Simplified approach to double jumps for fluorescing dipole-dipole interacting atoms

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Abstract. A simplified scheme for the investigation of cooperative effects in the quantum jump statistics of small numbers of fluorescing atoms and ions in a trap is presented. It allows the analytic treatment of three dipole-dipole interacting four-level systems which model the relevant level scheme of $Ba⁺$ ions. For the latter, a huge rate of double and triple jumps was reported in a former experiment and the huge rate was attributed to the dipole-dipole interaction. Our theoretical results show that the effect of the dipole-dipole interaction on these rates is at most 5% and that for the parameter values of the experiment there is practically no effect. Consequently it seems that the dipole-dipole interaction can be ruled out as a possible explanation for the huge rates reported in the experiment.

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1 Introduction

The dipole-dipole interaction between atoms and molecules is of fundamental importance in nature as it gives rise to the all pervading van der Waals force. In physics, cooperative effects in the radiative behaviour of atoms due to their mutual dipole-dipole interaction have also attracted considerable interest in the literature [1], and they may play a role for possible quantum computers based on trapped ions or atoms. Atoms exhibiting macroscopic light and dark periods in their fluorescence may provide a sensitive test for such cooperative effects. Such macroscopic light and dark periods can occur in a multi-level system if the electron is essentially shelved in a metastable state, thereby causing the photon emission to cease [2]. Two such systems accordingly exhibit a dark period, a bright period of the same intensity as that of a single system, and a bright period of double intensity. Three systems exhibit an additional bright period of threefold intensity. The dipole-dipole interaction may now alter the statistics of these periods.

In an experiment with two and three $Ba⁺$ ions [3,4] a large number of double and triple jumps, i.e. jumps by two or three intensity steps within a short resolution time, had been observed, by far exceeding the number expected for independent atoms. Theoretically, the quantitative explanation of such large cooperative effects for distances of the order of ten wave lengths of the strong transition proved difficult [5–10]. On the other hand, experiments with different ions showed no observable cooperative effects [11], in particular none were seen for Hg^+ for a distance of about 15 wave lengths [12]. More recently, effects similar to reference [3] were found in an experiment with $Ca⁺$ ions [13], in contrast to another, comparable, experiment [14]. Neither were cooperative effects found experimentally in an extensive analysis of the quantum jump statistics of two trapped Sr^+ ions [15]. Skornia et al. [16] recently put forward a new proposal for observing the dipole-dipole interaction of two V systems.

For two V systems numerical [17] and analytical [18] investigations of the effect of the dipole-dipole interaction showed an increase of up to 30% in the double jump rate when compared to independent systems. However, the systems used in the experimental setups of references [3,12,19] were not V systems so that a direct comparison between theory and experiment was not possible. For this reason the present authors extended their investigation to two other systems [20], namely a D shaped system modeling the $Hg⁺$ ions used in reference [12] and a four-level system (see Fig. 1) modeling the $Ba⁺$ ions of references [3,4]. For two D systems cooperative effects in the same order of magnitude as for the V systems were found for ion-distances of a few wavelengths of the laser-driven transition. For larger distances practically no effects where found, in agreement with the experiments [12] and with the results of reference [21]. In contrast, only negligible effects for a wide range of ion-distances were found for

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Fig. 1. (a) Relevant level scheme of $Ba⁺$ [3, 4]. For the effective four level system the circled levels are merged to a single level. (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 $|2\rangle$.

two of the four level-systems. Although this result contradicts the findings of references [3,4] a direct quantitative comparison with the experiments was not possible since explicit experimental data were only provided for three $Ba⁺$ ions. For this reason three of the D and V systems were investigated in reference [22]. In comparison to two of either systems the cooperative effects found in this case are considerably higher, namely up to 170% deviation from the case of independent atoms. However, since the complexity increases dramatically for higher-level systems, this approach could not be applied to three of the four-level systems which we use to describe the situation of reference [3].

In the present paper a simplified approach for the calculation of the transition rate will be presented with which three four-level systems can now be treated analytically. This approach is valid for atoms with a level structure in which the transitions between the different intensity periods take place incoherently, i.e. via decay or via incoherent driving. The transition rates for three dipole-interacting four-level systems will be calculated. Cooperative effects for this system are found to be less than 5% and negligible for the experimental parameters of reference [3]. Consequently it seems that the dipole-dipole interaction can be ruled out as a possible explanation for the huge effects measured in the latter experiment.

Fig. 2. Three-level system in D configuration with fast transitions (solid lines) and slow transitions (dashed lines).

In Section 2 the Bloch equation approach is recapitulated. On this basis the new method is presented in Section 3 and applied to the four-level systems in Section 4. In Section 5 the possibility of a translation of this method to V system type level structures is discussed.

