The intensity correlation function of “blinking” quantum systems

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Abstract. Explicit expressions are determined for the photon correlation function of “blinking” quantum systems, i.e. systems with different types of fluorescent periods. These expressions can be used for a fit to experimental data and for obtaining system parameters therefrom. For two dipole-dipole interacting \(V\) systems the dependence on the dipole coupling constant is explicitly given and shown to be particularly pronounced if the strong driving is reduced. We propose to use this for an experimental verification of the dipole-dipole interaction.

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1. Introduction

For fluorescing quantum systems one of the most important statistical quantities is given by the intensity correlation function, \(g(\tau)\), for photon counts \([1]\). Its behavior for small times \(\tau\) can indicate more classical or more quantum behavior and bunching or anti-bunching, depending on whether \(g(0) > 1\) or \(g(0) < 1\). Early investigations of the intensity correlation function of single two level systems \([2, 3, 4]\) led to the observation of nonclassical light \([5, 6]\). The intensity correlation function can also exhibit Rabi oscillations, and for \(V\) systems with metastable state it contains indications of light and dark periods (“blinking”) in the fluorescence \([7]\).

The importance of correlation functions partially stems from the relative ease with which they can be experimentally determined, partially due to the fact that the efficiency of the photon detector does not enter. Correlation functions were determined for single ions in a trap in experiments which were mainly carried out for the detection of quantum jumps in the fluorescence \([8, 9, 10, 11]\). In the last decade the same was achieved for single fluorescent molecules \([12, 13, 14]\). A theoretical determination of \(g(\tau)\) was obtained in closed form for three level systems \([15, 16, 17]\) and \(g(0)\) was calculated for two dipole-dipole interacting two-level systems \([18, 19, 20]\), but for more complicated multi-level systems it is usually done by the numerical solution of Bloch equations.
Because the intensity correlation function contains information on the system parameters, e.g. atomic constants like Einstein coefficients, these parameters could be obtained in principle from an experimentally determined $g(\tau)$ by fitting numerically calculated curves to the data. However, in general such a fit procedure needs many numerical runs and tends to be highly sensitive to experimental and numerical errors. With an algebraic expression a fit would be much easier and more reliable. Moreover, if algebraic expression were known for the intensity correlation function for multi-level systems one could study its behavior without recourse to many numerical runs for different parameter values. In the present paper such algebraic expressions of the correlation function will be given for fluorescing quantum systems with light and dark periods. As a generalization of a result for $V$ systems in [7], it is deduced from this algebraic expression that the correlation function for “blinking” systems with dark periods shows a “hump” for values of $\tau$ larger than the correlation times of the individual intensity periods. It is also pointed out that the existence of extended dark periods may considerably enhance the amplitude of Bloch oscillations.

As an application we study two dipole-dipole interacting $V$ systems and determine an algebraic expression for the corresponding intensity correlation function $g(\tau)$. This expression is used to study the behavior of $g(\tau)$ with respect to the strength of the dipole-dipole interaction. It is shown that the dipole-dipole interaction has an effect for small $\tau$ which is particularly pronounced for small values of the strong driving. We suggest to use this effect to experimentally verify the dipole-dipole interaction for atomic distances of a few wavelength of the strong transitions.

2. Algebraic expressions for intensity correlation functions

The temporal intensity correlation function for photon counts is defined as follows. In the steady state, let $G(\tau)$ be the joint probability density for detecting a photon both at time 0 and $\tau$, and let $I_{SS}$ denote the steady-state intensity (counts per unit time). Then $g(\tau)$ is defined as

$$g(\tau) = G(\tau)/I_{SS}^2.$$  \hspace{1cm} (1)

We consider now a fluorescing system with $n$ periods of different intensity $I_i$ and mean duration $T_i, i = 0, \ldots, n - 1$. A particular period usually corresponds to transitions within a simpler subsystem which is easier to treat than the complete system. In the following we therefore assume that within a given period $i$ the corresponding intensity correlation function $g_i(\tau)$ is known.

