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Inhibition of cooperative quantum jumps due to fast spontaneous decay

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Abstract. – A Quantum Monte Carlo wave function approach is used to rule out the possibility of cooperative effects in the quantum jump statistics of adjacent three-level ions of the type discussed in a series of experiments and theoretical investigations. By deriving analytical results and examining quantum jumps in various subspaces, we track this outcome to the fact that a fast spontaneous decay inhibits dipole-dipole–induced entanglement between the ions developing on a much slower time scale. This inhibition is demonstrated even for weak pumping of the fast transition. The Quantum Monte Carlo calculation thus sheds new light on the controversy arising from the findings of two recent conflicting experiments by Block *et al.* (*Eur. Phys. J. D*, **7** (1999) 461) and Donald *et al.* (*Europhys. Lett.*, **51** (2000) 388).

Introduction. – For some time there has been a controversy whether cooperative effects show up in the quantum jump statistics of a collection of atoms or ions placed in the vicinity of each other. Quantum jumps can be observed in a multilevel system of a single particle if one of the states is metastable [1–4]. For example, if the internal structure of the atom corresponds to the 3-level system of fig. 1, the electron is shelved in the metastable state $|2\rangle$ either due to spontaneous decay (in a Λ-scheme) or excitation by a laser (in a V-scheme); in both cases photons cease to be scattered on the fast $|1\rangle$-$|3\rangle$ transition [5–7]. Sauter *et al.* reported the observation of simultaneous quantum jumps of groups of ions within a given time interval at a rate much higher than expected for an ensemble of independently radiating ions [8]. Lewenstein *et al.* published a theoretical calculation stating that in a Λ-scheme collective behaviour in the quantum jump statistics of adjacent ions is not observable unless the particles are separated by an (unrealistically small) fraction of the shortest of the wavelengths involved [9]. Beige *et al.* and Addicks *et al.* derived cooperative quantum jump behaviour, scaling with the wavelength of the fast transition, if the ions are excited coherently in a V-configuration on both transitions [10, 11]. In spite of this result it remained unclear why collective effects in the quantum jump behaviour are not displayed if the ions are placed close to each other and coherently excited in a Λ-scheme only on the fast transition. An experiment similar to the assumptions of [9] was later performed by Itano *et al.* [12]. Here, two (or three) mercury ions, separated within a wavelength of the $|1\rangle$-$|2\rangle$ transition, did not show any deviation from

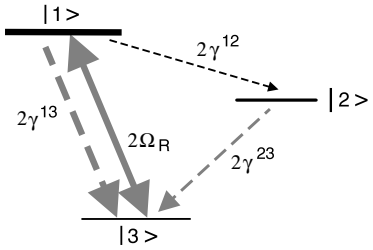

Fig. 1

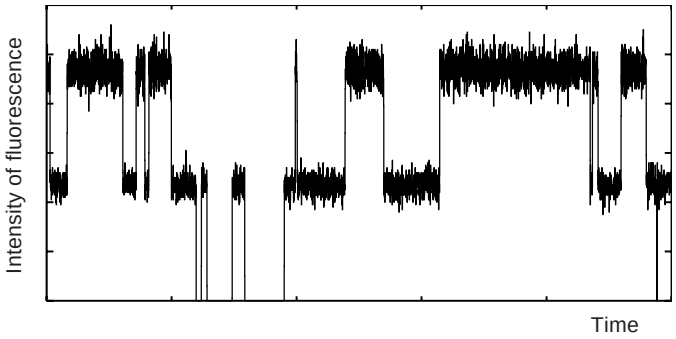

Fig. 2

Fig. 1 – Atomic 3-level Λ-scheme considered in this paper. Due to spontaneous decay quantum jumps occur towards the metastable state $|2\rangle$ at a rate $2\,\gamma^{12}$. From $|2\rangle$ the atom spontaneously decays towards $|3\rangle$ at a rate $2\,\gamma^{23}$.