2 Bloch equation approach

The fluorescence, i.e. the stochastic sequence of photon emissions, of a system consisting of a number of atoms with macroscopic bright and dark periods can be described by a telegraph process. This process is characterized by the transition rates between the different intensity periods. In references [18,20,22] they were calculated for different model level systems and different numbers of atoms using a perturbation approach based on the Bloch equation of the corresponding systems. This approach will be illustrated in the following by applying it to the simple case of a single three level system in a D-type configuration as depicted in Figure 2.

The Bloch equations can be written in the compact form [23]

$$
\dot{\rho} = -\frac{i}{\hbar} \left[H_{\text{cond}} \rho - \rho H_{\text{cond}}^{\dagger} \right] + \mathscr{R}(\rho) \tag{1}
$$

where H_{cond} is the conditional Hamiltonian of the quantum jump approach [24], for this system given by

$$
H_{\text{cond}} = \frac{\hbar}{2i} \left[(A_2 + A_3)|3\rangle\langle 3| + A_1|2\rangle\langle 2| \right] + \frac{\hbar \Omega_3}{2} \left[|1\rangle\langle 3| + |3\rangle\langle 1| \right] \tag{2}
$$

and $\mathcal{R}(\rho)$ is the the reset state,

$$
\mathscr{R}(\rho) = A_1|1\rangle\langle 2|\rho|2\rangle\langle 1| + A_2|2\rangle\langle 3|\rho|3\rangle\langle 2| + A_3|1\rangle\langle 3|\rho|3\rangle\langle 1|.\tag{3}
$$

The Rabi frequency Ω_3 and the Einstein coefficients A_1 , A_2 , A_3 are subject to the condition

$$
\Omega_3, A_3 \gg A_1, A_2. \tag{4}
$$

A detuning of the laser has been neglected for simplicity. If the small optical parameters A_1 , A_2 are neglected the system splits into independent subspaces. They are given by

$$
\mathcal{S}_0 = \{|2\rangle\}, \qquad \mathcal{S}_1 = \{|1\rangle, |3\rangle\}. \tag{5}
$$

These subspaces \mathscr{S}_i can be associated with the periods of intensity I_i in the sense that in a period I_i the system is mostly in the subspace \mathscr{S}_i . Taking a state $\rho_{0,i}$ in one of the subspaces \mathscr{S}_i at a time t_0 we calculate the state at a time $t_0 + \Delta t$ later in perturbation theory with respect to the small parameters. The time interval Δt used here should be long compared to the mean time between the emission of two photons but short compared to the length of the intensity periods,

$$
A_3^{-1}, \Omega_3^{-1} \ll \Delta t \ll A_1^{-1}, A_2^{-1}.
$$
 (6)

For the calculation the Bloch equation is written in a Liouvillean form,

$$
\dot{\rho} = \mathscr{L}\rho = \{\mathscr{L}_0(A_3, \Omega_3) + \mathscr{L}_1(A_1, A_2)\}\rho . \tag{7}
$$

The density matrix at time $t_0 + \Delta t$ is then given by [18]

$$
\rho(t_0 + \Delta t, \rho_{0,i}) = \rho_{ss,i} + \int_0^{\Delta t} d\tau \, e^{\mathcal{L}_0 \tau} \mathcal{L}_1 \rho_{ss,i} , \quad (8)
$$

where $\rho_{ss,i}$ is the quasi-steady state in subsystem \mathscr{S}_i , i.e. a steady state of *L*0. One can write

$$
\mathcal{L}_1 \rho_{ss,i} = \sum_{j=0}^{1} \alpha_{ij} \rho_{ss,j} \Delta t + \tilde{\rho},\tag{9}
$$

with $\tilde{\rho}$ containing the contributions from the eigenstates of \mathcal{L}_0 for non-zero eigenvalues. This leads to [20]

$$
\rho(t + \Delta t, \rho_i) = \rho_{ss,i} + \sum_{j=0}^{1} \alpha_{ij} \rho_{ss,j} \Delta t + (\epsilon - \mathcal{L}_0)^{-1} \tilde{\rho}. (10)
$$