The intensity correlation function $g(\tau)$ of the larger system can then be determined as follows. Let $P_i$ be the probability for the occurrence of period $i$ and let $P_{ij}(\tau)$ be the probability to have period $j$ at time $\tau$ provided one had period $i$ at $\tau = 0$. Now, if at time 0 the fluorescence is in period $i$, then, for $\tau \ll T_i$, one still is in period $i$ with high probability. Therefore, in this case, the joint probability density for detecting a photon at both times, 0 and $\tau$, is $\sum_i P_i G_i(\tau)$ where $G_i(\tau)$ is the joint probability density for
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period $i$. Since for small $\tau$ one has $P_{ii}(\tau) \simeq 1$ this can be replaced by
\[ \sum_i P_i P_{ii}(\tau) G_i(\tau), \quad \tau \ll T_i. \tag{2} \]

For larger $\tau$ one may end up in a period different from that at time 0. If $\tau$ is larger than the correlation times associated with $g_i(\tau)$ then, for initial period $i$ and final period $j$, the joint detection probability is $I_i P_{ij}(\tau) I_j$. Since for such $\tau$’s one has $G_j(\tau) = I_j^2$ one can write the complete joint detection probability as
\[ G(\tau) = \sum_{ij} P_i I_i P_{ij}(\tau) G_j(\tau)/I_j. \tag{3} \]

This includes equation (2) as a special case since for $\tau$ small one has $P_{ij}(\tau) \simeq 0$ for $i \neq j$.

In this general formula one still has to express $P_i$ and $P_{ij}(\tau)$ in terms of the transition rates $p_{ij}$ from period $i$ to period $j$. These transition rates themselves can be expressed by Einstein coefficients and Rabi frequencies. One has, in particular, $T_i = 1/\sum_{k \neq i} p_{ik}$. The $P_{ij}(\tau)$ are easily seen to obey rate equations, e.g.
\[ \dot{P}_{11}(\tau) = \left( -\sum_k p_{1k} \right) P_{11}(\tau) + p_{21} P_{12}(\tau) + \cdots + p_{n1} P_{1n}(\tau). \tag{4} \]

In general, with the matrix $B = (B_{ij})$,
\[ B_{ij} = p_{ij} - \delta_{ij} \sum_k p_{ik}, \tag{5} \]
and the matrix
\[ P(\tau) = \left( P_{ij}(\tau) \right) \tag{6} \]
one has
\[ \dot{P} = PB, \tag{7} \]
with the initial condition $P_{ij}(0) = \delta_{ij}$, or
\[ P(0) = I. \tag{8} \]

The solution of equation (7) with this initial condition can be written as
\[ P(\tau) = e^{B\tau}. \tag{9} \]
If $\mu_0, \ldots, \mu_{n-1}$ are the eigenvalues of $B$ (assumed distinct) then
\[ e^{B\tau} = \sum_{i=0}^{n-1} e^{\mu_i \tau} \prod_{\alpha \neq i} \frac{B - \mu_\alpha}{\mu_i - \mu_\alpha}. \tag{10} \]
The properties of the matrix $B$ are closely related to those of stochastic matrices \[24, 27\], and under quite general conditions $B$ has a single eigenvalue $\mu_0 = 0$ and eigenvalues $\mu_1, \ldots, \mu_{n-1}$ with negative real part. This will be assumed in the following. For up to four intensity periods, the $\mu_i$ can be determined in closed form.

To find the $P_i$’s, we note that for $\tau \to \infty$ the memory to the initial start is in general lost. Therefore, for any $\kappa$,
\[ P_i = P_{\kappa i}(\infty). \tag{11} \]
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Laplace transforming equation (9) yields

$$P(\infty) = \lim_{\epsilon \to +0} \epsilon (\epsilon - B)^{-1}.$$  \hspace{1cm} (12)

Thus the \( P_i \)'s can be calculated without knowledge of the \( \mu_i \)'s.