Fig. 2 – Quantum Monte Carlo simulation of the fluorescence record of two 3-level Λ-systems (with $\gamma^{13} \gg \gamma^{12}, \gamma^{23}$ and $\lambda_{12} \gg r \gg \lambda_{13}$) as a function of time. Three different levels of fluorescence can be distinguished, corresponding to zero, single and double intensity of the single-ion fluorescence.

the photon statistics of randomly radiating ions. Recently, the question whether collective quantum jumps exist in an ensemble of ions with a more complex Λ-scheme was again raised due to the outcome of an investigation made by Block *et al.* [13]. In this experiment, simultaneous jumps in a linear chain of ten calcium ions were observed much more frequently than expected from pure statistics. Since the coincidences of quantum jumps were not confined to adjacent ions, the authors speculated that unexplained long-range interactions between the ions in the linear crystal may exist. The results were rejected later by an experiment using the same ion species (and level structure) that was conducted under similar conditions by Donald *et al.* [14]. Here again, no evidence of ion-ion correlations was found.

As pointed out in [14], the question of collectivity in an ensemble of trapped ions is of importance in the context of quantum information processing. The advantage of using ions in a quantum computer relies heavily on the assumption that the interactions governing the behaviour of the particles in the trap are well understood in principle. Unexplained cooperativity of the trapped ions would seriously call this assumption into question [15].

This paper reinvestigates the problem of collective behaviour in the quantum jump statistics of closely separated dipole-dipole interacting ions in the Λ-configuration. The internal level structure of the ions corresponds to the 3-level scheme of fig. 1, investigated by Lewenstein *et al.* [9] and experimentally by Itano *et al.* [12]. For the analysis we make use of a Quantum Monte Carlo wave function calculation. In contrast to the master-equation approach employed by Lewenstein *et al.* [9], this has the advantage of allowing us to follow closely the dynamics of individual quantum systems as investigated here. Our calculations reconfirm the prediction that for this configuration no collective behaviour is observed in the quantum jump behaviour of two dipole-dipole interacting particles. We can show that this result holds for any pump power —weak or strong— taking fully into account the real and imaginary parts of the dipole-dipole interaction, level shifts and coherent evolution among the internal states of the two ions. The Quantum Monte Carlo calculation even allows us to carry out the jump statistics analytically. We can thus clearly understand why in fact cooperative behaviour is not displayed. The principal result is that due to the rapid decay on the fast $|1\rangle$-$|3\rangle$ transition any dipole-dipole–induced coherent evolution on the $|1\rangle$-$|2\rangle$ transition is brought to an abrupt

stop. This behaviour is in close analogy to the Quantum Zeno effect encountered in 3-level systems [16, 17].

In what follows, we first present the mathematical approach used to analyze the dynamics of the two-atom system (second section). Next, we derive the transition rates between the different subspaces of fluorescence corresponding to zero, single and double intensity of the single-ion fluorescence (third section). Finally, we reconsider the role played by the dipole-dipole interaction in view of the findings of the third section and give an interpretation of the final result (last section).

Model and method. – We consider two identical atoms, each with levels $|1\rangle$, $|2\rangle$ and $|3\rangle$, according to fig. 1. A resonant coherent pumping field interacts continuously with both atoms on the $|1\rangle$-$|3\rangle$ transition. In our analysis it is assumed that the level spacing $|1\rangle$-$|2\rangle$ is close enough so that the dipole-dipole interaction is effective on this transition; the $|1\rangle$-$|3\rangle$ transition is supposed to lie in the optical domain, where the distance between the two atoms is assumed to be much larger than the corresponding wavelength. We thus work in the limit

$$\lambda_{12} \gg r \gg \lambda_{13}\,, \tag{1}$$

which also implies that $r \gg \lambda_{23}$. Here λ_{nm} is the wavelength of the $|n\rangle$-$|m\rangle$ transition, where $n, m \in \{1, 2, 3\}$, $n < m$, and r denotes the ion-ion distance. Since a system is assumed where quantum jumps are observed, it is supposed that

$$\gamma^{13} \gg \gamma^{12}, \gamma^{23}\,, \tag{2}$$

with γ^{23} being sufficiently small so that for the atom in the metastable state $|2\rangle$ the dark period in the fluorescence signal can be clearly resolved by a photodetector.