The last term can be neglected and the coefficient α_{ij} can therefore be interpreted as transition rate p_{ij} from intensity period I_i to period I_j . They can be calculated by means of the dual eigenstates for eigenvalue 0 of \mathcal{L}_0 [20]. For a single D system the quasi-steady states are given by

$$
\rho_{\rm ss,0} = |2\rangle\langle 2|,\tag{11}
$$
\n
$$
\rho_{\rm ss,1} = \frac{1}{A_3^2 + 2\Omega_3^2} \left[(A_3^2 + \Omega_3^2)|1\rangle\langle 1| + \Omega_3^2|3\rangle\langle 3| + iA_3\Omega_3|1\rangle\langle 3| - iA_3\Omega_3|3\rangle\langle 1| \right]
$$
\n
$$
(12)
$$

for the dark and the light period respectively. The corresponding dual states are

$$
\rho_{\rm ss}^0 = |2\rangle\langle 2|
$$
, and $\rho_{\rm ss}^1 = |1\rangle\langle 1| + |3\rangle\langle 3|$. (13)

From (11) and (12) one finds

$$
\mathcal{L}_1 \rho_{\text{ss},0} = -A_1 |2\rangle\langle 2| + A_1 |1\rangle\langle 1| \tag{14a}
$$

and

$$
\mathcal{L}_{1}\rho_{ss,1} = -A_2 \frac{\Omega_3^2}{A_3^2 + 2\Omega_3^2} |3\rangle\langle 3| + A_2 \frac{\Omega_3^2}{A_3^2 + 2\Omega_3^2} |2\rangle\langle 2| -\frac{A_2}{2} \frac{A_3 \Omega_3}{A_3^2 + 2\Omega_3^2} (|1\rangle\langle 3| - |3\rangle\langle 1|). \tag{14b}
$$

The transition rates are then calculated from

$$
p_{ij} = \alpha_{ij} = \text{Tr}(\rho_{\text{ss}}^{j\dagger} \mathcal{L}_{1} \rho_{\text{ss},i})
$$
\n(15)

as

$$
p_{01} = \alpha_{01} = A_1 \tag{16}
$$

and

$$
p_{10} = \alpha_{10} = \frac{A_2 \Omega_3^2}{A_3^2 + 2\Omega_3^2},\tag{17}
$$

in agreement with the direct calculation of the transition rates via the quantum jump approach.

3 New simplified approach

Due to the increased number of levels involved, a calculation of the transition rates for three dipole-interacting four-level systems would, although in principal feasible with the methods introduced above, be even more laborious than for three three-level systems. It is, however, possible to read off the transition rates without having to carry out the full calculation. One only needs the quasi-steady states of the corresponding subsystems. In following this simpler approach will be presented.

By looking at equation (14) one realizes that the last step in the calculation, namely the projection onto the dual eigenstates, although formally more satisfactory, was actually not necessary in order to gain the final result. The transition rates are already present as prefactors for some of the density matrix elements. In fact, \mathscr{L}_1 can be interpreted as a transition operator. Applying it to some state of the system yields the density matrix elements which are modified by the weak decays multiplied by the corresponding decay rates. They are positive for density matrix element which gain population and negative for those which loose population due to the decay. In the case in which one started with $\rho_{ss,0} = |2\rangle\langle 2|$ one therefore has a term $-A_1|2\rangle\langle 2|$, which accounts for the loss of population of level $|2\rangle$, and a term $A_1|1\rangle\langle 1|$ for the corresponding gain of population in the ground state. When starting with $\rho_{ss,1}$ the Einstein coefficient A_2 for the decay from $|3\rangle$ to $|2\rangle$ has an additional factor $\Omega_3^2/(\Lambda_3^2 + 2\Omega_3^2)$ for the quasi-steady state population of level $|3\rangle$. The last two terms in equation (14b) are due to the decay of the coherences between $|1\rangle$ and $|3\rangle$.

From these considerations one is lead to a simple scheme for the evaluation of the transition rates. First one has to identify the different independent subspaces for vanishing weak decay rates and calculate the quasi-steady states in these subspaces as in the above Bloch equation approach. For a single D system these are the states $\rho_{ss,0}$ and $\rho_{ss,1}$ for the subsystems associated with the dark and bright period, respectively. By looking at the level scheme one can then determine the possible decay channels between the subsystems.In the present case this is a decay by A_2 from $|3\rangle$ to $|2\rangle$ and a decay by A_1 from $|2\rangle$ to $|1\rangle$.

Fig. 3. Level configuration of two D systems in the Dicke
hasis. Transitions with rate $A_0 + \text{Re} C_0$ (dotted arrows) and basis. Transitions with rate $A_2 \pm \text{Re } C_2$ (dotted arrows) and transitions with rate $A_1 \pm \text{Re } C_1$ (dashed arrows). Fast transitions (with $A_3 \pm \text{Re } C_3$) and line shifts due to detuning and to Im ^C*ⁱ* are omitted.

The transition rates are then given by these decay rates multiplied with the steady state population of the decaying level.

Physically this is quite intuitive: the transition rates are given by the corresponding decay rates multiplied by the mean occupation probabilities of the levels involved.

