Spontaneous-emission suppression via multiphoton quantum interference

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Abstract. The spontaneous emission is investigated for an effective atomic two-level system in an intense coherent field with frequency lower than the vacuum-induced decay width. As this additional low-frequency field is assumed to be intense, multiphoton processes may be induced, which can be seen as alternative transition pathways in addition to the simple spontaneous decay. The interplay of the various interfering transition pathways influences the decay dynamics of the two-level system and may be used to slow down the spontaneous decay considerably. We derive from first principles an expression for the Hamiltonian including up to three-photon processes. This Hamiltonian is then applied to a quantum mechanical simulation of the decay dynamics of the two-level system. Finally, we discuss numerical results of this simulation based on a rubidium atom and show that the spontaneous emission in this system may be suppressed substantially.

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1. Introduction

While spontaneous decay is a fundamental ingredient for many physical processes, many applications have been proposed recently where spontaneous processes are amongst the main limiting factors [1]. These schemes usually rely on the persistence of either population trapped in a specific state or of coherences on timescales long as compared to typical atomic timescales such as the lifetime of an atomic state. A well-known example for this restriction is the construction of a high frequency laser, where it is hard to reach a population inversion as the spontaneous decay is considerable on high frequency transitions. For the storage and the processing of quantum information, spontaneous emission is a major limitation, because it is necessary to avoid all possible sources of decoherence in these schemes. The same holds for the secure information transmission using quantum effects.
Because of the great interest in these applications, various schemes to modify the spontaneous dynamics of an atomic system have been proposed [2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. One ansatz is the quantum Zeno effect [2]. According to the measurement postulate, a system is projected into one of its eigenstates upon a measurement. If the measurements are repeated rapidly, the system evolution may effectively be stopped, because each measurement projects the system back into the initial state. The technical conditions on the brevity of the pulses though may not always be easily fulfilled for every transition, especially in the free vacuum. Another possibility is a modification of the vacuum surrounding the given atomic system, e.g. by an optical cavity [8]. If the vacuum is modified such that the mode density at the frequency of an atomic transition is decreased, spontaneous processes on the given transition may be suppressed. Here the control of the environmental modes with cavities is rather challenging in reality and not suitable to all schemes. Thirdly it is possible to find superpositions of more than one upper state which are stable [4] or almost stable [5, 6] against spontaneous decay. The suppression of the spontaneous decay here is due to quantum interference effects such as the cancellation of the amplitudes of several possible pathways between two system states [7]. In spite of the conceptual beauty, the disadvantage here often is the difficulty to provide convenient atomic systems which fulfill all conditions such as parallel transition dipole moments for interference to be present. Finally, a system driven by an electromagnetic field periodic in time is somewhat related to systems exhibiting spacial periodicity such as crystals. Thus schemes relying on spacial periodicity which involve a change of the probability for incoherent processes such as the Borrmann effect [13] or the suppression of nuclear reactions [14] may be transferred to laser-driven atomic systems [8]. Recently, this idea has also been applied to modify the decay of a three level system in V-configuration [9].

In [10], a scheme was proposed to slow down the spontaneous decay of the upper state population of an effective atomic two-level system considerably. This scheme makes use of an intense low-frequency laser field of constant frequency and intensity which is applied to the two-level system such that the frequency of the low-frequency field is lower than the decay width of the atomic transition. This additional field induces multiphoton transitions between the two atomic states under consideration, thus allowing for alternative pathways between the two states in addition to spontaneous transitions. In the literature, Hamiltonians describing two-photon processes have been discussed to a great extend [15, 16]. However these mainly apply to systems involving dipole-forbidden transitions. As our aim is to inhibit spontaneous decay, it is not sufficient to look at the spontaneous emission on dipole-forbidden transitions as these rates are very low naturally. For dipole-allowed transitions, two- and four-photon pathways vanish due to parity reasons. Therefore it is necessary to extend the analysis to third-order processes [17]. The leading-order corrections are five-photon processes, which we neglect as these have a low probability in the discussed parameter range.

Thus in this paper, we derive the Hamiltonian describing the interaction of the two-level system with both the vacuum modes and the additional low-frequency field mode
including up to three-photon processes from first principles by adiabatically removing the intermediate atomic states of the multiphoton processes. We calculate the coupling constants which were taken as free parameters in \cite{10} in terms of the properties of the atom and the laser field. Furthermore, the Hamiltonian includes Stark shift effects not discussed in \cite{10}, which however will turn out to be of no importance for the suppression scheme. This Hamiltonian is then applied to a quantum mechanical simulation of the decay dynamics of the two-level system. As it will turn out, the spontaneous decay may be slowed down considerably due to the additional transition pathways induced by the intense low-frequency field. In the final part, this is demonstrated using numerical results of this simulation based on rubidium atoms.

2. Derivation of the multiphoton Hamiltonian

The derivation starts from the usual Hamiltonian for an atomic system coupled to quantized electromagnetic fields \cite{1}. The atomic system consists of two atomic states (typically of opposite parity) which are considered as the ground state \( |1\rangle \) and the excited state \( |2\rangle \) of an two-level atom, which we will denote as the effective two-level system throughout this paper. To model possible multiphoton transitions between the two effective atomic levels, unspecified auxiliary states \( |j\rangle \) (\( j = 3, 4, \ldots \)) are required, which will be adiabatically eliminated throughout the analysis. These states might even be part of the continuum; the only condition on them is that they are sufficiently far away from the two main atomic states as will be explained in more detail below. Since there are no isolated two-level atomic systems, the presence of this states is always guaranteed.

The auxiliary states are necessary as in starting from first principles, i.e. the interaction Hamiltonian \( H_I = -\vec{d}\vec{E} \), we only allow for one-photon transitions between the two

\[
\begin{align*}
|2\rangle \quad |3\rangle, |4\rangle, \ldots
\end{align*}
\]

\[\begin{array}{c}
|1\rangle
\end{array}\]

**Figure 1.** The two-level atom and the auxiliary states \( |3\rangle, |4\rangle, \ldots \) which will be adiabatically removed in the derivation of the multiphoton Hamiltonian. The auxiliary states are show schematically; their relative position is not restricted to be above the two main atomic states. After the adiabatic elimination, the effective two-level system exhibits multiphoton processes as symbolized in the figure. Here, the short arrows depict interactions with the intense low-frequency field and the long arrows interactions with the vacuum field.
effective atomic states. Multiphoton processes are then introduced by considering series of one-photon processes from one of the two effective states to the other effective state via one or more of the auxiliary states, which within the adiabatic approximation occur simultaneously. The result will be a Hamiltonian involving the states of the effective two-level system only, which will turn out to be a generalization of the one used in [10].

The applied field consists of the low frequency field of frequency $\bar{\omega}$ and the vacuum field modes. The following analysis is similar to the one in [15], and the notation is close to [15, 10]. The system Hamiltonian is given by

$$H = H_0 + H_I,$$

$$H_0 = \hbar \omega_1 |1\rangle \langle 1| + \hbar \omega_2 |2\rangle \langle 2| + \sum_{j \notin \{1, 2\}} \hbar \omega_j |j\rangle \langle j| + \hbar \bar{\omega} b^\dagger b + \hbar \sum_k \omega_k a_k^\dagger a_k,$$

$$H_I = -\vec{d} \vec{E}.$$

Here, $\hbar \omega_1$, $\hbar \omega_2$ and $\hbar \omega_j$ ($j = 3, 4, \ldots$) are the energies of state $|1\rangle$, $|2\rangle$ and auxiliary state $|j\rangle$, respectively. $k$ is a multi index over all polarization modes and vacuum frequencies $\omega_k$. Throughout the analysis, we denote sums over all auxiliary states excluding the two effective atomic states $|1\rangle$, $|2\rangle$ by the sum subscript $j \notin \{1, 2\}$, where $j$ is the summation index. $b$ ($b^\dagger$) is a low-frequency field annihilation (creation) operator, $a_k$ ($a_k^\dagger$) annihilates (creates) a vacuum photon of mode $k$, $\vec{d}$ is the total atomic dipole operator, and $\vec{E}$ is the electric field given by

$$\vec{E} = i \left( \mathcal{E} (\vec{e} b - \vec{e}^* b^\dagger) + \sum_k \mathcal{E}_k (\vec{e}_k a_k - \vec{e}_k^* a_k^\dagger) \right).$$

The $\vec{e}, \vec{e}_k$ are polarization vectors, while the $\mathcal{E}, \mathcal{E}_k$ are field amplitudes. The frequency corresponding to the low-frequency field operators $b, b^\dagger$ is assumed to be different from the relevant frequencies for the vacuum photons, which are close to the atomic transition frequency of the effective two-level system, such that the low-frequency field operators commute with the relevant vacuum field operators. In the following analysis, we make use of the approximation that no multiphoton transitions involving two or more spontaneous photons with different frequencies are included. As in our system spontaneous one-photon transitions are possible, transitions involving more than one spontaneous photon are highly suppressed; see the processes considered and observed in [19]. However we include multiple spontaneous photons with the same frequency to account for Stark shifts, which arise from interactions of the atom with the electromagnetical field as a sequence of an absorption and an emissions (or vice versa) which does not change the state of the atomic system. Also we consider all possible combinations of interactions with the low-frequency field as it is intense.