The two ions are supposed to be at fixed positions $\boldsymbol{r}_i, i = 1, 2$, and the field outside the laser beam is assumed to be in the vacuum state. The dipole matrix elements of the atoms are defined as $\boldsymbol{d}_{nm} := e\langle n|x|m\rangle$, where $|n\rangle$, $|m\rangle$ denote the internal states of the ions and e corresponds to the electron charge. The atomic raising and lowering operators for the internal states of the i-th ion are given by $\sigma_i^{nm} := |n\rangle_i\langle m|$. For the time steps Δt used in the Quantum Monte Carlo simulations we need the time development of the system under the condition that no photon be emitted in the time interval $[t, t + \Delta t]$. The transformation of the state vector of the system $|\Psi(t)\rangle$ during the coherent evolution is computed from a modified Schrödinger equation employing a non-Hermitian conditional Hamiltonian. Details of this approach can be found in [18,19]. The conditional Hamiltonian including the on-resonance ion-laser interaction is found in second-order perturbation theory to be [10]

$$H_{\mathrm{cond}} = \frac{\hbar}{\mathrm{i}}\left[\sum_{\substack{n,m=1\\ n<m}}^{3}\sum_{i,j=1}^{2}\gamma_{ij}^{nm}\sigma_i^{nm}\sigma_j^{mn} + \sum_{i=1}^{2}\mathrm{i}\Omega_{\mathrm{R}}\left(\sigma_i^{13} + \sigma_i^{31}\right)\right]. \tag{3}$$

Here, $2\gamma_{ij}^{nm}$ is the Einstein A-coefficient for the $|n\rangle \leftrightarrow |m\rangle$ transition for $i = j$ and $2\Omega_{\mathrm{R}}$ is the Rabi frequency due to the continuous laser exciting the $|1\rangle$-$|3\rangle$ transition. For $i \neq j$ γ_{ij}^{nm} is the complex parameter which describes the strength of the dipole-dipole interaction. In the Markov approximation γ_{12}^{nm} can be calculated analytically [20]:

$$\gamma_{12}^{nm} = \frac{3}{2}\gamma_{11}^{nm}\mathrm{e}^{\mathrm{i}k_{nm}r}\left[\frac{1}{\mathrm{i}k_{nm}r}\left(1 - \cos^2\theta_{nm}\right) + \left(\frac{1}{(\mathrm{i}k_{nm}r)^2} + \frac{1}{(\mathrm{i}k_{nm}r)^3}\right)\left(1 - 3\cos^2\theta_{nm}\right)\right], \tag{4}$$

where $r = |\boldsymbol{r}_1 - \boldsymbol{r}_2|$ is the distance between the two ions, θ_{nm} the angle between $\boldsymbol{d}_{nm}$ and $\boldsymbol{r}_1 - \boldsymbol{r}_2$ and $k_{nm} = \frac{2\pi}{\lambda_{nm}}$. For further calculations we define $\gamma^{nm} = \gamma_{11}^{nm} = \gamma_{22}^{nm}$, $\gamma_{\mathrm{dd}}^{nm} := \mathrm{Re}\left(\gamma_{12}^{nm}\right)$

and $\Omega_{\rm dd}^{nm} := {\rm Im}\,(\gamma_{12}^{nm})$. The values of $\gamma_{\rm dd}^{nm}(r)$ and $\Omega_{\rm dd}^{nm}(r)$ tend to zero for $r \to \infty$, but $\Omega_{\rm dd}^{nm}$ diverges for $r \to 0$. From this behaviour it is not evident *a priori* why no consequences of the dipole-dipole interaction on the $|1\rangle$-$|2\rangle$ transition between the two ions are derived from the master-equation approach.

The evolution of the state vector between two emissions is given by

$$\frac{\partial}{\partial t}|\Psi\rangle = \frac{i}{\hbar} H_{\rm cond}|\Psi\rangle\,. \tag{5}$$

The quantum jumps of the atomic system are represented by the six operators,

$$\left.\begin{array}{rcl} J_{mn}^{s} & = & \sigma_1^{mn} + \sigma_2^{mn} \\ J_{mn}^{a} & = & \sigma_1^{mn} - \sigma_2^{mn} \end{array}\right\} n,m \in \{1,2,3\}; \qquad n < m\,, \tag{6}$$

with their corresponding probabilities,

$$\begin{array}{rcl} P(J_{mn}^{s}) & = & 2\,(\gamma^{mn} + \gamma_{\rm dd}^{mn})\,\langle\Psi|J_{mn}^{s\,\dagger} J_{mn}^{s}|\Psi\rangle\,, \\ P(J_{mn}^{a}) & = & 2\,(\gamma^{mn} - \gamma_{\rm dd}^{mn})\,\langle\Psi|J_{mn}^{a\,\dagger} J_{mn}^{a}|\Psi\rangle\,, \end{array} \tag{7}$$

for the atomic system being in state $|\Psi\rangle$ just before the jump.