The question is now if this approach can be extended to more complicated systems, especially to dipole-interacting D systems and to the four-level system for the description of $Ba⁺$. This is indeed possible. For two dipole-interacting D systems for example the possible decays can be read off Figure 3 which shows the level scheme in the Dicke basis given by

$$
|g\rangle = |1\rangle|1\rangle, \quad |e_2\rangle = |2\rangle|2\rangle, \quad |e_3\rangle = |3\rangle|3\rangle
$$

\n
$$
|s_{ij}\rangle = \frac{1}{\sqrt{2}} (|i\rangle|j\rangle + |j\rangle|i\rangle),
$$

\n
$$
|a_{ij}\rangle = \frac{1}{\sqrt{2}} (|i\rangle|j\rangle - |j\rangle|i\rangle).
$$
 (18)

The easiest case is the transition rate p_{01} for a transition from a dark period to a period of intensity I_1 . Here the relevant transitions are from $|e_2\rangle$ to $|s_{12}\rangle$ and $|a_{12}\rangle$. The corresponding decay rates are $A_1 + \text{Re } C_1$ and $A_1 - \text{Re } C_1$, respectively, with the dipole-dipole coupling parameters C_i given explicitly in reference [20]. The quasi-steady state population of $|e_2\rangle$ is unity, so the transition rate is $p_{01} =$ $2A_1$, in agreement with the result of reference [20]. The other transition rates are a bit more complicated. For p_{10} one has to take into account the decays from $|s_{23}\rangle$ and $|a_{23}\rangle$ to $|e_2\rangle$, for p_{12} the decays from $|s_{23}\rangle$ and $|a_{23}\rangle$ to $|s_{13}\rangle$ and from $|s_{12}\rangle$ and $|a_{12}\rangle$ to $|g\rangle$, and for p_{21} the decays from $|e_3\rangle$ to $|s_{23}\rangle$ and $|a_{23}\rangle$ and from $|s_{13}\rangle$ and $|a_{13}\rangle$ to $|s_{12}\rangle$ and $|a_{12}\rangle$. Multiplying for each decay the decay rate by the steady state population of the initial level and adding up the different contributions then yields the same results for the transition rates as obtained by the Bloch equation approach in reference [20]. The same is also true for three dipole-interacting D systems [22].

4 Three dipole-interacting four-level systems

An application of the simplified method to the four-level system describing $Ba⁺$ is also possible. As depicted in Figure 1 the transition from a bright to a dark period is a two step process for this system, first an excitation to level $|4\rangle$ by incoherent light with the rate W and then a decay to level $|2\rangle$ with the Einstein coefficient A_2 . Instead of a single Einstein coefficient one therefore has to use the product of the incoherent transition rate W with the branching ratio $A_2/(A_2+A_4)$ for a decay from state $|4\rangle$ to state $|2\rangle$ for this transition. Then everything works as in the case of the D systems and one confirms the results for a single four-level system already known from the Bloch equation approach [20].

Consequently it is also possible to obtain the transition rates for three four-level systems which would be rather involved to do with the Bloch equation approach. The Bloch equations can be written in the compact form

$$
\dot{\rho} = -\frac{i}{\hbar} \Big[H_{\text{cond}} \rho - \rho H_{\text{cond}}^{\dagger} \Big] + \mathcal{R}_W(\rho) + \mathcal{R}(\rho) \quad (19)
$$

$$
\equiv \{ \mathcal{L}_0 + \mathcal{L}_1(A_1, W) \} \rho,
$$

where $\mathscr{R}_W(\rho)$ describes the incoherent driving as in reference [25] and is given by

$$
\mathcal{R}_W(\rho) = W \sum_{i=1}^3 \left(S_{i4}^+ \rho S_{i4}^- + S_{i4}^- \rho S_{i4}^+ \right) , \qquad (20)
$$

with

$$
\begin{aligned} S_{i1}^+ &= |2\rangle_{ii}\langle 1|, \qquad S_{i2}^+ = |4\rangle_{ii}\langle 2|, \qquad S_{i3}^+ = |3\rangle_{ii}\langle 1| \\ S_{i4}^+ &= |4\rangle_{ii}\langle 1|, \qquad & \text{and} \qquad & S_{ij}^- = S_{ij}^{+\dagger} \ . \end{aligned}
$$