The steady-state fluorescence intensity is

$$I_{SS} = \sum_i P_i I_i.$$  \hspace{1cm} (13)

Therefore we obtain

$$g(\tau) = \frac{G(\tau)}{I_{SS}^2} = \frac{\sum_{ij} P_i I_i P_{ij}(\tau) g_j(\tau)}{(\sum_i P_i I_i)^2}. \hspace{1cm} (14)$$

so that for larger \( \tau \) the dependence on \( \tau \) of \( g(\tau) \) is solely governed by the statistics of the individual periods.

\begin{figure}[h]
\centering
\includegraphics[width=0.4\textwidth]{figure1.png}
\caption{V system with metastable level 2 and Einstein coefficient \( A_3 \) for level 3. \( \Omega_2 \) and \( \Omega_3 \) are the Rabi frequencies of the two lasers driving the weak 1-2 transition and the strong 1-3 transition, respectively.}
\end{figure}

A first simple example is a system with dark \( (I_0 = 0) \) and light \( (I_1 \neq 0) \) periods, such as the V system of figure 1 with a metastable level or a \( \Lambda \) system 28, 29, 30, 31, 32. The mean period durations are \( T_0 = 1/p_{01} \) and \( T_1 = 1/p_{10} \), respectively. Because \( I_0 = 0 \), equation (13) becomes

$$g(\tau) = \frac{1}{P_1} P_{11}(\tau) g_1(\tau), \hspace{1cm} (15)$$

with \( g_1(\tau) \) the correlation function of the two-level subsystem. A simple calculation yields

$$P_{11}(\tau) = \frac{T_1}{T_0 + T_1} + \frac{T_0}{T_0 + T_1} e^{-\left(\frac{1}{T_0} + \frac{1}{T_1}\right)\tau}$$  \hspace{1cm} (16)

$$P_1 = \frac{T_1}{T_0 + T_1}. \hspace{1cm} (17)$$

For a \( V \) system equation (13) agrees with the result of (17) if the correlation time for \( g_1(\tau) \) is much smaller than \( T_1 \).

For \( \tau \ll T_1 \) one has \( P_{11}(\tau) = 1 \) and \( g(\tau) = g_1(\tau)/P_1 \). Since \( P_1 < 1 \) it follows that, for small \( \tau \), \( g(\tau) \) is just \( g_1(\tau) \) blown up by the factor \( 1/P_1 \). In particular, possible Bloch oscillations of \( g_1(\tau) \) become enhanced in \( g(\tau) \) if \( T_0 \) increases. Moreover, for \( \tau \) values
larger than the correlation time of the two-level subsystem one has $g_1(\tau) = 1$ so that there is a "hump" larger than 1 in $g(\tau)$. In the case of a $V$ system this hump was already noted in [1].

Such a hump is a general feature for any system with a dark period, as will now be shown by means of equation (14). Indeed, for $\tau$ larger than the correlation times of $g_i(\tau)$ but much less than the $T_i$'s one has $P_{ij} = \delta_{ij}$, resulting in $g(\tau) = \sum_i P_i I_i^2 / (\sum \alpha P_\alpha I_\alpha)^2$. By Schwarz's inequality one obtains $(\sum \alpha P_\alpha^{1/2} P_\alpha^{1/2} I_\alpha)^2 \leq (\sum \alpha P_\alpha) (\sum \alpha P_\alpha I_\alpha^2)$. Since $\sum \alpha P_\alpha = 1 - P_0 < 1$ the statement follows. This does not mean, however, that $g(\tau)$ always stays above 1 for all subsequent values of $\tau$. There are systems where it dips below 1 again and then approaches its asymptotic value 1 from below.

In the following, equation (13) for $g(\tau)$ will be studied for an example with three different fluorescence periods and shown to be highly accurate.