The coherent evolution between two quantum jumps according to (5) and the reduction of the state vector in the case of a quantum jump given by (6) and (7) allow one to model the complete dynamics of the two-atom system.

Fluorescence phases. – The atomic system can be divided into three subspaces, each belonging to different intensities of fluorescence (see fig. 2). We define

$$B_2 := \{|1,1\rangle, |1,3\rangle, |3,1\rangle, |3,3\rangle\}\,, \tag{8}$$

$$B_1 := \{|1,2\rangle, |2,1\rangle, |2,3\rangle, |3,2\rangle\}\,, \tag{9}$$

$$D := \{|2,2\rangle\}\,. \tag{10}$$

If the system is in any state of subspace B_2, both ions are fluorescing; if the system is in any state of subspace B_1, only one ion is fluorescing; D is the dark state, where no fluorescence photons are emitted. The time evolution between two quantum jumps is determined by the conditional Hamiltonian (3). As can be seen from (3), (5) and (8)-(10), any state $|\Psi\rangle$ which is in one of the subspaces B_2, B_1 or D at time t remains in the same subspace for any time $t+\tau$ if no jump of type (6) takes place in between. Therefore, the only possibility to change the intensity of fluorescence and thus the subspaces $\{B_2, B_1, D\}$ is if one of the quantum jumps (6) occurs. For the initial state $|\Psi\rangle$ in B_2 it is clear that only jumps of type J_{21}^{s} or J_{21}^{a} take the system out of B_2. If the initial state $|\Psi\rangle$ is in B_1, there are two possibilities of leaving this subspace: a) one of the jumps of type J_{32}^{s} or J_{32}^{a} will return the system back to B_2 or b) one of the jumps of type J_{21}^{s} or J_{21}^{a} puts the system into the dark state. For the dark state $|2,2\rangle$ only a jump of type J_{32}^{s} or J_{32}^{a} can take place, transferring the system towards B_1.

To clarify how the dipole-dipole interaction influences the quantum jump behaviour of the system, we need to calculate these transition rates explicitly. For example, for a state vector $|\Psi\rangle$ being part of subspace B_2, we can write

$$|\Psi\rangle(t) = b_1|1,1\rangle + b_2|1,3\rangle + b_3|3,1\rangle + b_4|3,3\rangle\,, \tag{11}$$

with $\sum_{i=1}^{4} |b_i|^2 = 1$. From (11) we can calculate the probability of a quantum jump from B_2 to B_1 as

$$P_{21} := P(B_2 \to B_1) = P(J_{21}^{s}) + P(J_{21}^{a}) = 2\gamma^{12}\left(2|b_1|^2 + |b_2|^2 + |b_3|^2\right)\,. \tag{12}$$

It can be seen that the rate P_{21} is independent of the ion distance r. This means that the length of the fluorescence periods when both ions are radiating is not affected by the dipole-dipole interaction between the two particles, irrespective of the employed laser power.

Next we calculate the transition rates P_{10} and P_{12}. For a state $|\Psi\rangle$ in B_1 we can write

$$|\Psi\rangle = c_1|1,2\rangle + c_2|2,1\rangle + c_3|3,2\rangle + c_4|2,3\rangle\,, \tag{13}$$

with $\sum_{i=1}^{4}|c_i|^2 = 1$. From (13), it is then found that

$$\begin{aligned} P_{10} &:= P(B_1 \to D) = P(J_{21}^s) + P(J_{21}^a) \\ &= 2\gamma^{12}\left(|c_1|^2 + |c_2|^2\right) + 4\gamma_{\rm dd}^{12}|c_1||c_2|\cos\left(\phi_{12}\right), \end{aligned} \tag{14}$$

$$\begin{aligned} P_{12} &:= P(B_1 \to B_2) = P(J_{32}^s) + P(J_{32}^a) \\ &= 2\gamma^{23}\left(|c_1|^2 + |c_2|^2 + |c_3|^2 + |c_4|^2\right) + 4\gamma_{\rm dd}^{23}|c_3||c_4|\cos\left(\phi_{34}\right), \end{aligned} \tag{15}$$

where $\phi_{ij} := \arg(c_j) - \arg(c_i)$ denotes the phase difference between c_i and c_j. As can be seen from (14) and (15), P_{10} and P_{12} depend via ϕ_{ij} as well as $\gamma_{\rm dd}^{12}$ and $\gamma_{\rm dd}^{23}$ on the dipole-dipole interaction. This will be analyzed in detail in the next section.

