3^2+4\Delta_3^2)+(A_3^2+4\Delta_3^2)(|C_3|^2+2A_3\mathrm{Re}\,C_3+4\Delta_3\,\mathrm{Im}\,C_3)}{(A_2+A_4)(A_3^2+2\Omega_3^2+4\Delta_3^2)^2+(A_3^2+4\Delta_3^2)(|C_3|^2+2A_3\mathrm{Re}\,C_3+4\Delta_3\,\mathrm{Im}\,C_3)}$$
$$=2A_2W\left[\frac{A_3^2+\Omega_3^2+4\Delta_3^2}{(A_2+A_4)[A_3^2+2\Omega_3^2+4\Delta_3^2]}+2\,\mathrm{Re}\,C_3\frac{A_3\Omega_3^2(A_3^2+4\Delta_3^2)}{(A_2+A_4)[A_3^2+2\Omega_3^2+4\Delta_3^2]^3}+4\,\mathrm{Im}\,C_3\frac{\Delta_3\Omega_3^2(A_3^2+4\Delta_3^2)}{(A_2+A_4)[A_3^2+2\Omega_3^2+4\Delta_3^2]^3}\right]$$
$$+O(C_3^2). \tag{30d}$$

It is seen that p_{01}, p_{10}, and p_{12} are independent of the coupling parameters and are thus the same as for noninteracting systems.

These results for two four-level systems show great similarity with those for the two D systems of the proceeding section. In both cases only p_{21} depends to first order on C_3, the coupling parameter associated with the laser-driven transition. However, cooperative effects are significantly smaller for the two four-level systems. For fixed laser detuning, the effect of C_3 becomes maximal for $\Omega_3=\frac{1}{2}\sqrt{\sqrt{5}-1}\sqrt{A_3^2+4\Delta_3^2}$. For this value of Ω_3, Fig. 5 shows the transition rate p_{21} from a double intensity period to a unit-intensity period and the double-jump rate n_{DJ} over the relative distance r/λ_3, with the other parameters as in the experiment [3,4]. Despite the optimal choice of the Rabi frequency Ω_3 the deviations from the value for non-interacting systems are very small. Already for a distance of about a wavelength λ_3, they are not more than 1% for p_{21} when compared to noninteracting systems, while for n_{DJ} they are less than 1‰.

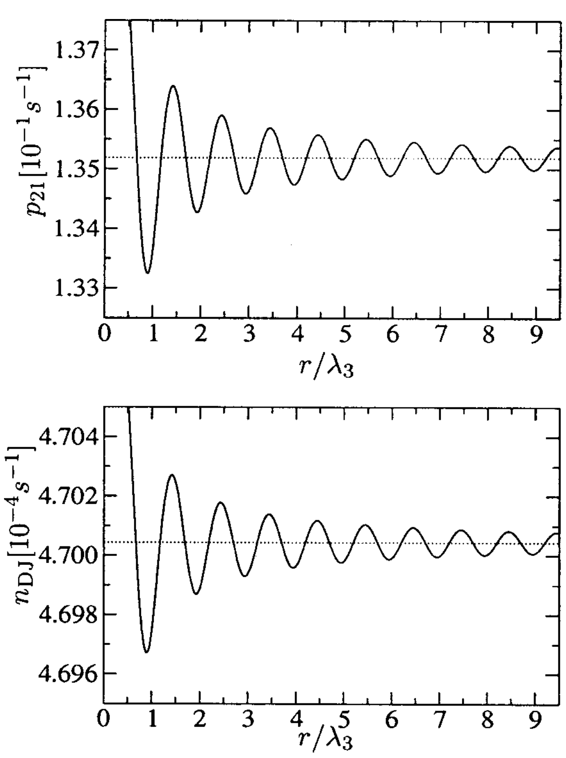

FIG. 5. Transition rate p_{21} and double-jump rate n_{DJ} for dipole-interacting four-level systems, with optimal $\Omega_3=\frac{1}{2}\sqrt{\sqrt{5}-1}\sqrt{A_3^2+4\Delta_3^2}$ and all other parameters as in the experiment [3]. The dashed lines show the case of independent systems.

IV. CONCLUSIONS

We have investigated the effect of the dipole-dipole interaction for two fluorescing systems with macroscopic light and dark periods, first for three-level D configurations and then for four-level systems. The three-level D configuration models the relevant levels of Hg^+ used in the experiments of Ref. [13,23], and the four-level configuration is an effective model for Ba^+, used in the experiments of Ref. [3,19]. For these systems one has macroscopic light and dark periods, and their statistics can be a sensitive test of the dipole-dipole interaction. We have explicitly calculated the transition rates between the different light and dark periods by employing the Bloch equations as well as a direct quantum jump approach. From the transition rates the double jump rates are obtained.