3. Application to two dipole-dipole interacting atoms

We consider two dipole-dipole interacting $V$ systems as in figure 1 at a fixed distance $r$, with one laser driving the strong 1-3 transition and another the weak 1-2 transition. Such a system exhibits three fluorescence periods, a dark period $I_0 = 0$, a period $I_1$ and a double intensity period $I_2 \approx 2I_1$. The transition rates $p_{ij}$ between the periods are known [33] and are given in the Appendix. One has $T_0 = 1/p_{01}$, $T_1 = 1/(p_{10} + p_{12})$, $T_2 = 1/p_{21}$, and $p_{02} = p_{20} = 0$. The distance dependent complex dipole-dipole coupling constant $C_3$ is also given in the Appendix. For $r \rightarrow \infty$ the system behaves as two independent, non-interacting, fluorescing $V$ systems, and then the periods $I_2$, $I_1$ and $I_0$ correspond to two, one or no atom radiating. Because of $I_0 = 0$, equation (13) simplifies to

$$g(\tau) = \left\{P_1 I_1^2 P_{11}(\tau) g_1(\tau) + P_1 I_1 I_2 P_{12}(\tau) g_2(\tau) + P_2 I_1 I_2 P_{21}(\tau) g_1(\tau) + P_2 I_2^2 P_{22}(\tau) g_2(\tau) \right\} / (P_1 I_1 + P_2 I_2)^2 .$$

(18)

Here, $g_1(\tau)$ is the usual correlation function of a single two-level system and $g_2(\tau)$ that of two two-level systems which are dipole-dipole interacting. These correlation functions are given in the Appendix. The transition probabilities $P_{ij}(\tau)$ from period $i$ to period $j$ in time $\tau$ are easily calculated from equation (9) and are also given in the Appendix, as is $P_i$, the probability for the occurrence of period $i$.

The accuracy of equation (18) is checked for two values of $\Omega_3$ in figure 2 for a coupling constant $\text{Re} \ C_3 = -0.09 \ A_3$. If the strong laser and the atomic dipole moments are perpendicular to the atomic connecting line this corresponds to a local maximum at an atomic distance $r = 2.7 \lambda$, where $\lambda$ is the wave length of the strong 3-1 transition. The agreement is excellent and becomes even better if one takes higher orders of $C_3$ into account, instead of only the first order as done here.

The usefulness of equation (18) is twofold. First, it can be used for a fit to experimental data to obtain atomic parameters and mean period lengths. Experimentally, correlations functions are in general much easier to determine than
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Figure 2. $g(\tau)$ for two dipole-interacting $V$ systems with an atomic distance $r = 2.7 \lambda$. The result of equation (18) (solid line) is compared with a numerical calculation (dashed line). The hump after $\tau > 10 A_3$ is clearly visible. The difference for small $\tau$ comes from the restriction to first-order terms in $C_3$. (a) $\Omega_3 = 0.3 A_3$ and $\Omega_2 = 0.005 A_3$; (b) $\Omega_3 = 5.0 A_3$ and $\Omega_2 = 0.05 A_3$. 
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period lengths. For such a fit one could also employ numerical solutions for \( g(\tau) \) obtained with the quantum regression theorem [34] or with the quantum jump approach [35, 36, 37, 20]. However, the numerical approach is in general much more sensitive to experimental errors than a fit based on an analytic expression.

The second use of equation (18) lies in the feasibility to study the behavior of \( g(\tau) \) for all parameter values simultaneously, without having to perform many numerical runs and possibly overlooking interesting parameter values. This will be demonstrated here by exhibiting a possible experimental test of the dipole-dipole interaction.