The interaction part of the Hamiltonian may be expanded as follows:

$$H_I = -\vec{d} \vec{E} = -\sum_{n,m} |n\rangle \langle n| \bar{d} |m\rangle \langle m| \vec{E} = -\sum_{n,m} \bar{d}_{nm} \sigma_{nm} \vec{E}$$

$$= -\left( \bar{d}_{12} \sigma_{12} + \bar{d}_{21} \sigma_{21} \right) \vec{E} - \sum_{j \notin \{1, 2\}} \left( \bar{d}_{1j} \sigma_{1j} + \bar{d}_{j1} \sigma_{j1} + \bar{d}_{2j} \sigma_{2j} + \bar{d}_{j2} \sigma_{j2} \right) \vec{E}$$
\[ - \sum_{j,j' \notin \{1,2\}} \vec{d}_{jj'} \sigma_{jj'} \vec{E}. \quad (5) \]

\[ \sigma_{nm} = \langle n | m \rangle \text{ is the atomic transition operator, } \vec{d}_{nm} = \langle n | \vec{d} | m \rangle \text{ is the dipole moment corresponding to transition } n \leftrightarrow m. \text{ Some of these dipole moments may be zero due to symmetry reasons, however as this depends on the configuration of the auxiliary states, we keep this most general form of the Hamiltonian. A specific example for this will be given in section 3.2 where we discuss rubidium as a model system. In this expansion, the first part in equation (5) corresponds to direct transitions between the ground and the excited state by dipole coupling to one of the electric field components (i.e. one-photon transitions). The second addend couples both the ground and the excited state to one of the auxiliary states. After an adiabatic elimination of these intermediate states, this part will turn out to represent multiphoton transitions. Obviously n-photon transitions with } n > 2 \text{ require transitions amongst the auxiliary states; thus these effects are accounted for by the last part in equation (5). These last two parts together with the intense low-frequency field lead to a system evolution which qualitatively differs from the well-known Mollow-type evolution for two-level systems resonantly driven by a laser field [18]. It is important to note that a higher order multiphoton treatment based on the above Hamiltonians involves operator ordering issues, as in the following analysis the atomic transition operators involving auxiliary states will be represented by expressions containing photon operators. If these issues are not taken into account, the resulting Hamiltonians are not self-adjoint. Thus strictly speaking, expressions like

\[ (\vec{d}_{1j} \sigma_{1j} + \vec{d}_{j1} \sigma_{j1}) (\vec{e}b - \vec{e}^* b^\dagger) \]

in the above Hamiltonians are understood to be read as

\[ \vec{d}_{j1} \sigma_{j1} (\vec{e}b - \vec{e}^* b^\dagger) + (\vec{e}b - \vec{e}^* b^\dagger) \vec{d}_{1j} \sigma_{1j}. \quad (7) \]

As the above equation equation (5) is difficult to solve exactly, we use a perturbative approach which consists of an expansion in the number of interactions with the auxiliary levels. The lowest order Hamiltonian thus does not include any interactions with the auxiliary levels and may therefore be written as

\[ H_i^{(1\text{-photon})} = - \left( \vec{d}_{12} \vec{E} \sigma_{12} + \vec{d}_{21} \sigma_{21} \vec{E} \right). \quad (8) \]

This Hamiltonian simply describes the coupling of the vacuum- and the low-frequency field to the effective two-level atom and does not contain contributions of the auxiliary states. To account for two-photon processes, we have to consider transitions from the ground or the excited state into and out of the auxiliary states, but no transitions amongst the auxiliary states. For this we introduce the transition operators \( \sigma_{1j}^{(0)}, \sigma_{2j}^{(0)} \) and their conjugates, where the super index (0) denotes the lowest order approximation whose equations of motion do not include any contribution of the auxiliary states. Only the operators involving the auxiliary states are approximated, as the operators connecting only the ground and the excited state contain the main evolution of the system and therefore have to be taken into account to all orders. Thus the two-photon
Hamiltonian is given by
\[ H_i^{(2\text{-photon})} = -\left( \tilde{d}_{12} \tilde{E} \sigma_{12} + \tilde{d}_{21} \sigma_{21} \tilde{E} \right) - \sum_{j \notin \{1,2\}} \left( \tilde{d}_{ij} \tilde{E} \sigma_{ij}^{(0)} + \tilde{d}_{j1} \sigma_{j1}^{(0)} \tilde{E} \right) + \tilde{d}_{2j} \tilde{E} \sigma_{2j}^{(0)} + \tilde{d}_{j2} \sigma_{j2}^{(0)} \tilde{E} \].

As it will be shown in section 2.1, the transition operators \( \sigma_{ij}^{(0)}, \sigma_{ij}^{(1)} \) and their conjugates consist of sums of products of a transition operator \( \sigma_{ij} \) \( (i, j \in \{1, 2\}) \) and a photon annihilation or creation operator. Thus it may easily be seen that contributions to the Hamiltonian in equation (9) containing transition operators to one of the auxiliary states describe two-photon processes. Including up to three-photon processes, a transition from one of the two effective atomic levels to one of the auxiliary levels may be followed by a transition to another auxiliary state. Thus the three-photon Hamiltonian may be expressed in terms of the first order transition operators \( \sigma_{ij}^{(1)}, \sigma_{ij}^{(2)} \) and their conjugates, whose equations of motion contain either lowest-order transition operators to the auxiliary state or transitions between the two effective atomic states. Also, transition operators between two auxiliary states are possible in lowest order; transition operators to one of the auxiliary states may be expressed in terms of the first order transition operators to one of the effective atomic states. This yields
\[ H_i^{(3\text{-photon})} = -\left( \tilde{d}_{12} \tilde{E} \sigma_{12} + \tilde{d}_{21} \sigma_{21} \tilde{E} \right) - \sum_{j, j' \notin \{1,2\}} \left( \tilde{d}_{ij} \tilde{E} \sigma_{ij}^{(1)} + \tilde{d}_{j1} \sigma_{j1}^{(1)} \tilde{E} + \tilde{d}_{2j} \tilde{E} \sigma_{2j}^{(1)} + \tilde{d}_{j2} \sigma_{j2}^{(1)} \tilde{E} \right) \]
as the three-photon Hamiltonian. Here, the operator ordering was not applied to the term containing a double sum over \( j, j' \) for notational simplicity as it will turn out to be irrelevant for the present analysis. For higher order processes, \( H_i^{(n\text{-photon})} \) with \( n > 3 \) may be obtained similarly. Introducing the coupling constants
\[ \lambda_{ij} = -\frac{i \mathcal{E} \tilde{d}_{ij} \tilde{E}}{\hbar}, \quad \lambda_{ijk} = -\frac{i \mathcal{E}_k \tilde{d}_{ij} \tilde{E}_k}{\hbar} \]
where the transition dipole moments are assumed to be real, the Hamiltonians may be written as
\[ H_i^{(1\text{-photon})} = \hbar \left\{ \lambda_{12} b + \lambda_{12}^* b^\dagger + \sum_k \left( \lambda_{12k} a_k + \lambda_{12k}^* a_k^\dagger \right) \right\} \sigma_{12} + \text{h.c.}, \]
\[ H_i^{(2\text{-photon})} = H_i^{(1\text{-photon})} + \hbar \sum_{m=1}^2 \sum_{j \notin \{1,2\}} \left\{ \lambda_{mj} b + \lambda_{mj}^* b^\dagger + \sum_k \left( \lambda_{mjk} a_k + \lambda_{mjk}^* a_k^\dagger \right) \right\} \sigma_{mj}^{(0)} + \text{h.c.}, \]
\[ H_i^{(3\text{-photon})} = H_i^{(1\text{-photon})} + \hbar \sum_{m=1}^2 \sum_{j \notin \{1,2\}} \left\{ \lambda_{mj} b + \lambda_{mj}^* b^\dagger + \sum_k \left( \lambda_{mjk} a_k + \lambda_{mjk}^* a_k^\dagger \right) \right\} \sigma_{mj}^{(1)} + \hbar \sum_{j, j' \notin \{1,2\}} \left\{ \lambda_{jj'} b + \lambda_{jj'}^* b^\dagger + \sum_k \left( \lambda_{jj'k} a_k + \lambda_{jj'k}^* a_k^\dagger \right) \right\} \sigma_{jj'}^{(0)} + \text{h.c.}. \]
2.1. Atomic transition operators

2.1.1. Equations of motion  The equations of motion for the various operators involved may be obtained using the Heisenberg equation

$$\frac{d}{dt}O = \frac{i}{\hbar}[H, O]$$

(15)

where $O$ is an operator in the Heisenberg picture. For the transition operator $\sigma_{ij}$ with $i = 1, j \notin \{1, 2\}$ - which is one of the elements that occurs in the above Hamiltonians in equations (13, 14) - this may be expanded as follows to the lowest two orders in the interaction with the auxiliary states:

$$\dot{\sigma}_{1j}^{(0)} = i \omega_{1j} \sigma_{1j}^{(0)} + \frac{i}{\hbar} \left( \tilde{d}_{1j} \sigma_{11} + \tilde{d}_{2j} \sigma_{12} \right) \vec{E},$$

(16)

$$\dot{\sigma}_{1j}^{(1)} = i \omega_{1j} \sigma_{1j}^{(1)} + \frac{i}{\hbar} \left( \tilde{d}_{1j} \sigma_{11} + \tilde{d}_{2j} \sigma_{12} \right) \vec{E} - \frac{i}{\hbar} \left( \tilde{d}_{21} \vec{E} \sigma_{2j}^{(0)} - \sum_{n \notin \{1, 2\}} \tilde{d}_{jn} \sigma_{1n}^{(0)} \vec{E} \right),$$

(17)

where $\omega_{nm} = \omega_n - \omega_m$ ($n, m \in \mathbb{N}$). The zeroth order equation does not include references to the auxiliary states, while the first order includes operators connecting one of the main atomic states with an auxiliary state in zeroth order. Note that for simplicity we omitted an addend containing the transition operator $\sigma_{nj}^{(0)}$ in the first-order equation, as it will turn out to be zero (see equation (28)). Higher order expressions may be obtained similarly. For $i = 2, j \notin \{1, 2\}$, we have:

$$\dot{\sigma}_{2j}^{(0)} = i \omega_{2j} \sigma_{2j}^{(0)} + \frac{i}{\hbar} \left( \tilde{d}_{1j} \sigma_{21} + \tilde{d}_{2j} \sigma_{22} \right) \vec{E},$$