The conditional Hamiltonian, without detuning, and the reset state in this case are given by

$$
H_{\text{cond}} = \sum_{i=1}^{3} \sum_{j=1}^{4} \frac{\hbar}{2i} A_j S_{ij}^+ S_{ij}^- + \sum_{i=1}^{3} \frac{\hbar}{2} \left[\Omega_3 S_{i3}^- + \text{h.c.} \right] + \sum_{\substack{k,l=1 \ k < l}}^{3} \sum_{j=1}^{4} \frac{\hbar}{2i} C_{kl}^{(j)} \left(S_{kj}^+ S_{lj}^- + S_{lj}^+ S_{kj}^- \right) \tag{21}
$$

and

$$
\mathcal{R}(\rho) = \sum_{i=1}^{3} \sum_{j=1}^{4} A_j S_{ij}^{-} \rho S_{ij}^{+} + \sum_{\substack{k,l=1 \ k < l}}^{3} \sum_{j=1}^{4} \text{Re } C_{kl}^{(j)} \left(S_{kj}^{-} \rho S_{lj}^{+} + S_{lj}^{-} \rho S_{kj}^{+} \right), \quad (22)
$$

where

$$
C_{kl}^{(j)} = \frac{3A_j}{2} e^{i a_{kl}^{(j)}} \left[\frac{1}{i a_{kl}^{(j)}} (1 - \cos^2 \theta_{kl}) + \left(\frac{1}{a_{kl}^{(j)2}} - \frac{1}{i a_{kl}^{(j)3}} \right) (1 - 3 \cos^2 \theta_{kl}) \right]
$$
(23)

is the coupling parameter which describes the dipoledipole interaction between atom k and atom l for the transition connected with the Einstein coefficient A_j , with θ_{kl} being the angle between the dipole moments and the line connecting the atoms. The dimensionless parameter $a_{kl}^{(j)} = 2\pi r_{kl}/\lambda_j$ is given by the inter-atomic distance r_{kl} multiplied by the wave number $2\pi/\lambda_j$ of this transition. In order to get a maximal effect of the dipole-dipole interaction we assume as in [22] that the atoms form an equilateral triangle (i.e. $r_{kl} = r$) and that $\theta_{kl} = \pi/2$. Then $C_{kl}^{(j)}$ becomes the C_j of reference [20].

The quasi-steady states are already known from the calculations for three three-level systems. As in reference [22] one can use a symmetrized basis analogous to the Dicke basis for two atoms. This leads to the states

$$
|s_{ijk}\rangle = \frac{1}{\sqrt{6}}(|i\rangle|j\rangle|k\rangle + |j\rangle|k\rangle|i\rangle + |k\rangle|i\rangle|j\rangle + |i\rangle|k\rangle|j\rangle + |j\rangle|i\rangle|k\rangle + |k\rangle|j\rangle|i\rangle), \qquad (24a)
$$

$$
|a_{ijk}\rangle = \frac{1}{\sqrt{6}}(|i\rangle|j\rangle|k\rangle + |j\rangle|k\rangle|i\rangle + |k\rangle|i\rangle|j\rangle
$$

$$
- |i\rangle|k\rangle|j\rangle - |j\rangle|i\rangle|k\rangle - |k\rangle|j\rangle|i\rangle), \qquad (24b)
$$

$$
|b_{ijk}\rangle = \frac{1}{\sqrt{12}}(2|i\rangle|j\rangle|k\rangle - |j\rangle|k\rangle|i\rangle - |k\rangle|i\rangle|j\rangle
$$

$$
+2|i\rangle|k\rangle|j\rangle-|j\rangle|i\rangle|k\rangle-|k\rangle|j\rangle|i\rangle), \qquad (24c)
$$

$$
|c_{ijk}\rangle = \frac{1}{2} (|j\rangle |k\rangle |i\rangle - |k\rangle |i\rangle |j\rangle - |j\rangle |i\rangle |k\rangle + |k\rangle |j\rangle |i\rangle), \qquad (24d)
$$

$$
|d_{ijk}\rangle = \frac{1}{\sqrt{12}} (2|i\rangle|j\rangle|k\rangle - |j\rangle|k\rangle|i\rangle - |k\rangle|i\rangle|j\rangle
$$

- 2|i\rangle|k\rangle|j\rangle + |j\rangle|i\rangle|k\rangle + |k\rangle|j\rangle|i\rangle), (24e)

$$
|e_{ijk}\rangle = \frac{1}{2} (|j\rangle|k\rangle|i\rangle - |k\rangle|i\rangle|j\rangle + |j\rangle|i\rangle|k\rangle - |k\rangle|j\rangle|i\rangle), \qquad (24f)
$$