4. Possible experimental verification of the dipole-dipole interaction

For \( \tau \ll T_1, T_2 \), one has \( P_{ij}(\tau) = \delta_{ij} \), so that equation (18) reduces to

\[
g(\tau) = \frac{P_{11}I_1^2 g_1(\tau) + P_{22}I_2^2 g_2(\tau)}{(P_{11}I_1 + P_{22}I_2)^2}.
\]

(19)

In particular, one has \( g_1(0) = 0 \), and \( g_2(0) \) for angle-averaged detection is given in equation (A.4) of the Appendix. Inserting this and the other quantities into equation (19) one obtains

\[
g(0) = \frac{2P_2(A_3^2 + 2\Omega_3^2)(A_3^2 + (\text{Re}C_3)^2)N}{A_3^2(P_1N + 2P_2(A_3^2 + 2\Omega_3^2)(A_3^2 + 2\Omega_3^2 + A_3\text{Re}C_3))}
\]

(20)

with \( N = (A_3^2 + 2\Omega_3^2)^2 + A_3^2|C_3|^2 + 2A_3^2\text{Re}C_3 \). To first order in the coupling constant \( C_3 \) this becomes

\[
g(0) = \frac{1}{2} - \frac{A_3}{2} \frac{(A_3^2 + \Omega_3^2)^2 + \Omega_3^4}{(A_3^2 + \Omega_3^2)^2(A_3^2 + 2\Omega_3^2)\text{Re}C_3}.
\]

(21)

By varying \( \Omega_3 \) one has a noticeable change of \( g(0) \), which depends on how large \( \text{Re}C_3 \) is. For small \( \tau \), the temporal behavior of \( g(\tau) \) is a combination of that of \( g_1(\tau) \) and \( g_2(\tau) \) and can be experimentally resolved. Therefore \( g(0) \) should be measurable and the presence of the dipole-dipole interaction detectable. The deviation of \( g(0) \) from 1/2, i.e. from that for non-interacting atoms, is greatest for small \( \Omega_3 \) and drops off to zero for increasing \( \Omega_3 \). In figure 3 we have plotted \( g(0) \) for various values of \( \Omega_3 \) and for \( \text{Re}C_3 = 0.2 A_3 \), \(-0.1 A_3 \), \(0.1 A_3 \), \(-0.09 A_3 \). If the strong laser and the atomic dipole moments are perpendicular to the atomic connecting line this corresponds to the atomic distance \( r = 1.2 \lambda \), 1.7 \( \lambda \), 2.2 \( \lambda \), 2.7 \( \lambda \), respectively. Since \( \text{Re}C_3 \) oscillatingly drops off to zero for increasing atomic distance, its influence on \( g(0) \) diminishes for increasing atomic distance. As shown in figure 4 the effect is about a factor of two smaller in the case of two dipole-dipole interacting two-level systems.

To show that the temporal behavior of \( g(\tau) \) should be experimentally resolvable we have plotted \( g(\tau) \) in figure 3 for various values of \( \Omega_3 \) and for \( \text{Re}C_3 = -0.09 A_3 \), which corresponds to an atomic distance of \( r = 2.7 \lambda \) for perpendicular laser incidence.
5. Discussion

It has been shown for systems with different fluorescent periods that their intensity correlation function can be reduced to those of simpler subsystems and to quantities that govern the stochastic behavior of the different periods. This is a considerable simplification and allows the determination of an algebraic expression for the intensity correlation function which usually have to be calculated numerically via the quantum
regression theorem or via the quantum jump approach.

One of the advantages of an algebraic expression is the feasibility of studying its behavior for all parameters simultaneously, without having to make numerical runs for different sets of parameters and possibly overlooking interesting values. Another advantage is that it is easier to fit experimental data to an algebraic expression and to obtain unknown atomic parameters or period durations by such a fit rather than fitting to numerically determined expression since the latter procedure is usually much more sensitive to experimental and numerical errors.

It has been shown in this paper that the correlation function for “blinking” systems with dark periods shows a “hump” larger than 1 for values of \( \tau \) larger than the correlation times of the individual intensity periods. This generalizes a similar result for a \( V \) system in [7]. It has also been pointed out that the existence of extended dark periods may considerably enhance the amplitude of Bloch oscillations.

As an application we have derived an expression for the intensity correlation function of two dipole-dipole interacting \( V \) systems and have studied its behavior with respect to the strength of the dipole-dipole interaction. We have shown that there is a significant trace of the dipole-dipole interaction in \( g(\tau) \) for small \( \tau \) which is particularly pronounced for small values of the strong driving. This effect of the dipole-dipole interaction should be experimentally verifiable for atomic distances of a few wavelength of the strong transitions. For two dipole-dipole interacting two-level systems the effect is about a factor of two smaller.
Appendix A.