(18)

$$\dot{\sigma}_{2j}^{(1)} = i \omega_{2j} \sigma_{2j}^{(1)} + \frac{i}{\hbar} \left( \tilde{d}_{1j} \sigma_{21} + \tilde{d}_{2j} \sigma_{22} \right) \vec{E} - \frac{i}{\hbar} \left( \tilde{d}_{21} \vec{E} \sigma_{1j}^{(0)} - \sum_{n \notin \{1, 2\}} \tilde{d}_{jn} \sigma_{2n}^{(0)} \vec{E} \right).$$

(19)

For $i, j \notin \{1, 2\}$ the corresponding expression in lowest order simply becomes

$$\dot{\sigma}_{ij}^{(0)} = i \omega_{ij} \sigma_{ij}^{(0)}.$$

(20)

2.1.2. First-order transition operators  The basic tool to eliminate the auxiliary states from the equation of motion is the adiabatic integration. For this, the equations of motion are written in terms of slowly varying operators which are denoted by the corresponding symbol with a tilde and which may be seen as an interaction picture representation of the operator:

$$\tilde{\sigma}_{12} = \sigma_{12} e^{i(\omega + \omega_k)t}, \quad \tilde{\sigma}_{1j} = \sigma_{1j} e^{-i\omega_{1j}t},$$

$$\tilde{\sigma}_{ij} = \sigma_{ij}, \quad \tilde{\sigma}_{2j} = \sigma_{2j} e^{-i\omega_{2j}t},$$

$$\tilde{b} = b e^{i\omega t}, \quad \tilde{a}_k = a_k e^{i\omega_k t}. $$

(21)

The adiabatic approximations with superscripts $(0), (1)$ are transferred as their full counterparts. The definition of the slowly varying operators is chosen as in [15] to allow for a comparison of the results. This for example yields using equations (11), (14) and
The first addend of equation (22) is shown to illustrate the basic idea of a diabatic integration:

Now we integrate over time, using the partial integration rule. For simplicity, only the changing operators $\tilde{a}_k$ are dropped, yielding using (21) and including all addends:

\[
\tilde{\sigma}^{(0)}_{1j} = -i \int \lambda_{j1} \tilde{\sigma}_{11} \tilde{b} e^{-i(\omega_{1j} + \bar{\omega})t} dt + (\text{other addends})
\]

\[
= \lambda_{j1} \tilde{\sigma}_{11} \tilde{b} e^{-i(\omega_{1j} + \bar{\omega})t} - \int \frac{\lambda_{j1} e^{-i(\omega_{1j} + \bar{\omega})t}}{(\omega_{1j} + \bar{\omega})} \left\{ \frac{d}{dt} (\tilde{\sigma}_{11} \tilde{b}) \right\} dt + (\text{other addends}).
\]

(23)

In the adiabatic integration one now assumes that the time evolution of the slowly changing operators

\[
\frac{d}{dt} (\tilde{\sigma}_{11} \tilde{b})
\]

(24)

(which is typically of the order of the product of the atomic decay rate or the Rabi frequencies involved) is low as compared to the oscillation of the exponential function. This is fulfilled if the Rabi frequencies involved are not too large and if the intermediate states $|j\rangle$ ($j \notin \{1, 2\}$) are sufficiently far away from the atomic ground and excited state, which we assume in the following. Therefore the integral on the right hand side of equation (23) may be dropped, yielding using (21) and including all addends:

\[
\tilde{\sigma}^{(0)}_{1j} = \frac{\lambda_{j1} \tilde{\sigma}_{11} \tilde{b}}{(\omega_{1j} + \bar{\omega})} e^{-i\bar{\omega}t} + \frac{\lambda_{j2} \tilde{\sigma}_{12} \tilde{b}}{(\omega_{1j} + \omega + \omega_{1j})} e^{-i(2\bar{\omega} + \omega) t}
\]

\[
+ \frac{\lambda^{*}_{j1} \tilde{\sigma}_{21} \tilde{b}}{(\omega_{1j} - \omega)} e^{i\omega t} + \frac{\lambda^{*}_{j2} \tilde{\sigma}_{22} \tilde{b}}{(\omega_{1j} + \omega) (\omega_{1j} + \bar{\omega})} e^{-i(2\bar{\omega} + \omega) t} + \frac{\lambda^{*}_{j2} \tilde{\sigma}_{22} \tilde{a}}{(\omega_{1j} - \omega)} e^{i\omega t}.
\]

(25)

as the lowest order adiabatic approximation for this atomic transition operator. A similar calculation yields

\[
\tilde{\sigma}^{(0)}_{2j} = \frac{\lambda_{j2} \tilde{\sigma}_{22} \tilde{b}}{(\omega_{2j} + \bar{\omega})} e^{-i\bar{\omega}t} + \frac{\lambda^{*}_{j2} \tilde{\sigma}_{22} \tilde{b}}{(\omega_{2j} - \omega)} e^{i\omega t} + \frac{\lambda_{j2} \tilde{\sigma}_{22} \tilde{a}}{(\omega_{2j} + \omega)} e^{-i\bar{\omega}t}
\]

\[
+ \frac{\lambda^{*}_{j2} \tilde{\sigma}_{22} \tilde{a}}{(\omega_{2j} - \omega)} e^{i\omega t} + \frac{\lambda_{j2} \tilde{\sigma}_{22} \tilde{b}}{(\omega_{2j} + \omega)} e^{-i(2\bar{\omega} + \omega) t} + \frac{\lambda^{*}_{j2} \tilde{\sigma}_{22} \tilde{a}}{(\omega_{2j} - \omega)} e^{i(2\bar{\omega} + \omega) t}.
\]

(26)

From equation (20), we obtain for $i, j \notin \{1, 2\}$ in the slowly varying operator frame:

\[
\tilde{\sigma}^{(0)}_{ij} = 0.
\]

(27)
As the auxiliary states are far from resonance with the applied frequencies, in lowest order of the adiabatic approximation we thus have \[ \tilde{\sigma}^{(0)}_{ij} = 0 = \sigma^{(0)}_{ij} \] for \( i, j \notin \{1, 2\} \). Another way of seeing this is that due to the vanishing time derivative, in this order of approximation the populations of the auxiliary states remain constant. As these populations are zero if the population initially is distributed over the ground and the excited state, they are empty at all times.

These results are sufficient to calculate the effective two-photon Hamiltonian equation (9) and to obtain the first order expressions for the transition operators in equations (17) and (19). As the expressions in equations (25) and (26) only contain transition operators involving the ground and the excited state, the elimination of the auxiliary states both from the two-photon Hamiltonian and from the first order expressions for the transition operators is obvious. Note however that the dipole moments connecting the ground and the excited atomic state to the auxiliary states remain in the equations influencing the various coefficients or coupling parameters.

2.1.3. Higher-order transition operators To calculate the first order transition operators, we we insert the lowest-order transition operators equations (25) and (26) and their conjugates in the equations of motion for the first order operators equations (17) and (19). Then the equations are transferred to the slowly changing operator picture using the transformation rules equations (21). The resulting expression for \( \dot{\tilde{\sigma}}^{(1)}_{1j}, \dot{\tilde{\sigma}}^{(1)}_{2j} \) and their conjugates may again be adiabatically integrated yielding \( \sigma^{(1)}_{1j}, \sigma^{(1)}_{j1}, \sigma^{(1)}_{2j} \) and \( \sigma^{(1)}_{j2} \). In addition to the corresponding operator of zeroth order, each of these operators contains 56 addends as first order contribution, 28 of which depend on a sum over auxiliary intermediate atomic states \( |n\rangle \). For example, we have
\[ \sigma^{(1)}_{1j} = \sigma^{(0)}_{1j} + A^{22}_{1j} + A^{21}_{1j} + \sum_{n \notin \{1, 2\}} (A^{11}_{1j}(n) + A^{12}_{1j}(n)) . \]

The operators \( A_{ij} \), which are proportional to \( \sigma_{ij} \), and the other first order transition operators may be found in Appendix A.

2.2. Effective Hamiltonians

In this section, we will use the transition operators derived in the last section to give an explicit representation of the two-photon and the three-photon Hamiltonian in equations (9) and (10). The resulting Hamiltonian will turn out to be equivalent to the Hamiltonian used in [10]. However here we obtain expressions for the coupling constants \( g_k, \bar{g}_i \) introduced as free parameters in the Hamiltonian in [10] and extend the analysis to include Stark shift contributions to the Hamiltonian. The role of the Stark shifts for the decay dynamics of the effective two-level atom will be discussed in section 3.
2.2.1. Two-photon Hamiltonian  Using the rotating-wave approximation, we drop all terms oscillating with frequencies of the order of $\omega_k$ or higher in the two-photon Hamiltonian in equation (23). Thus we neglect counter-rotating interactions with the vacuum field, but do not apply the rotating-wave approximation to the interaction of the atom with the low-frequency field. Transferring the resulting expression back to the Heisenberg picture adopted in the initial equations equation (5) using the transformation rules in equations (21), we obtain

$$H_f^{(2\text{-photon})} = \hbar \sum_k \left\{ \alpha_0 a_k \sigma_{21} + \alpha_1 b_k a_k \sigma_{21} + \alpha_2 b^\dagger_k a_k \sigma_{21} + \text{h.c.} \right\}$$

$$+ \hbar \sum_k (\alpha_3 a_k^\dagger a_k + \alpha_4 a_k^\dagger a_k^\dagger) \sigma_{11} + \hbar (\alpha_5 b^\dagger b + \alpha_6 b^\dagger b + \alpha_{11} b b + \alpha_{11}^* b^\dagger b^\dagger) \sigma_{11}$$