 $i < j < k$; $i, j, k = 1, \ldots, 4$, in the case where all three atoms are in different states. For the remaining states one gets for $i, j = 1, \ldots, 4, i \neq j$,

$$
|s_{ijj}\rangle = \frac{1}{\sqrt{3}} (|i\rangle|j\rangle|j\rangle + |j\rangle|j\rangle|i\rangle + |j\rangle|i\rangle|j\rangle)
$$
 (25a)

$$
|b_{ijj}\rangle = \frac{1}{\sqrt{6}} (2|i\rangle|j\rangle|j\rangle - |j\rangle|j\rangle|i\rangle - |j\rangle|i\rangle|j\rangle)
$$
 (25b)

$$
|c_{ijj}\rangle = \frac{1}{\sqrt{2}} (|j\rangle|j\rangle|i\rangle - |j\rangle|i\rangle|j\rangle)
$$
 (25c)

if two atoms are in the same state and

$$
|g\rangle = |1\rangle|1\rangle|1\rangle, \quad |e_i\rangle = |i\rangle|i\rangle|i\rangle \quad \text{for} \quad i = 2, 3, 4 \quad (26)
$$

if all three atoms are in the same state. The quasi-steady states for intensity periods I_0 to I_2 are, by symmetry, given by

$$
\rho_{\rm ss,0} = |e_2\rangle\langle e_2|
$$
\n
$$
\rho_{\rm ss,1} = \frac{1}{3} \{ \rho_{\rm ss}^{1D} \otimes |2\rangle_{22} \langle 2| \otimes |2\rangle_{33} \langle 2|
$$
\n
$$
+ |2\rangle_{11} \langle 2| \otimes \rho_{\rm ss}^{1D} \otimes |2\rangle_{33} \langle 2| + |2\rangle_{11} \langle 2| \otimes |2\rangle_{22} \langle 2| \otimes \rho_{\rm ss}^{1D} \}
$$
\n(27b)

$$
\rho_{\text{ss},2} = \frac{1}{3} \sum_{i=1}^{3} \rho_{\text{ss},2}^{\text{2D}} \otimes |2\rangle_{ii} \langle 2|,\tag{27c}
$$

where ρ_{ss}^{1D} is the quasi-steady state of one D system in the $\{|1\rangle, |3\rangle\}$ subspace and $\rho_{ss,2}^{2D}$ is the quasi-steady state in the subspace corresponding to double intensity of two D systems. The state $\rho_{ss,3}$ is rather complicated. Therefore only the populations of the relevant levels will be given, i.e.

$$
\langle g|\rho_{\rm ss,3}|g\rangle = \frac{1}{N} \Big[\{ (A_3^2 + \Omega_3^2) \big[(A_3^2 + \Omega_3^2)^2 + 3A_3^2B \big] + 2A_3 \big[|C_3|^2 |A_3 + C_3|^2 + B^2 \big] \} \Big], \tag{28a}
$$

$$
\langle s_{113} | \rho_{\text{ss},3} | s_{113} \rangle = \frac{\Omega_3^2}{N} \left[(A_3^2 + \Omega_3^2)(3A_3^2 + \Omega_3^2) + 3A_3^2B \right] \tag{28b}
$$

$$
\langle b_{113} | \rho_{\text{ss},3} | b_{113} \rangle = \langle c_{113} | \rho_{\text{ss},3} | c_{113} \rangle = \frac{\Omega_3^4}{N} (A_3^2 + \Omega_3^2) \tag{28c}
$$

$$
\langle s_{133} | \rho_{ss,3} | s_{133} \rangle = \frac{\Omega_3^4}{N} (3A_3^2 + \Omega_3^2)
$$
 (28d)

$$
\langle e_3|\rho_{\text{ss},3}|e_3\rangle = \langle b_{133}|\rho_{\text{ss},3}|b_{133}\rangle = \langle c_{133}|\rho_{\text{ss},3}|c_{133}\rangle = \frac{\Omega_3^6}{N}
$$
\n(28e)

with

$$
N = \{(A_3^2 + 2\Omega_3^2) [(A_3^2 + 2\Omega_3^2)^2 + 3A_3^2B] + 2A_3 [|C_3|^2 |A_3 + C_3|^2 + B^2] \}
$$

and

$$
B = |C_3|^2 + 2A_3 \text{Re}\, C_3.
$$

Now the procedure is the same as described in the previous section for two D systems and one obtains

$$
p_{01} = 3A_1 \qquad p_{12} = 2A_1 \qquad p_{23} = A_1 \qquad (29a)
$$

and

$$
p_{10} = \frac{A_2 W (A_3^2 + \Omega_3^2)}{(A_2 + A_4)[A_3^2 + 2\Omega_3^2]}
$$
(29b)

$$
p_{21} = \frac{2A_2 W}{A_2 + A_4} \left[\frac{A_3^2 + \Omega_3^2}{A_3^2 + 2\Omega_3^2} + 2 \operatorname{Re} C_3 \frac{A_3^3 \Omega_3^2}{[A_3^2 + 2\Omega_3^2]^3} \right]
$$
(29c)

$$
p_{32} = \frac{3A_2 W}{A_2 + A_4} \left[\frac{A_3^2 + \Omega_3^2}{A_3^2 + 2\Omega_3^2} + 4 \operatorname{Re} C_3 \frac{A_3^3 \Omega_3^2}{[A_3^2 + 2\Omega_3^2]^3} \right]
$$
(29d)