For two D systems the effect of the dipole-dipole interaction is of similar magnitude as for two V systems investigated earlier [2] and shown to be up to 30 % for distances of the order of a wavelength of the strong transition and about 5% around ten wave lengths, when compared to independent systems. For longer distances they are practically absent and this is in agreement with the experimental results of Ref. [13]. We have also recovered the special case of Ref. [20], where distances satisfying $\lambda_1,\ \lambda_3\ll r\ll\lambda_2$ were considered and where an argument for the nondependence on the dipole coupling constant C_2 was given. Here we have shown that this holds to first order and that the explicitly determined second-order terms are negligibly small.

For the effective model of two Ba^+ systems our results yield very small and hardly observable cooperative effects for the double-jump rate. This is at odds with the experimental result in Ref. [3]. Our method also applies to three Ba^+ ions, but this is more tedious and requires another paper. Also a theoretical investigation of the experiments with Ca^+ [14,15] is possible with our method. For this the calculations have to be carried over to a level scheme which models the levels of Ca^+.

A further conclusion of our work is the observation that the magnitude of cooperative effects due to the dipole-dipole interaction sensitively depends on how the metastable level is populated.

APPENDIX: QUANTUM JUMP APPROACH FOR TWO D SYSTEMS

The procedure will first be explained for a single D system which has just two types of periods, light and dark ones. From its level configuration in Fig. 1 it is evident that the onset of a dark period is preceded by a photon from the $|3\rangle$–$|2\rangle$ transition, with frequency ω_2. Hence, starting at $t_0 = 0$ in $|1\rangle$, the probability density for the next photon to occur at time t and to come from the $|3\rangle$–$|2\rangle$ transition is

$$w_{1\omega_2}(t) = A_2 |\langle 3| \mathrm{e}^{-\mathrm{i}H_{\mathrm{cond}}t/\hbar}|1\rangle|^2, \tag{A1}$$

since H_{cond} gives the time development between photon emissions [22]. Then its time integral,

$$P_{\omega_2} = \int_0^\infty \mathrm{d}t\, w_{1\omega_2}(t), \tag{A2}$$

is the probability for the next emitted photon to come from the $|3\rangle$–$|2\rangle$ transition. Now, let the photon rate in a light period be denoted by I_L. Then, after each photon of the light period the system is reset to the ground state and thereafter, with probability P_{ω_2}, emits a photon from the $|3\rangle$–$|2\rangle$ transition. Hence the transition rate from a light to a dark period is

$$p_{10} = I_L P_{\omega_2}. \tag{A3}$$

This can be carried over to two dipole interacting D systems as follows. We consider an emission trajectory and assume to be in a particular intensity period say, of unit intensity. In contrast to a single D system, the reset state after a photon emission in this period is not always quite the same, but it is reasonable to start from $\rho_{\mathrm{ss},1}$ and to use

$$\overline{\rho_1} \equiv \{(A_3 + \mathrm{Re}\, C_3) R_+^{(3)} \rho_{\mathrm{ss},1} R_+^{(3)\dagger} + (A_3 - \mathrm{Re}\, C_3) R_-^{(3)} \rho_{\mathrm{ss},1} R_-^{(3)\dagger}\}/\mathrm{Tr}(\cdot) \tag{A4}$$

as an average reset state. The transition to a double-intensity period is marked by a photon from the $|2\rangle$–$|1\rangle$ transition, and therefore the probability density for such a transition, starting from the above reset state, is

$$w_{1\omega_1}(t) = \mathrm{Tr}\{(A_1 + \mathrm{Re}\, C_1) R_+^{(1)} \mathrm{e}^{-iH_{\mathrm{cond}}t/\hbar} \overline{\rho_1} \mathrm{e}^{iH_{\mathrm{cond}}^\dagger t/\hbar} R_+^{(1)\dagger} + (A_1 - \mathrm{Re}\, C_1) R_-^{(1)} \mathrm{e}^{-iH_{\mathrm{cond}}t/\hbar} \overline{\rho_1} \mathrm{e}^{iH_{\mathrm{cond}}^\dagger t/\hbar} R_-^{(1)\dagger}\}. \tag{A5}$$

Integration over t gives the total transition probability, denoted by $P_{1\omega_1}$. The photon rate in a period of unit intensity is that of two dipole interacting two-level systems and is given by [27]

$$I_{\mathrm{ss}}^{(2)} = 2 \frac{\Omega_3^2 [A_3(A_3^2 + 2\Omega_3^2 + 4\Delta_3^2) + \mathrm{Re}\, C_3 (A_3^2 + 4\Delta_3^2)]}{(A_3^2 + 2\Omega_3^2 + 4\Delta_3^2)^2 + (A_3^2 + 4\Delta_3^2)(|C_3|^2 + 2A_3 \,\mathrm{Re}\, C_3 + 4\Delta_3 \,\mathrm{Im}\, C_3)}. \tag{A6}$$