$$+ \hbar \sum_k (\alpha_7 a_k^\dagger a_k + \alpha_8 a_k a_k^\dagger) \sigma_{22} + \hbar (\alpha_9 b^\dagger b + \alpha_{10} b^\dagger b + \alpha_{12} b b + \alpha_{12}^* b^\dagger b^\dagger) \sigma_{22},$$

where the coefficients are given by $\alpha_0 = \lambda_{12k}$,

$$\alpha_1 = \sum_{j \not\in \{1,2\}} \left( \frac{\lambda_{2j} \lambda_{j1k}}{(\Delta - \Delta_j)} - \frac{\lambda_{2j} \lambda_{j1k}}{\Delta_j} - \frac{\lambda_{2j} \lambda_{j1k}}{(\Delta_j + \omega_k - \omega)} + \frac{\lambda_{2j} \lambda_{j1k}}{(\Delta - \Delta_j - \omega_k + \omega)} \right),$$

$$\alpha_2 = \sum_{j \not\in \{1,2\}} \left( \frac{\lambda_{j1k} \lambda_{j2j}}{(\Delta - \Delta_j)} - \frac{\lambda_{j1k} \lambda_{j2j}}{\Delta_j} - \frac{\lambda_{j1k} \lambda_{j2j}}{(\Delta_j - 2\omega)} + \frac{\lambda_{j1k} \lambda_{j2j}}{(\Delta - \Delta_j - \omega_k - \omega)} - \frac{\lambda_{j1k} \lambda_{j2j}}{(\Delta_j + \omega_k - \omega)} \right),$$

$$\alpha_3 = \sum_{j \not\in \{1,2\}} -\frac{2\lambda_{j1k} \lambda_{j1k}^*}{\Delta_j}, \quad \alpha_4 = \sum_{j \not\in \{1,2\}} \frac{2\lambda_{j1k} \lambda_{j1k}^*}{\Delta_j + 2\omega},$$

$$\alpha_5 = \sum_{j \not\in \{1,2\}} -\frac{2\lambda_{j1k} \lambda_{j1k}^*}{\Delta_j + \omega_k - \omega}, \quad \alpha_6 = \sum_{j \not\in \{1,2\}} -\frac{2\lambda_{j1k} \lambda_{j1k}^*}{\Delta_j + \omega_k + \omega},$$

$$\alpha_7 = \sum_{j \not\in \{1,2\}} \frac{2\lambda_{j2k} \lambda_{j2k}^*}{\Delta_j - \Delta_j + \omega_k + \omega}, \quad \alpha_8 = \sum_{j \not\in \{1,2\}} \frac{2\lambda_{j2k} \lambda_{j2k}^*}{\Delta_j - \Delta_j - \omega_k + \omega},$$

$$\alpha_9 = \sum_{j \not\in \{1,2\}} \frac{2\lambda_{j2j} \lambda_{j2j}^*}{\Delta_j - \Delta_j + 2\omega}, \quad \alpha_{10} = \sum_{j \not\in \{1,2\}} \frac{2\lambda_{j2j} \lambda_{j2j}^*}{\Delta_j - \Delta_j},$$

$$\alpha_{11} = \sum_{j \not\in \{1,2\}} \left( -\frac{\lambda_{j1j} \lambda_{j1j}}{(\Delta_j + \omega_k - \omega)} - \frac{\lambda_{j1j} \lambda_{j1j}}{(\Delta_j + \omega_k + \omega)} \right),$$

$$\alpha_{12} = \sum_{j \not\in \{1,2\}} \left( \frac{\lambda_{j2j} \lambda_{j2j}}{(\Delta - \Delta_j)} + \frac{\lambda_{j2j} \lambda_{j2j}}{(\Delta - \Delta_j + 2\omega)} \right),$$

and the detunings are defined as

$$\Delta = \omega_2 - \omega_1 - (\omega_k + \omega), \quad \Delta_j = \omega_j - \omega_k.$$  

Here, the terms proportional to $\alpha_0$ are the usual one-photon Hamiltonian parts. The next addends including $\alpha_1, \alpha_2$ are two-photon transitions which will turn out to be of most interest for the following analysis. Terms with $\alpha_3, \ldots, \alpha_{10}$ may be interpreted as Stark shifts due to the presence of the two electromagnetic field modes. This Hamiltonian is an extension to the one obtained in [15] in that it includes more
transitions such as direct one-photon transitions due to the vacuum and also field ordering effects. These ordering effects are the reason why for example $\alpha_3$ and $\alpha_4$ here are not the same as other than in [15]. The addends proportional to $\alpha_{11}, \alpha_{12}$ are somewhat generalized Stark shifts in that they involve the population operators $a_{11}$ and $a_{22}$. Usually they do not appear in effective multiphoton Hamiltonians as they may be dropped in a rotating wave approximation if the frequency of the involved photons is large enough. For example, the addend with $\alpha_{11}$ stems from a transition $b |j\rangle\langle 1|$ followed by a transition $b |1\rangle\langle j|$, one of which is counter-rotating. But here $b$ represents a low-frequency photon such that these terms may not be dropped a priori. However as discussed later the numerical simulations indicate that they do not disturb the effects described in [10]. One hint that may help to understand this is that from a quantum mechanical point of view, these contributions account for a distribution of the system state over the various Fock states of the low-frequency field without inducing atomic transitions. The trapping effect however does not rely on a specific configuration of the low-frequency field modes such as a concentration of the states to only few of the Fock states.

The interpretation of the general structure of this Hamiltonian is straightforward. In each addend of the $\alpha_l$ ($l = 0, \ldots, 12$), the number of $\lambda$ coefficients is equal to the number of photons exchanged. The various processes involved may also be read off from the $\lambda$ coefficients, as for example in $\alpha_1$ in equation (31), which describes a transition of the effective atom from the ground state $|1\rangle$ to the excited state $|2\rangle$ together with the absorption of both a low-frequency and a spontaneous photon ($b a_{k} a_{21}$ in Hamiltonian equation (30)). The first addend stems from a transition between $|1\rangle$ and an intermediate state $|j\rangle$ via a vacuum-induced transition ($\lambda_{j1k}$) and a low frequency field photon absorption between $|j\rangle$ and $|2\rangle$ ($\lambda_{2j}$). The third addend is due to a transition between $|1\rangle$ and an intermediate state $|j\rangle$ via a low frequency field photon absorption ($\lambda_{j1}$) and a vacuum-induced transition between $|j\rangle$ and $|2\rangle$ ($\lambda_{2jk}$).

2.2.2. Three-photon Hamiltonian Using the results of section 2.1.3 in the expression for the three-photon Hamiltonian equation (10) yields after a calculation as in section 2.2.1 for the two-photon Hamiltonian the following three-photon Hamiltonian:

$$H^{(3\text{-photon})}_I = H^{(2\text{-photon})}_I + \hbar \sum_k \left\{ (\beta_1 b b \dagger + \beta_2 b \dagger b + \beta_3 b b b + \beta_4 b \dagger b \dagger) a_k \sigma_{12} + \text{h.c.} \right\}. \quad (40)$$

Thus as expected the one- and two-photon processes of this Hamiltonian are identical to the ones in the two-photon Hamiltonian equation (30). All additional terms in equation (10) are three-photon processes. The terms with $\beta_1, \beta_2$ are corrections to $\alpha_0$ in the two-photon Hamiltonian, as they effectively induce the same transition. $\beta_3$ and $\beta_4$ are coefficients to the three-photon transition parts, which will be of most interest in the simulation of the decay dynamics. Third-order Stark effects vanish due to symmetry reasons. Introducing the detuning $\Delta_n = \omega_{n1} - \omega_k$, the explicit expressions for the $\beta$ coefficients are given as follows (Note that the sums over $j$ and $n$ have been omitted for notational simplicity; all occurrences of these indices are summed over all auxiliary states).
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\[ \beta_1 = \frac{\lambda_{12} \lambda_{12k} \lambda_{12j}^*}{\Delta_j (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{12} \lambda_{2jk} \lambda_{2j}^*}{\Delta_j (\Delta - \Delta_j + \omega_k + 2\omega)} + \frac{\lambda_{ij} \lambda_{jnk} \lambda_{jnk}^*}{\Delta_n (\Delta_j + \omega_k)} \\
+ \frac{\lambda_{ij} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k - \omega)} + \frac{(\Delta - \Delta_n) (\Delta - \Delta_j - \omega)}{(\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jn}^*}{\Delta_n (\Delta_j + \omega_k)} \\
+ \frac{\lambda_{ij} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jk} \lambda_{ik}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jn}^*}{\Delta_n (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jn}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jk} \lambda_{ik}^*} \\
+ \frac{\lambda_{ij} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jn}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jk} \lambda_{ik}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jn}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jk} \lambda_{ik}^*} \right) \right) \\
(41) \\

\beta_2 = \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k - \omega)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k + \omega)} \\
+ \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k - \omega)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k + \omega)} \\
+ \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} \right) \right) \\
(42) \\

\beta_3 = \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k - \omega)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k + \omega)} \\
+ \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k - \omega)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k + \omega)} \\
+ \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} \right) \right) \\
(43) \\

\beta_4 = \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k - \omega)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k + \omega)} \\
+ \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k - \omega)} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k + \omega)} \\
+ \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_j (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} + \frac{\lambda_{ij} \lambda_{jn} \lambda_{jk} \lambda_{ik}^*}{\Delta_n (\Delta - \Delta_j + \omega_k) + \lambda_{ij} \lambda_{jn} \lambda_{jn}^*} \right) \right) \\
(43)
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Figure 2. The first four transitions involved in the coupling constant $\beta_3$ corresponding to the transition $ba^\dagger_1 \sigma_{12}$. The solid lines denote interactions with the low-frequency field, the dashed line denotes a vacuum photon emission. All paths start in state $|2\rangle$ and end in state $|1\rangle$. The lengths of the arrows are not related to the frequencies of the involved photons.