The remaining transition rate P_{01} is found to be

$$P_{01} := P(D \to B_1) = P(J_{32}^s) + P(J_{32}^a) = 4\gamma^{23}, \tag{16}$$

i.e. the probability of leaving the dark state $|2,2\rangle$ is simply twice the decay rate $2\gamma^{23}$ of the individual atom system. This transition rate is independent of dipole-dipole interaction.

Note that the transition rate $P_{20} := P(B_2 \to D)$ is zero for any state $|\Psi\rangle \in B_2$. For the system under investigation there is thus no probability of a simultaneous jump $|\Psi\rangle \to |2,2\rangle$ for $|\Psi\rangle \in B_2$ occurring.

Dipole-dipole interaction. – From the calculated rates (12), (14)-(16) above, it is apparent that the only influence of the dipole-dipole interaction on the transition probabilities is to be expected when leaving subspace B_1. To analyze (14) and (15) any further, let us investigate the phase difference ϕ_{ij} more closely. For that purpose, let us introduce the functions f_i, $i = 1, \ldots, 4$:

$$f_i := \frac{\rm d}{{\rm d}t}\tan(\arg(c_i)) = \frac{\rm d}{{\rm d}t}\frac{{\rm Im}(c_i)}{{\rm Re}(c_i)}\,. \tag{17}$$

These can be calculated explicitly from (3) in the limit (1), *i.e.* for $\gamma_{\rm dd}^{13} \ll \gamma^{13}$. If no quantum jump occurs, the conditional Hamiltonian $H_{\rm cond}$ is found to drive the system into a state where $f_1 - f_2 = 0$ and $f_3 - f_4 = 0$, *i.e.* into a state where ϕ_{12} and ϕ_{34} are not changed any further by $H_{\rm cond}$ (see fig. 3). The solutions of ϕ_{12} and ϕ_{34} corresponding to the condition $f_1 - f_2 = f_3 - f_4 = 0$ can immediately be calculated. One obtains

$$c_1 = c_2\,, \qquad c_3 = c_4\,, \qquad c_1 = -c_2\,, \qquad c_3 = -c_4\,, \tag{18}$$

with the corresponding fixed phase relations $\phi_{12} = \phi_{34} = 0, \pi$. On a time scale given according to (3) and (5) by $\frac{1}{|\gamma_{\rm dd}^{12}|}$ $\left(\frac{1}{|\gamma_{\rm dd}^{23}|}\right)$, c_1 and c_2 (c_3 and c_4) will thus acquire the same modulus, with equal or opposite sign. This means that the interference terms in (14) and (15) do *not* disappear; instead, they are built up to their maximum values in the course of the coherent evolution between two subsequent quantum jumps.

On the other hand, *all* spontaneous emissions which may take place have to be considered. The most probable decay is the decay on the fast $|1\rangle$-$|3\rangle$ transition, occurring on time scales