The temporal intensity correlation function for a single two-level system is given by

\[ g_1(\tau) = 1 - e^{-\frac{3}{4} A\tau} \left( \cos \gamma \tau + \frac{3A}{4\gamma} \sin \gamma \tau \right) \]  

(A.1)

with \( \gamma = \sqrt{16\Omega^2 - A^2/4} \), and the mean intensity is

\[ I_1 = \frac{A\Omega^2}{A^2 + 2\Omega^2} \]  

(A.2)

We have used the approach of [20] and Eq. (27) of [36] to determine the corresponding expression for two dipole-dipole interacting two-level systems and find

\[ g_2(\tau) = 1 - \frac{1}{2} e^{-\frac{3}{4} A\tau} \left( \cos \gamma \tau + \frac{3A}{4\gamma} \sin \gamma \tau \right) \]

\[ -\frac{A \Re C}{2(A^2 + 2\Omega^2)} e^{-\frac{3}{4} A\tau} \left[ \left( 4\Omega^2 + \frac{A(A^2 + 2\Omega^2)(A^2 - 22\Omega^2)}{16\gamma^2} \right) \cos \gamma \tau \right. \]

\[ \left. - \left( \frac{512\Omega^6 + 41A^6 + 2A^2\Omega^2(776\Omega^2 - 391A^2)}{64\gamma^3} + \frac{(A^2 - 6\Omega^2)(A^2 + 2\Omega^2)}{4\gamma} \right) \sin \gamma \tau \right] \]

(A.3)

to first order in the dipole-dipole coupling constant \( C \). To all orders in \( C \) one has [19, 20]

\[ g_2(0) = \frac{A^2 + (\Re C)^2}{2A^2} \left[ 1 + \frac{A(A(\Im C)^2 - 4\Omega^2\Re C)}{(2\Omega^2 + A(A + \Re C))^2} \right] \]

(A.4)

and

\[ I_2 = \frac{2A\Omega^2(2\Omega^2 + A(A + \Re C))}{(A^2 + 2\Omega^2)^2 + A^2\Re C(2A + \Re C) + A^2(\Im C)^2} \].

(A.5)

The dipole-dipole coupling constant \( C \) is given by [20, 39, 40]

\[ C = \frac{3A}{2} e^{ikr} \left[ \frac{1}{ikr} \left( 1 - \cos^2 \vartheta \right) + \left( \frac{1}{(kr)^2} - \frac{1}{i(kr)^3} \right) \left( 1 - 3 \cos^2 \vartheta \right) \right] , \]

(A.6)

where \( \vartheta \) denotes the angle between the dipole moments of the atoms and their connection line and \( k \) is the wave number of the strong transition. We assume \( \vartheta = \pi/2 \) for maximal values of \( C \). In Sections 3 and 4 these expressions are used with \( A = A_3, \Omega = \Omega_3 \) and \( C = C_3 \).

For systems with two different light periods and a dark period the relevant transition probabilities, \( P_{ij}(\tau) \), from period \( i \) to period \( j \) in time \( \tau \) are found from equation (9). For physical reasons it is assumed that \( p_{02} \) and \( p_{20} \) vanish. The eigenvalues of the matrix \( B \) are then \( \mu_0 = 0 \) and

\[ \mu_{1,2} = -\frac{1}{2}(p_{01} + p_{10} + p_{12} + p_{21}) \pm \frac{1}{2} \sqrt{(p_{01} + p_{10} - p_{12} - p_{21})^2 + 4p_{10}p_{12}} \].