\begin{align}
&+ \frac{\lambda_{21}^* \lambda_{j1}^* \lambda_{j1}^*}{(\Delta_j + 2\omega_k - \bar{\omega})(\Delta - \Delta_j - \omega_k + \bar{\omega})} + \frac{\lambda_{j2}^* \lambda_{jn}^* \lambda_{n1}^*}{(\Delta - \Delta_j + \bar{\omega})(\Delta - \Delta_n - \omega_k + \bar{\omega})} \\
&+ \frac{\lambda_{j2}^* \lambda_{jn}^* \lambda_{n1}^*}{(\Delta - \Delta_n - \omega_k + \bar{\omega})(\Delta - \Delta_j - \omega_k + 2\bar{\omega})}.
\end{align}

(44)

The structure of the coefficients is similar to the one in the two-photon Hamiltonian, and the contributing transition pathways may be read off each addend. Figure 2 shows the first four processes contributing to $\beta_3$. This coefficient corresponds to the transition $aba^\dagger_1 \sigma_{12}$ in the Hamiltonian equation (40). For example, the first addend in $\beta_3$ in equation (43) describes a path starting by a transition from the excited state $|2\rangle$ to the auxiliary state $|n\rangle$ induced by a low-frequency field absorption, followed by a transition to the auxiliary state $|j\rangle$ induced by a low-frequency field absorption, and then a vacuum photon emission with a transition to the ground state $|1\rangle$. The other transition pathways may be read off the addends accordingly. It is important to note that the frequency of the spontaneous photon emitted or absorbed in the multiphoton processes is always close to the atomic transition frequency, because the sum of the frequencies of all photons (with a relative sign between emitted and absorbed photons) is required to be close to the atomic transition frequency for a spontaneous emission to occur. This will be shown using a Wigner-Weisskopf-like calculation in section 3.

For any application of the above Hamiltonian equation (40), it is important to be able to decide which intermediate states need to be taken into account in the analysis. First of all, the number of exchanged photons restricts the atomic level space. For example in a three-photon transition from a $S$ (angular momentum $l = 0$) to a $P$ ($l = 1$) state, states with $l > 2$ are not possible as intermediate states. The two important parameters which decide about the relevance of the remaining intermediate states are the transition dipole moments which connect the state in question to other relevant states, and the frequency separation to the other states. A reasonable parameter involving these quantities is e.g. the ratio $\beta_i/\alpha_0$ ($i = 1, \ldots, 4$), i.e., the coupling strength of the
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...multiphoton transitions relative to the single photon transition strength. By evaluating the expression for $\beta_i$ for each combination of the intermediate states $j$ and $n$ separately, it is possible to compare the relative weights of the various pathways as e.g. shown in figure 2. Then the states which give rise to pathways with small relative contribution can be neglected. For this, it is not necessary to evaluate all possible combinations. If $m$ is the principal quantum number of an intermediate state which is higher than the principal quantum numbers of the ground and the excited state, then the contribution of the corresponding intermediate state with principal quantum number $m + 1$ can be expected to be lower than the contribution of the state with quantum number $m$ due to the larger frequency separation and due to a lower transition dipole moment.

3. Decay dynamics of the effective two-level system

3.1. General considerations

To further understand the various contributions in the derived Hamiltonian, and to analyze the modification of the decay dynamics due to the additional multiphoton transition pathways we apply the three-photon Hamiltonian equation (40) in a quantum mechanical simulation of the effective two-level atom subject to an intense low-frequency field and the vacuum modes. For this, we rearrange the free and the interaction part of the three-photon Hamiltonian as follows:

$$H^{(3\text{-photon})}_0 = \hbar \omega b \dagger b + \hbar \sum_k \omega_k a \dagger_k a_k$$

$$+ \hbar \left\{ \omega_1 + \alpha_5 b \dagger b + \alpha_6 b \dagger b \dagger + \sum_k (\alpha_3 a \dagger_k a_k + \alpha_4 a_k a \dagger_k) \right\} \sigma_{11}$$

$$+ \hbar \left\{ \omega_2 + \alpha_9 b \dagger b + \alpha_{10} b \dagger b \dagger + \sum_k (\alpha_7 a \dagger_k a_k + \alpha_8 a_k a \dagger_k) \right\} \sigma_{22},$$

(45)

$$H^{(3\text{-photon})}_{\text{vac}} = \hbar \sum_k \left\{ \alpha_0 + \alpha_1 b \dagger b + \alpha_2 b \dagger b \dagger + \beta_1^* b \dagger b \right\} a_k \sigma_{21} + \text{h.c.},$$

(46)

$$H^{(3\text{-photon})}_b = \hbar \left( \alpha_{11} b b \sigma_{11} + \alpha_{12} b b \sigma_{22} \right) + \text{h.c.}.$$  

(47)

$H^{(3\text{-photon})}_0$ is the effective free Hamiltonian, including the Stark shifts of the two atomic levels. $H^{(3\text{-photon})}_{\text{vac}}$ describes the interaction with the vacuum field, i.e. the spontaneous decay. The contributions involving operators $b$ and $b \dagger$ in this part account for multiphoton transitions consisting of one interaction with the vacuum field and one or more interactions with the low-frequency field. $H^{(3\text{-photon})}_b$ contains terms due to the low frequency field alone which drive the effective system.

In order to calculate the system evolution we notice that $H^{(3\text{-photon})}_b$ contains multiphoton processes only such that its coupling constants $\alpha_i$ ($i \in \{11, 12\}$) are moderate even for an intense low-frequency field. Thus in the regime where the previous approximations such as the adiabatic elimination are valid, the evolution which gives
rise to the damping of the system (i.e. the coupling to the vacuum) may be evaluated separately from external driving fields as it is common practice in quantum optical calculations, see e.g. Chapter 8.6.1 in [20]. Thus in our case, we evaluate the system with \( H_0^{(3\text{-photon})} + H_{\text{vac}}^{(3\text{-photon})} \) alone and then combine the resulting equations of motion with the ones resulting from the driving part \( H_b^{(3\text{-photon})} \). This approximation holds if \( || H_b^{(3\text{-photon})} || \ll || H_0^{(3\text{-photon})} || \), which means that coupling constants in \( H_b^{(3\text{-photon})} \) have to be low as compared to the atomic transition frequency of the effective two-level system.

First, we transfer the vacuum interaction part of the Hamiltonian \( H_{\text{vac}}^{(3\text{-photon})} \) to the interaction picture with respect to the free part \( H_0^{(3\text{-photon})} \). To understand the Stark shift contribution, we transfer the partial Hamiltonian \( a_k^\dagger b^n \sigma_{21} \) as an example:

\[
 a_k^\dagger b^n \sigma_{21} \Rightarrow a_k^\dagger b^n \sigma_{21} e^{-i\omega t} S_a S_b
\]

(48)

with

\[
 S_a = e^{-i\sum_i(a_i^\dagger a_i)(\alpha_7 + \alpha_8)(a_i^\dagger a_i - \delta_{kl})} e^{i\sum_i(\alpha_3 + \alpha_4) a_i^\dagger a_i},
\]

\[
 S_b = e^{-i(\alpha_9 + \alpha_{10})b^i b + n)} e^{i(\alpha_5 + \alpha_6) b^i b}
\]

(49)

(50)

as the Stark shift contributions due to spontaneous photons \( S_a \) and due to coherent low-frequency photons \( S_b \). The two exponential functions in each of the contributions describe the Stark shift of the upper level \( |2\rangle \) and of the lower level \( |1\rangle \), respectively. As expected, the shifts are proportional to the number of photons in the respective modes. The difference in the photon number factors for the two states (e.g. \( b^i b + n \) as compared to \( b^i b \)) is due to the fact that the example Hamiltonian part induces changes in the photon numbers. To evaluate their importance, these contributions have to be compared to the exponential factor in equation (48). There is at most one photon in the vacuum modes \( a_i^\dagger a_i \in \{0, 1\} \), thus \( S_a \) may be safely neglected. For the low-frequency field, it is reasonable to assume a coherent state with a large mean number of photons \( N \), as this represents an intense laser field. As the relative photon number distribution width decreases as \( N^{-1/2} \) for coherent states, and as \( 0 \leq n \leq 2 \) in the vacuum part of the three-photon Hamiltonian, we approximate \( b^i b \approx b^i b + n \approx N \) and thus have

\[
 S_a S_b \approx e^{-i(\alpha_9 + \alpha_{10} - \alpha_5 - \alpha_6) N}
\]

(51)

as the Stark shift contribution. It is important to note that the simulated model system does not lose its quantum character in adopting these approximations, which are less inspired by physical than by numerical reasoning, as we keep the photon operators and the distinguishable Fock states for the electromagnetical fields. The above argument holds for all terms in \( H_{\text{vac}}^{(3\text{-photon})} \) with the same result, thus these shifts may be taken care of by introducing an effective atomic transition frequency

\[
 \omega = \omega_2 - \omega_1 + N(\alpha_9 + \alpha_{10} - \alpha_5 - \alpha_6).
\]

(52)

Then the vacuum interaction part becomes

\[
 V_{\text{vac}}^{(3\text{-photon})} = \hbar \sum_k \left( \alpha_0 + \alpha_1 b e^{-i\omega t} + \alpha_2 b^\dagger e^{i\omega t} + \beta_1^* b b^\dagger + \beta_2^* b^i b \\
 + \beta_3^* b^i b^\dagger e^{2i\omega t} + \beta_4^* b b e^{-2i\omega t} \right) a_k \sigma_{21} e^{i(\omega - \omega_k)t} + \text{h.c. .}
\]