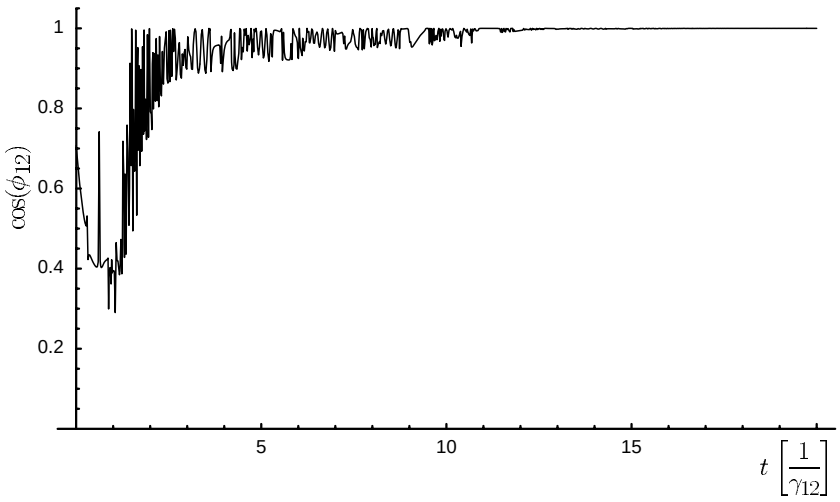

Fig. 3 – Evolution of the term $\cos(\phi_{12})$ as a function of time (in units of $(\gamma^{12})^{-1}$), obtained by Quantum Monte Carlo simulation. Parameters are $r = 0.3\lambda$, $\gamma^{23} = 20\,\gamma^{12}$, $\Omega_{\rm R} = 20\,\gamma^{13}$.

$\frac{1}{|\gamma^{13}|}$. As there is no preferred direction of photon emission, the phases have to be reset after each spontaneous decay $|1\rangle \rightarrow |3\rangle$ to a random value. In order to obtain the *averaged* transition rates P_{10} and P_{12}, we have therefore to integrate (14) and (15) over all possible phase relations ϕ_{12} and ϕ_{34}. In this case, in the limit (2), the interference terms in (14) and (15) vanish: resetting of the phase differences ϕ_{12} and ϕ_{34} takes place at a much faster rate than the change and build-up of the transition rates to their time-invariant (and extremal) value. Integrating (14) and (15) over all possible phase relations simply leads to

$$P_{10} \;=\; 2\gamma_{12}\left(|c_1|^2 + |c_2|^2\right)\,, \tag{19}$$

$$P_{12} \;=\; 2\gamma_{23}\left(|c_1|^2 + |c_2|^2 + |c_3|^2 + |c_4|^2\right)\,. \tag{20}$$

According to (19) and (20) no effect relating to the dipole-dipole interaction and depending on the ion-ion distance r remains. This result is in close analogy to the Quantum Zeno effect [16, 17], where the build-up of coherences in the course of the coherent evolution of the system is suppressed by resetting the state vector. As this effect is inherent in any 3-level system corresponding to the Λ-scheme of fig. 1 and satisfying (1) and (2), a master-equation treatment has to yield the same result [9]. This basic mechanism of the 3-level Λ-system might also be at the heart of more complex schemes and thus help to understand the contradicting outcomes of refs. [13] and [14].

Note that a different result is obtained when in addition the two ions are excited coherently on the $|3\rangle$-$|2\rangle$ transition. In this case oscillations of the transition rates P_{21}, P_{10} and P_{12} may appear in phase with $\gamma^{13}_{\rm dd}$ up to distances of $10\lambda_{13}$ (which may still be outside limit (1)) [10,11].

Conclusion. – In conclusion, we reinvestigated the quantum jump behaviour of two dipole-dipole interacting atoms placed in the vicinity of each other in the limit $\lambda_{12} \gg r \gg \lambda_{13}$. The internal level structure of the two atoms corresponds to the level scheme of fig. 1, where the condition $\gamma^{13} \gg \gamma^{12}, \gamma^{23}$ is imposed. This investigation is motivated by contradictory outcomes of recent experiments [8, 12–14] as well as by our own work with In^+ [21, 22]. For the analysis we employ a Quantum Monte Carlo wave function approach which apart from numerical simulations allows to obtain analytical results. These agree with the outcome

of a previous investigation [9] that for ion separations $r \gg \lambda_{13}$ no collective behaviour is observable even though $r \ll \lambda_{12}$. In contrast to the master-equation approach employed in [9], a clear understanding is obtained why in fact no cooperative behaviour is displayed. The principal result is that due to the rapid decay on the fast transition any dipole-dipole–induced entanglement between the two ions leading to cooperative behaviour is suppressed. This result can be shown to hold for any pump power —weak or strong— taking fully into account the real and imaginary parts of the dipole-dipole interaction, level shifts and coherent evolution among the internal states of the two ions. In particular, it turns out that it is not the strong saturation of the $|1\rangle$-$|3\rangle$ transition that is responsible for this result but the fast decay on the $|1\rangle$-$|3\rangle$ transition. This process therefore also occurs for weak pump power and results from the widely different time scales of the two natural lifetimes involved.

* * *

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