Fig. 4. Double jump rate n_{DJ} and for three dipole-interacting four-level systems with experimental parameter values of reference [19]. Dotted line: independent systems. Cooperative effects are less than 1 for distances larger than λ_3 .

as transition rates up to first order in C_3 . The exact results including detuning are given in the appendix. The approximations to first order in C_3 have the same structure as for three dipole-interacting three-level systems given in reference [22]. Basically this means an increase of cooperative effects by a factor of two compared to two atoms. In terms of these transition rates the double and triple jump rate, i.e. the rate of two or three subsequent jumps within a short time window T_W , are then given by [20]

$$
n_{\rm DJ} = 2 \frac{p_{01} p_{21} p_{32} (p_{01} + p_{12})}{p_{21} p_{32} (p_{01} + p_{10}) + p_{01} p_{12} (p_{23} + p_{32})} T_W \quad (30)
$$

and

$$
n_{\text{TJ}} = 2 \frac{p_{01}p_{10}p_{12}p_{21}p_{23}p_{32}}{p_{21}p_{32}(p_{01} + p_{10}) + p_{01}p_{12}(p_{23} + p_{32})} T_W^2 \tag{31}
$$

In Figure 4 a plot of n_{DJ} for the experimental parameter values of reference [19] is shown. The effects of the dipoledipole interaction are negligibly small in particular for experimental distances of about ten times the wavelength λ_3 of the strong transition. Without detuning Δ_3 , maximal cooperative effects are obtained for $\Omega_3 = \frac{1}{2}\sqrt{\sqrt{5}-1}A_3$. This case is shown in Figure 5 for the triple jump rate n_{TJ} . For inter-atomic distances larger than one wavelength λ_3 of the strong transition cooperative effects are less than 5% and again rapidly decreasing for larger distances. For non-zero detuning the maximally achievable effects have about the same value. Also one has to bear in mind that, as in reference [22], this result has to be seen as an upper limit for all possible configurations in the trap. Large cooperative effects, i.e. enhancements of the double and triple jump rate by several orders of magnitude, can therefore not be explained by the dipole-dipole interaction. Furthermore one sees that the first order results of equation (29) are a very good approximation to the exact transition rates given in the appendix.

Fig. 5. Triple jump rate n_{TJ} for three dipole-interacting four-level systems. Parameter values as in Figure 4 except for $\Delta_3 = 0$ and $\Omega_3 = \frac{1}{2}\sqrt{\sqrt{5}-1}A_3$ for maximal effects. Dotted
line: independent systems, dashed line: up to first order. Coline: independent systems, dashed line: up to first order. Cooperative effects are less than 5% for distances larger than λ_3 .

5 V system and similar level schemes

From the previous results the question arises whether the method presented here is also applicable to level systems like the V system, i.e. systems in which the transition between different light and dark periods results from a coherent excitation. It turns out that for these systems the situation is much more complicated. For a single V system for example, $\mathscr{L}_1(\Omega_2)$ contains coherences between the ground state $|1\rangle$ and the metastable state $|2\rangle$. Therefore $\mathscr{L}_1\rho_i$ has no component in the subspace of eigenstates of \mathscr{L}_0 for eigenvalue zero and the state at time $t_0 + \Delta t$ in the Bloch equation approach is thus given by [18]

$$
\rho(t_0 + \Delta t, \rho_{0,i}) = \rho_{\text{ss},i} + (\epsilon - \mathcal{L}_0)^{-1} \mathcal{L}_1 \rho_{\text{ss},i}.
$$
 (32)

An explicit evaluation of this expression for a single V system starting with ρ_1 not only leads to terms proportional to the quasi-steady state population of the ground state but also to terms proportional to the quasi-steady state coherence between ground state and excited state.

The situation gets even more involved for dipoleinteracting V systems. Here the term $(\epsilon - \mathcal{L}_0)^{-1}$ gives rise to additional factors which depend in a very complicated way on C_3 . This is in contrast to the D and the four-level system, for which the C_3 dependence in the transition rates is solely due to the C_3 dependence of the quasi-steady states. The physical reason for this is that the efficiency of the laser driving is influenced by the dipole interaction, for example via additional detunings. Therefore the mechanism of jumps in the light intensity based on laser driven transitions is much more complex than for jumps based on spontaneous decay and incoherent driving so that the method outlined above is applicable only in the latter case.