(53)
This expression is equivalent to the interaction Hamiltonian in [10], while we do not introduce the generalized ladder operators \( \sigma_+^{(j)} \) and \( \sigma_-^{(j)} \) \((j \in \mathbb{Z})\) here which were used in the semiclassical approximation in [10]. We proceed with the ansatz for the wavefunction

\[
|\Psi(t)\rangle = \sum_n E_n(t) \ket{2, n, 0} + \sum_n \sum_{k'} G_{n}^{k'}(t) \ket{1, n, k'}. \tag{54}
\]

Here the first index in the kets denotes the atomic state, the second slot represents the number of photons in the low-frequency field, and the last entry is either 0 for the vacuum without photons or \( k \) for a single photon in mode \( k \). As described earlier, we first derive an equation of motion for the state amplitudes due to the vacuum part of the Hamiltonian \( V_{\text{vac}}^{(3-\text{photon})} \), which we denote by \( E_{n}^{\text{vac}} \) and \( G_{n}^{k, \text{vac}} \). In a second step, we calculate the equations corresponding to the driving part of the Hamiltonian \( V_{b}^{(3-\text{photon})} \) with amplitudes \( E_{n}^{b} \). These two sets of equations of motion are then summed to give the equations for the full amplitudes \( E_{n} \) and \( G_{n}^{k} \). Inserting the ansatz equation (54) in the Schrödinger equation with Hamiltonian \( V_{\text{vac}}^{(3-\text{photon})} \) yields as vacuum part of the equations of motion for the state amplitudes

\[
i\hbar \frac{d}{dt} E_{n}^{\text{vac}} = \langle 2, n, 0| V_{\text{vac}}^{(3-\text{photon})}|\Psi\rangle = \hbar \sum_{k} \left\{ (\alpha_{0} + \beta_{1}^{*}(n + 1) + \beta_{2}^{*} n) \, G_{n}^{k, \text{vac}} \\
+ \alpha_{1} \sqrt{n + 1} \, G_{n+1}^{k, \text{vac}} \, e^{-i\omega t} + \alpha_{2} \sqrt{n} \, G_{n-1}^{k, \text{vac}} \, e^{i\omega t} \\
+ \beta_{1}^{*} \sqrt{(n + 1)(n + 2)} \, G_{n+2}^{k, \text{vac}} \, e^{-2i\omega t} \\
+ \beta_{2}^{*} \sqrt{n(n - 1)} \, G_{n-2}^{k, \text{vac}} \, e^{2i\omega t} \right\} e^{i(\omega - \omega_{k})t}, \tag{55}
\]

\[
i\hbar \frac{d}{dt} G_{n}^{k, \text{vac}} = \langle 1, n, k| V_{\text{vac}}^{(3-\text{photon})}|\Psi\rangle = \hbar \left\{ (\alpha_{0}^{*} + \beta_{1}(n + 1) + \beta_{2} n) \, E_{n}^{\text{vac}} + \alpha_{1}^{*} \sqrt{n} \, E_{n-1}^{\text{vac}} \, e^{i\omega t} \\
+ \alpha_{2} \sqrt{n + 1} \, E_{n+1}^{\text{vac}} \, e^{-i\omega t} + \beta_{4} \sqrt{n(n - 1)} \, E_{n-2}^{\text{vac}} \, e^{2i\omega t} \\
+ \beta_{3} \sqrt{(n + 1)(n + 2)} \, E_{n+2}^{\text{vac}} \, e^{-2i\omega t} \right\} e^{-i(\omega - \omega_{k})t}. \tag{56}
\]

Formally integrating (56) and inserting the result in (55) allows to obtain an equation of motion for the upper state amplitudes \( E_{n}^{\text{vac}} \) of the form

\[
\frac{d}{dt} E_{n}^{\text{vac}} = -\int_{0}^{t} \sum_{k} \sum_{l=-n-4}^{n+4} C_{l}(t, t') E_{l}^{\text{vac}}(t') \, dt' \tag{57}
\]

where \( C_{l}(t, t') \) are time dependent coefficients depending on the coupling constants \( \alpha_{i} \) and \( \beta_{j} \). Expanding equation (57), it turns out that all addends on the right hand side are of the form

\[
A = -\int_{0}^{t} \sum_{k} R(\omega_{k}) \lambda_{rs} S^{*}(\omega_{k}) \lambda_{xy} e^{in\omega t} e^{i\omega t'} e^{i(\omega - \omega_{k} + \kappa \omega)(t-t')} E_{n+\delta}(t') \, dt'. \tag{58}
\]

Here, \( r, s, x, y, \mu, \nu, \kappa \) and \( \delta \) are integers depending on the specific addend. \( R(\omega_{k}) \lambda_{rs} \) and \( S(\omega_{k}) \lambda_{xy} \) are one of the coefficients \( \alpha_{i} \) \((i = 0, 1, 2)\) or \( \beta_{j} \) \((j = 1, 2, 3, 4)\) respectively, written as products of the dipole moment corresponding to the spontaneous transition
\( \lambda_{rs} \) and \( \lambda_{xy} \) and of the remaining factors \( R(\omega) \) and \( S(\omega) \). In a Wigner-Weisskopf-like calculation, this generic contribution may be evaluated to give

\[
A = \frac{-R(\omega + \kappa \bar{\omega})S(\omega + \kappa \bar{\omega})}{6\hbar\epsilon_0 c^3 \pi} \frac{\tilde{d}_{rs} \tilde{d}_{xy}}{|\tilde{d}_{rs}| \cdot |\tilde{d}_{xy}|} (\omega + \kappa \bar{\omega})^3 E_{n+3}^{vac}(t) e^{i(\mu + \nu)\omega t} = -R(\omega + \kappa \bar{\omega})S(\omega + \kappa \bar{\omega}) p_{rs}^{xy} e^{i(\mu + \nu)\omega t} \frac{1}{2} \sqrt{\Gamma_{rs}(\omega + \kappa \bar{\omega})}\frac{1}{\Gamma_{xy}(\omega + \kappa \bar{\omega})} E_{n+3}^{vac}(t). \tag{59}
\]

Here \( \Gamma_{ij}(x) \) is the spontaneous decay rate of transition \( i \leftrightarrow j \) with transition frequency modified to \( x \), i.e.

\[
\Gamma_{ij}(x) = \left( \frac{1}{4\pi\epsilon_0} \frac{4|d_{ij}|^2\omega_{ij}^3}{3\hbar c^3} \right) x^3 = \bar{\Gamma}_{ij} x^3 \omega_{ij}^3,
\]

where \( \bar{\Gamma}_{ij} \) is the spontaneous decay rate of transition \( i \leftrightarrow j \) with transition frequency \( \omega_{ij} \), and

\[
p_{rs}^{xy} = \frac{\tilde{d}_{rs} \tilde{d}_{xy}}{|\tilde{d}_{rs}| \cdot |\tilde{d}_{xy}|} \tag{60}
\]

is a prefactor describing the amount of quantum interference possible between transition \( r \leftrightarrow s \) and transition \( x \leftrightarrow y \). It is zero if the dipole moments are orthogonal and reaches its maximum value of 1 (-1) for parallel (antiparallel) dipole moments. Thus we find the usual form of a square root of the product of the two corresponding spontaneous decay rates as characteristic for vacuum induced interference effects [5]. The \( R \) and \( S \) merely are prefactors which are present because the corresponding process is a multiphoton process. Assuming as for the Stark shift contributions that the photon number distribution width of the low-frequency field is negligible as compared to the number of photons \( N \), i.e. \( n \approx n + 1 \approx n - 1 \approx \ldots \approx N \), one may introduce the low-frequency field Rabi frequency in equation (57) given by

\[
\Omega_{ij} = 2\lambda_{ij} \sqrt{n + 1} \tag{61}
\]

for transition \( i \leftrightarrow j \). This finally leads to a system of equations for the upper state populations given by

\[
\frac{d}{dt} E_{n}^{vac}(t) = -c_0 E_{n}(t) - c_1 e^{i\omega t} E_{n-1}^{vac}(t) - c_2 e^{-i\omega t} E_{n+1}^{vac}(t) - c_3 e^{2i\omega t} E_{n-2}^{vac}(t)
- c_4 e^{-2i\omega t} E_{n+2}^{vac}(t) - c_5 e^{3i\omega t} E_{n-3}^{vac}(t) - c_6 e^{-3i\omega t} E_{n+3}^{vac}(t)
- c_7 e^{4i\omega t} E_{n-4}^{vac}(t) - c_8 e^{-4i\omega t} E_{n+4}^{vac}(t), \tag{62}
\]

where the \( c_i \) (\( i = 1 \ldots 8 \)) are constant coefficients, whose specific form which may be obtained by expanding equation (57) and (59) is omitted here as e.g. \( c_0 \) contains of several hundred addends in its most general form. The generalized Stark shift contributions in the driving part \( V_b^{(3-photon)} \) of the Hamiltonian give rise to an equation of motion for the upper atomic state amplitude which is given by

\[
i\frac{d}{dt} E_{n}^{b}(t) = \langle 2, n, 0| V_b^{(3-photon)}(t)|\Psi(t) \rangle = d_3 e^{2i\omega t} E_{n-2}^{b}(t) + d_4 e^{-2i\omega t} E_{n+2}^{b}(t) \tag{63}
\]

with constant coefficients \( d_i \) (\( i = 3, 4 \)). Thus the total equation of motion of the upper atomic state amplitude \( E_n \) is given by the sum of the right hand side of the
two contributions of equations (62) and (63) upon replacement of the amplitudes $E_n^{\text{vac}}$ and $E_n^b$ by $E_n$:

$$
\frac{d}{dt} E_n(t) = -c_0 E_n(t) - c_1 e^{i\omega t} E_{n-1}(t) - c_2 e^{-i\omega t} E_{n+1}(t) - (c_3 + i d_3) e^{2i\omega t} E_{n-2}(t) - (c_4 + i d_4) e^{-2i\omega t} E_{n+2}(t) - c_5 e^{3i\omega t} E_{n-3}(t) - c_6 e^{-3i\omega t} E_{n+3}(t) - c_7 e^{4i\omega t} E_{n-4}(t) - c_8 e^{-4i\omega t} E_{n+4}(t). \tag{64}
$$

These equations are a generalization of equations (3) in [10], which means that they have the same structure, but different coefficients $c_i (i = 0, \ldots, 8)$ and $d_i (i = 3, 4)$ than in [10]. The difference is due to the fact that here we include Stark shifts and do not use the semiclassical sum over all possible low-frequency photon numbers as in [10]. Some effects of these differences will be discussed in section 3.2.2. In particular, it will turn out later that the driving terms which may be interpreted as generalized Stark shift terms do not change the decay dynamics of the simulated system in a notable manner.