(A.7)

From this one obtains by means of equation (10)

\[ P_{11}(\tau) = \frac{P_{01}p_{21}}{\mu_1\mu_2} - \frac{e^{i\mu_1\tau}}{\mu_1(\mu_1 - \mu_2)} \left( p_{10}(p_{21} + \mu_1) + p_{12}(p_{01} + \mu_1) \right) \]
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\[ P_{12}(\tau) = \frac{p_{01}p_{12}}{\mu_1\mu_2} + \frac{p_{12}e^{i\mu_1\tau}}{\mu_1(\mu_1 - \mu_2)}(p_{01} + \mu_1) - \frac{p_{12}e^{i\mu_2\tau}}{\mu_2(\mu_1 - \mu_2)}(p_{01} + \mu_2) \]  

(A.8)

\[ P_{21}(\tau) = \frac{p_{01}p_{21}}{\mu_1\mu_2} + \frac{p_{21}e^{i\mu_1\tau}}{\mu_1(\mu_1 - \mu_2)}(p_{01} + \mu_1) - \frac{p_{21}e^{i\mu_2\tau}}{\mu_2(\mu_1 - \mu_2)}(p_{01} + \mu_2) \]  

(A.9)

\[ P_{22}(\tau) = \frac{p_{01}p_{12}}{\mu_1\mu_2} + \frac{p_{21}e^{i\mu_1\tau}}{\mu_1(\mu_1 - \mu_2)}(p_{12} + p_{21} + \mu_2) - \frac{p_{21}e^{i\mu_2\tau}}{\mu_2(\mu_1 - \mu_2)}(p_{12} + p_{21} + \mu_1) \]  

(A.10)

and, for \( \tau \to \infty \), by means of equation (4)

\[ p_0 = \frac{p_{10}p_{21}}{p_{10}p_{21} + p_{01}p_{12} + p_{01}p_{21}} \]  

(A.12)

\[ p_1 = \frac{p_{10}p_{21} + p_{01}p_{12} + p_{01}p_{21}}{p_{10}p_{21} + p_{01}p_{12} + p_{01}p_{21}} \]  

(A.13)

\[ p_2 = \frac{p_{10}p_{21} + p_{01}p_{12} + p_{01}p_{21}}{p_{10}p_{21} + p_{01}p_{12} + p_{01}p_{21}} \]  

(A.14)

For two dipole-dipole interacting \( V \) systems with metastable state (figure 1) the \( p_{ij} \) have been calculated in [33]. For this \( V \) system the dipole-dipole coupling constant \( C_2 \) can be replaced by 0 since \( A_2 \) is very small. For simplification we have also put \( A_2 = 0 \) which does not change the overall results. For zero detuning the transition rates \( p_{ij} \) between the three periods are then given in [33] as

\[ p_{01} = \frac{2A_3\Omega_3^2}{\Omega_3^2} \]  

(A.15)

\[ p_{10} = \frac{A_3\Omega_2^2}{(A_3^2 + 2\Omega_3^2)\Omega_3^2} \]  

(A.16)

\[ p_{12} = \Omega_2^2 \left[ \frac{A_3^2}{\Omega_3^2} + \text{Re}C_3 \frac{2A_3^2}{(A_3^2 + 2\Omega_3^2)\Omega_3^2} \right] \]  

(A.17)

\[ p_{21} = \Omega_2^2 \left[ \frac{2A_3^2}{(A_3^2 + 2\Omega_3^2)\Omega_3^2} + \text{Re}C_3 \frac{4A_3^4(A_3^2 + 4\Omega_3^2)}{(A_3^2 + 2\Omega_3^2)^3\Omega_3^2} \right] \]  

(A.18)

to second order in \( \Omega_2 \) and first order in \( C_3 \), with the remaining \( p_{ij} \) being zero. The calculation of the expressions to all orders in \( C_3 \) and for nonzero detuning is also given in [33]. For the period durations one has \( T_0 = 1/p_{01}, T_1 = 1/(p_{10} + p_{12}) \) and \( T_2 = 1/p_{21} \). It is noteworthy that \( P_1 \) becomes independent of \( \Omega_2 \), up to second order and to all orders of \( C_3 \).

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