Thus p_{12} is given by

$$p_{12} = I_{\mathrm{ss}}^{(2)} P_{1\omega_1}. \tag{A7}$$

In a similar way one obtains p_{10} and p_{21}. The transition rate p_{01} can be directly read off from the no-photon probability $e^{-2A_1 t}$. One obtains the same results as in Sec. II when one expands in the small parameters. In the case $\lambda_1, \lambda_3 \ll r \ll \lambda_2$ one can put $C_1 = C_3 = 0$ and one obtains, for example,

$$p_{12} = A_1 \left(1 + \frac{\mathrm{Im}\, C_2^2}{A_3^2 + 2\Omega_3^2 + 4\Delta_3^2} \right). \tag{A8}$$

[1] For references cf., e.g., those in Ref. [2].
[2] S. Addicks, A. Beige, M. Dakna, and G.C. Hegerfeldt, Eur. Phys. J. D **15**, 393 (2001).
[3] T. Sauter, R. Blatt, W. Neuhauser, and P.E. Toschek, Opt. Commun. **60**, 287 (1986).
[4] T. Sauter, Ph. D. thesis, Universität Hamburg, 1987 (unpublished).
[5] B.H.W. Hendriks and G. Nienhus, J. Mod. Opt. **35**, 1331 (1988).
[6] M. Lewenstein and J. Javanainen, Phys. Rev. Lett. **59**, 1289 (1987).
[7] M. Lewenstein and J. Javanainen, IEEE J. Quantum Electron. **42**, 1403 (1988).
[8] G.S. Agarwal, S.V. Lawande, and R. D'Souza, IEEE J. Quantum Electron. **24**, 1413 (1988).
[9] S.V. Lawande, Q.V. Lawande, and B.N. Jagatap, Phys. Rev. A **40**, 3434 (1989).
[10] C.R. Fu and C.D. Gong, Phys. Rev. A **45**, 5095 (1992).
[11] R.C. Thompson, D.J. Bates, K. Dholakia, D.M. Segal, and D.C. Wilson, Phys. Scr. **46**, 285 (1992).
[12] D.J. Berkeland, D.A. Raymondson, and V.M. Tassin, e-print physics/0304013.
[13] W.M. Itano, J.C. Bergquist, and D.J. Wineland, Phys. Rev. A **38**, 559 (1988).

[14] M. Block, O. Rehm, P. Seibert, and G. Werth, Eur. Phys. J. D **7**, 461 (1999).
[15] C.J.S. Donald, D.M. Lucas, P.A. Barton, M.J. McDonnell, J.P. Stacey, D.A. Stevens, D.N. Stacey, and A.M. Steane, Europhys. Lett. **51**, 388 (2000).
[16] For references cf., e. g., Ref. [17].
[17] A. Beige and G.C. Hegerfeldt, Phys. Rev. A **59**, 2385 (1999).
[18] C. Skornia, J. von Zanthier, G.S. Agarwal, E. Werner, and H. Walther, Phys. Rev. A **64**, 053803 (2001).
[19] T. Sauter, W. Neuhauser, R. Blatt, and P.E. Toschek, Phys. Rev. Lett. **57**, 1696 (1986).
[20] C. Skornia, J. von Zanthier, G.S. Agarwal, E. Werner, and H. Walther, Europhys. Lett. **56**, 665 (2001).
[21] In Ref. [3] it is stated ''We have also demonstrated, with the light of two or three ions in the trap observed, that simultaneous quantum jumps use to happen at a rate large exceeding random coincidence.''
[22] G.C. Hegerfeldt and T.S. Wilser, in *Proceedings of the Second International Wigner Symposium, 1991*, edited by H.D. Doebner, W. Scherer, and F. Schroeck (World Scientific, Singapore, 1992), p. 104; G.C. Hegerfeldt, Phys. Rev. A **47**, 449 (1993); G.C. Hegerfeldt and D.G. Sondermann, Quantum Semiclassic. Opt. **8**, 121 (1996); for a review cf. M.B. Plenio and P.L. Knight, Rev. Mod. Phys. **70**, 101 (1998); the quantum jump approach is essentially equivalent to the Monte Carlo wavefunction approach of J. Dalibard, Y. Castin, and K. Mølmer, Phys. Rev. Lett. **68**, 580 (1992), and to the quantum trajectories of H. Carmichael, *An Open Systems Approach to Quantum Optics*, Lecture Notes in Physics Vol. m18 (Springer, Berlin, 1993).
[23] W.M. Itano, J.C. Bergquist, R.G. Hulet, and D.J. Wineland, Phys. Rev. Lett. **59**, 2732 (1987).
[24] G.C. Hegerfeldt, Phys. Rev. A **47**, 449 (1993).
[25] A. Beige, Ph.D. thesis, Universität Göttingen, 1997 (unpublished).
[26] G.C. Hegerfeldt and M.B. Plenio, Phys. Rev. A **47**, 2186 (1993).
[27] A. Beige and G.C. Hegerfeldt, Phys. Rev. A **58**, 4133 (1998).