To simulate the system behavior, one has to choose an initial number of photons $N$ in the low-frequency mode and a possible range of deviations $\Delta_N$ from this number. The set of state amplitudes considered in the analysis is then chosen as \{\{E_n(t)|N - \Delta_N \leq n \leq N + \Delta_N\}\}. In the equations of motion for these state amplitudes according to equation (64), all references to states outside the chosen set are neglected. Thus $\Delta_N$ has to be chosen large enough such that the outermost states are barely populated during the calculated evolution time to avoid numerical artefacts due to the artificially added borders of the simulated level space. As initial condition we choose

$$
|\Psi(0)\rangle = |2, \alpha, 0\rangle = \frac{1}{P} e^{-|\alpha|^2/2} \sum_{n=2-W}^{N+W} \frac{\alpha^n}{\sqrt{n!}} |2, n, 0\rangle. \tag{65}
$$

Thus the atom is in the excited state, the vacuum is assumed empty and the low frequency field is in a coherent state $|\alpha\rangle$ which simulates a strong quantized laser field. The field parameter $\alpha$ is given by

$$
\alpha = \sqrt{N} e^{i\varphi}, \tag{66}
$$

as $|\alpha|^2$ is the expectation value of the photon number in a coherent state. The phase $\varphi$ accounts for the possible complexity of $\alpha$. As only upper state amplitudes $E_n(t)$ with $N - \Delta_N \leq n \leq N + \Delta_N$ are considered in the analysis, also the initial photon number distribution of the coherent state has to be restricted. Thus $W$ is a cutoff of the photon number distribution width chosen such that $W < \Delta_N$. Again, this avoids population losses at the borders of the simulated Hilbert space. $P$ is a normalization constant such that $\langle\Psi(0)|\Psi(0)\rangle = 1$, which is required because of the cutoff width $W$. As a rough consistency check, the range parameters $\Delta_N$ and $W$ must be chosen large enough for the specific system parameters such that increasing these values does not affect the result. After solving this set of $(2\Delta_N + 1)$ coupled ordinary differential equations the total upper state population may be obtained as

$$
\Pi(t) = \sum_{n=N-\Delta_N}^{N+\Delta_N} |E_n(t)|^2. \tag{67}
$$
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\[ |8\rangle = 6P_{3/2} \]
\[ |7\rangle = 6P_{1/2} \]
\[ |3\rangle = 6S_{1/2} \]
\[ |5\rangle = 4D_{3/2} \]
\[ |6\rangle = 4D_{5/2} \]
\[ |2\rangle = 5P_{3/2} \]
\[ |4\rangle = 5P_{1/2} \]
\[ |1\rangle = 5S_{1/2} \]

Figure 3. Partial level scheme of rubidium taken as the simulated model system. The spontaneous emission of transition $|2\rangle \rightarrow |1\rangle$ is to be slowed down by the discussed scheme. The figure is not drawn to scale.

This population may then be compared to the exponential decay for the two-state system without an additional low-frequency field.

3.2. Rubidium as an example

3.2.1. Model system To demonstrate the feasibility of the scheme, we use rubidium as our model system. The simulated atomic levels are shown in figure 3. The population is assumed to be in the $5P_{3/2}$ state initially. Without additional fields, this state decays to the $5S_{1/2}$ state with a decay rate of $\Gamma_{21} = 37.5 \cdot 10^6$ s$^{-1}$ [21]. As discussed in the previous section, within the adiabatic approximation the intermediate states are never populated, see equation (28). Thus it is possible to include states even if there are decay channels leading out of the simulated level space. Also, low spontaneous decay rates do not in general mean that the corresponding transitions do not need to be taken into account, as the low rates may be due to the small energy spacing between two states. The dipole moments however which are important for the low-frequency field may be comparable to the other transition dipole moments. In the simulation, only dipole-allowed transitions are considered. This amounts to a simplification of the equation of the upper state amplitudes equation (64) by e.g. eliminating $c_1$ and $c_2$ which rely on dipole-forbidden transitions, as in addition to the spontaneous photon a second photon is exchanged with the low-frequency field. The energies of the various states are given by [21]

\[ E_1 = 0 \text{ eV} , \quad E_2 = 1.589 \text{ eV} , \]
\[ E_3 = 2.496 \text{ eV} , \quad E_4 = 1.560 \text{ eV} , \]
\[ E_5 = 2.400 \text{ eV} , \quad E_6 = 2.400 \text{ eV} , \]
\[ E_7 = 2.940 \text{ eV} , \quad E_7 = 2.950 \text{ eV} . \]

The energy separation $(E_5 - E_6)$ is about 55 MHz; the main transition has a frequency of $\omega_{21} = 3.84 \cdot 10^{14}$ Hz. We consider decay rates as obtained by theoretical calculations in [21], where also the required branching ratios of the various decay pathways from the
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Figure 4. Dependence of the total upper state population on $\Omega_{12}$. The chosen values are $\Omega_{12} = 1; 3; 5; 8; 10; 12 [10^{12} \text{Hz}]$. The graph order corresponds to the Rabi frequency where the highest graph corresponds to the largest driving strength. The graph for $\Omega_{12} = 1 \cdot 10^{12} \text{Hz}$ is almost on top of the dashed reference curve.

excited states are given:

\[
\begin{align*}
\hat{\Gamma}_{21} &= 37.5 \cdot 10^6 \text{ s}^{-1}, & \hat{\Gamma}_{41} &= 35.6 \cdot 10^6 \text{ s}^{-1}, \\
\hat{\Gamma}_{32} &= 12.9 \cdot 10^6 \text{ s}^{-1}, & \hat{\Gamma}_{34} &= 6.6 \cdot 10^6 \text{ s}^{-1}, \\
\hat{\Gamma}_{52} &= 2.0 \cdot 10^6 \text{ s}^{-1}, & \hat{\Gamma}_{54} &= 10.7 \cdot 10^6 \text{ s}^{-1}, \\
\hat{\Gamma}_{62} &= 11.9 \cdot 10^6 \text{ s}^{-1}, & \hat{\Gamma}_{71} &= 2.4 \cdot 10^6 \text{ s}^{-1}, \\
\hat{\Gamma}_{73} &= 4.3 \cdot 10^6 \text{ s}^{-1}, & \hat{\Gamma}_{75} &= 2.4 \cdot 10^6 \text{ s}^{-1}, \\
\hat{\Gamma}_{81} &= 2.8 \cdot 10^6 \text{ s}^{-1}, & \hat{\Gamma}_{83} &= 4.5 \cdot 10^6 \text{ s}^{-1}, \\
\hat{\Gamma}_{85} &= 0.2 \cdot 10^6 \text{ s}^{-1}, & \hat{\Gamma}_{86} &= 1.7 \cdot 10^6 \text{ s}^{-1}.
\end{align*}
\]

For the transitions which are listed in [22], the above values are in reasonable agreement with the data reported there. The system parameters for the figures are chosen as $N = 10^6$, $\Delta N = 15000$, $W = 500$, $\phi = 0$, $\bar{\omega} = 0.1 \text{ MHz}$ and $p_{rs}^{xy} = 1$ if not stated otherwise. Here $N$ only affects the initial population of the low-frequency Fock states. It is chosen independent of the field strength of the low-frequency field in order to estimate the dependence of the population dynamics on the initial conditions. The value $p_{rs}^{xy} = 1$ has been chosen for simplicity, but is not required to for our scheme to work. A calculation with a $p$-value of 0.5 reduces the trapping duration e.g. of the top curve in figure 4 by less than a factor of 3, such that the remaining effect is still considerable. For this one should note that other than in most previously studied systems exhibiting quantum interference effects [4, 5, 6, 7], in our setup non-zero values for $p_{rs}^{xy}$ can be found in any atomic system, which is due to the fact that here the two transitions do not need to have a similar transition frequency or a common atomic state. To speed up the numerical calculations, the driving terms in equation (63) are suppressed by the replacement

\[d_i \rightarrow \varrho d_i \ (i = 3, 4)\] (68)
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Figure 5. Dependence of the population trapping on the low-frequency field frequency $\bar{\omega}$. Chosen values are $\bar{\omega} = 10^2; 10^4; 10^6; 10^7; 10^8; 10^9$ Hz. (a) $\bar{\omega} = 10^2; 10^4$ Hz; (b) $\bar{\omega} = 10^6$ Hz; (c) $\bar{\omega} = 10^7$ Hz and (d) $\bar{\omega} = 10^8; 10^9$ Hz. The two curves in (d) can be distinguished as the one corresponding to the lower frequency $\bar{\omega}$ slightly wiggles. The dashed curve is the reference.

in equation (64), where we choose $\rho = 1/1000$ in the following calculations. For this one should note that for the above parameters the simulation consists of $2\Delta N + 1 = 30001$ coupled complex differential equations. The validity of this suppression is discussed in section 3.2.2. In the following figures 4-7, dashed lines are reference curves given by $\exp(-\hat{\Gamma}_{21}t)$.