6 Conclusions

In this paper we have presented a simplified approach for the calculation of the transition rates between periods of

$$
p_{10} = \frac{A_2 W (A_3^2 + \Omega_3^2 + 4 \Delta_3^2)}{(A_2 + A_4)[A_3^2 + 2 \Omega_3^2 + 4 \Delta_3^2]}
$$
(33a)

$$
p_{21}=\frac{2A_2 W}{A_2+A_4}\frac{(A_3^2+ \Omega_3^2+4 \Delta_3^2)(A_3^2+2 \Omega_3^2+4 \Delta_3^2)+ (A_3^2+4 \Delta_3^2)(|C_3|^2+2 A_3 \mathrm{Re}\, C_3-4 \Delta_3 \mathrm{Im}\, C_3)}{(A_3^2+2 \Omega_3^2+4 \Delta_3^2)^2+(A_3^2+4 \Delta_3^2)(|C_3|^2+2 A_3 \mathrm{Re}\, C_3-4 \Delta_3 \mathrm{Im}\, C_3)}
$$

$$
= \frac{2A_2W}{A_2 + A_4} \left[\frac{A_3^2 + \Omega_3^2 + 4\Delta_3^2}{A_3^2 + 2\Omega_3^2 + 4\Delta_3^2} + 2\operatorname{Re}C_3 \frac{A_3 \Omega_3^2 (A_3^2 + 4\Delta_3^2)}{[A_3^2 + 2\Omega_3^2 + 4\Delta_3^2]^3} - 4\operatorname{Im}C_3 \frac{\Delta_3 \Omega_3^2 (A_3^2 + 4\Delta_3^2)}{[A_3^2 + 2\Omega_3^2 + 4\Delta_3^2]^3} \right] + \mathcal{O}(C_3^2). \tag{33b}
$$

$$
p_{32} = \frac{3A_2W}{A_2 + A_4}
$$

\n
$$
\times \frac{(A_3^2 + B_3^2 + 4A_3^2)[(A_3^2 + 2B_3^2 + 4A_3^2)^2 + 3(A_3^2 + 4A_3^2)B] + 2(A_3^2 + 4A_3^2)[[C_3]^2|A_3 - 2iA_3 + C_3]^2 + B(B_3^2 + B_3^2)}{(A_3^2 + 2B_3^2 + 4A_3^2)[(A_3^2 + 2B_3^2 + 4A_3^2)^2 + 3(A_3^2 + 4A_3^2)B] + 2(A_3^2 + 4A_3^2)[[C_3]^2|A_3 - 2iA_3 + C_3]^2 + B^2]}
$$

\n
$$
= \frac{3A_2W}{A_2 + A_4} \left[\frac{A_3^2 + B_3^2 + 4A_3^2}{A_3^2 + 2B_3^2 + 4A_3^2} + 4 \text{Re } C_3 \frac{A_3B_3^2(A_3^2 + 4A_3^2)}{[A_3^2 + 2B_3^2 + 4A_3^2]^3} - 8 \text{Im } C_3 \frac{A_3B_3^2(A_3^2 + 4A_3^2)}{[A_3^2 + 2B_3^2 + 4A_3^2]^3} \right] + \mathcal{O}(C_3^2)
$$
(33c)

different intensity of a system of dipole-dipole interacting atoms which show macroscopic quantum jumps in their fluorescence. This method works for atoms with level configurations in which the transition between the different intensity periods is based on incoherent processes. Results previously obtained with other methods are recovered by the new approach.

In addition, the new method has allowed the calculation of the transition rates for three interacting four-level systems modeling the the relevant level structure of Ba⁺ ions. This allows a direct comparison with the experiment of reference [19]. This experiment reported an enhancement of the double and triple jump rate by several orders of magnitude and this was explained through cooperative effects due to the dipole-dipole interaction between the ions. With the present results it is seen that this cannot be the explanation for the reported enhancement. Cooperative effects can indeed be found for this system but they are much smaller, namely only maximal 5% of the values for independent atoms. For the parameter values of the experiment they are practically absent.

Appendix A: Exact transition rates including detuning

As mentioned above the transition rates between the different intensity periods can be calculated exactly in C_3 and with inclusion of a possible detuning of the laser Δ_3 with respect to the corresponding atomic transition. The result for the downward rates is

see equations (33a–33c) above

with $B = |C_3|^2 + 2A_3 \text{Re } C_3 - 4\Delta_3 \text{Im } C_3$. The upward rates of equation (29a) are already the exact results since they are independent of C_3 and Δ_3 .

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