3.2.2. Results The first figure 4 shows the total upper state population $\Pi(t)$ for different values of $\Omega_{12}$. The chosen values for this Rabi frequency are $\Omega_{12} = 1; 3; 5; 8; 10; 12 \times 10^{12}$ Hz. As expected an increasing Rabi frequency of the low-frequency field increases the amount of trapping. The reason for this is that the relative probability of low-frequency-field-assisted transitions increases with an increasing field strength. However the Rabi frequency must not be chosen too high as otherwise the adiabatic approximation is not valid any longer. Also higher-order processes with more than three exchanged photons have to be considered if the Rabi-frequency is chosen too large. The Rabi frequency $\Omega_{12} = 10^{13}$ Hz chosen in most of the figures is about $1/40$ of the atomic transition frequency $\omega_{21}$, which is well within the validity range of the applied approximations.

Figure 5 shows the role of the low-frequency field frequency $\bar{\omega}$. The chosen values are $\bar{\omega} = 10^2; 10^4; 10^6; 10^7; 10^8; 10^9$ Hz, while the other parameters are chosen as in figure 4 with $\Omega_{12} = 10^{13}$ Hz. As long as $\bar{\omega}$ is low as compared to the natural decay width $\hat{\Gamma}_{12}$, the result is independent of $\bar{\omega}$ and the population plots are on top of each other. For higher frequencies, the population trapping decreases until the oscillation due to $\bar{\omega}$ is visible for $\bar{\omega} \lesssim \hat{\Gamma}_{12}$. The behavior of the system in the limit of small frequencies $\bar{\omega}$ is somewhat different from the behavior found in [10]. There the trapping was found to improve with decreasing $\bar{\omega}$ until in the limit $\bar{\omega} \rightarrow 0$ the decay was completely stopped. As already discussed in [10], this is the expected behavior for the system simulated in [10] which is equivalent to a system with near-degenerate upper states [23]. But while in
our model in [10] all other parameters were kept constant in changing $\bar{\omega}$, in the present analysis other parameters such as the various coupling strengths also change with $\bar{\omega}$, thus leading to a different behavior. Still the slowing down of the spontaneous emission is most pronounced for low values of $\bar{\omega}$. However this is a key ingredient of all similar quantum interference effects [23] which may be depicted as follows: The smaller the field frequency is, the harder is it to distinguish between the various interference pathways, which leads to stronger quantum interference effects. As the scheme relies on the fact that photons with nonzero frequency may be exchanged during atomic transitions, the singular case of zero frequency, i.e. a static field, is excluded from our analysis as in [10].

In figure 6 the phase $\phi$ of the initial low-frequency field coherent state in equation (65) is varied. The chosen values are $\phi = -0.25; 0; 0.25; 0.5; 0.75; 1[\pi]$. The phase of the initial coherent state crucially influences the decay dynamics of the effective two-level system. By choosing the phase, the effective spontaneous decay varies between the usual decay rate $\tilde{\Gamma}_{21}$ and the maximum trapping for $\phi = 0; \pi$. This may be understood from equation (64) which shows that the decay dynamics of the state amplitude $E_n$ depends on the neighboring amplitudes $E_{n\pm m}$ ($m = 1, \ldots, 4$) which change relative to $E_n$ with varying phase $\phi$.

Figure 7 (a) shows that the initial photon distribution width of the coherent low-frequency laser field does not influence the result of the numerical calculation notably. This is a consistency check of the approximate coherent state in the ansatz for the wavefunction equation (65). The chosen values are $W = 100, 300, 500, 1000, 2000, 3000$; the other parameters are chosen as in figure 4 with $\Omega_{12} = 10^{13}$ Hz.

Figure 7 (b) shows that the number of photons of the coherent low-frequency laser field does not visibly influence the result of the numerical simulation. To evaluate the influence of the initial conditions, this value was chosen independent from the intensity of

**Figure 6.** Dependence of the population trapping on the phase of the initial low-frequency field coherent state. The chosen values are $\phi = -0.25; 0; 0.25; 0.5; 0.75; 1[\pi]$. Plots are (a) for $\phi = 0.5\pi$ and the dashed reference, (b) for $\phi = -0.25\pi; 0.75\pi$, (c) for $\phi = 0.25\pi$, and (d) for $\phi = 0; \pi$. 
Figure 7. Consistency checks of the simulation. (a) Dependence of the population trapping on the initial laser distribution width $W$. The chosen values are $W = 100, 300, 500, 1000, 2000, 3000$. (b) Dependence of the population trapping on the number of photons in the low-frequency mode considered in the simulation. The chosen values are $N = 10^4, 10^5, 10^6, 10^7, 10^8, 10^9$. (c) Dependence of the population trapping on the driving strength considered in the simulation. The chosen values are $\varrho = 10^{-5}; 10^{-4}; 10^{-3}; 10^{-2}; 10^{-1}; 1$. (d) Dependence of the population trapping on the number of simulated photon number states. The chosen values are $\Delta N = 3000, 5000, 8000, 10000, 12000, 15000$. In all subfigures, all plots but the dashed reference are on top of each other.

the low-frequency field. The chosen values are $N = 10^4, 10^5, 10^6, 10^7, 10^8, 10^9$; the other parameters are chosen as in figure 4 with $\Omega_{12} = 10^{13}$ Hz. Increasing $N$ broadens the initial photon number distribution. However if $N$ is large enough, neighboring photon number states have similar amplitudes up to a possible phase. This does not change notably for increasing $N$, and as the equation of motion for state amplitude $E_n$ depends on the neighboring amplitudes $E_{n\pm m}$ ($m = 1, \ldots, 4$) as discussed before, the results are independent of $N$ if it is not too small.

Figure 4 (c) shows the dependence of the population trapping on the damping factor $\varrho$ of the driving terms which was introduced in equation (68) to speed up the numerical calculations. The chosen values are $\varrho = 10^{-5}; 10^{-4}; 10^{-3}; 10^{-2}; 10^{-1}; 1$; the other parameters are chosen as in figure 4 with $\Omega_{12} = 10^{13}$ Hz. The Figure does not show a dependence on the damping of the driving strength. This may be explained along the lines of the interpretation of figures 4 (a) and (b), as these driving terms merely account for a broadening of the photon number distribution of the low-frequency field. The trapping mechanism however does not rely on a specific distribution width. Also, the driving terms are due to the same interactions as the ones which lead to the Stark shift discussed before. The last consistency check is shown in figure 4 (d), where the number of simulated photon number states $\Delta N$ is varied. The chosen values are $\Delta N = 3000, 5000, 8000, 10000, 12000, 15000$; the other parameters are chosen as in figure 4 with $\Omega_{12} = 10^{13}$ Hz. Again, there is no visible dependence on $\Delta N$ for the chosen values which shows that $\Delta N = 15000$ is large enough to eliminate errors due to border losses.
4. Summary

In summing up, we have derived an explicit expression for the multiphoton Hamiltonian describing the interaction of an atomic two-level system with both the vacuum field and an additional intense low-frequency laser field including up to three-photon processes. This Hamiltonian was used in a quantum mechanical simulation of the decay dynamics of a two level atom subject to the intense low-frequency laser field and the vacuum field modes. Using this simulation it was shown that the usual spontaneous decay found on one of the transitions in rubidium may be decelerated considerably by a suitably chosen low-frequency field.

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Appendix A. First order transition operators

In this appendix explicit expressions for the first-order transition operators calculated in section 2.1.3 are given. These transition operators may be written as

$$
\sigma_{ij}^{(1)} = \sigma_{ij}^{(0)} + A_{ij}^{22} + A_{ij}^{21} + \sum_{n \notin \{1,2\}} A_{ij}^{11}(n) + A_{ij}^{12}(n),
$$

$$
\sigma_{ij}^{(1)} = \sigma_{ij}^{(0)} + A_{ij}^{11} + A_{ij}^{12} + \sum_{n \notin \{1,2\}} A_{ij}^{22}(n) + A_{ij}^{21}(n).
$$

The corresponding operators $\sigma_{ij}^{(1)}$ and $\sigma_{ij}^{(1)}$ may be obtained by conjugation. The operators $A_{ij}^{kl}$ ($i, j = 1, 2$) are proportional to $\sigma_{kl}$. They are given by

$$
A_{ij}^{11}(n) = \mathcal{A}(1, n),
$$

where

$$
\mathcal{A}(\alpha, n) = \frac{\lambda_{jnk}^* \lambda_{nak} a_k^* a_k \sigma_{aa}}{(\omega_\alpha - \omega_j) (\omega_\alpha - \omega_n - \omega_k)} + \frac{\lambda_{nak} \lambda_{jnk}^* a_k^* a_k \sigma_{aa}}{(\omega_\alpha - \omega_j) (\omega_\alpha - \omega_n + \omega_k)} + \frac{\lambda_{jnk}^* \lambda_{nak} b_k^* b_k \sigma_{aa}}{(\omega_\alpha - \omega_j) (\omega_\alpha - \omega_n - \bar{\omega})} + \frac{\lambda_{nak} \lambda_{jnk}^* b_k^* b_k \sigma_{aa}}{(\omega_\alpha - \omega_j - 2\bar{\omega}) (\omega_\alpha - \omega_n - \bar{\omega})} + \frac{\lambda_{jnk}^* \lambda_{nak} a_k^* b_k^* \sigma_{aa}}{(\omega_\alpha - \omega_n - \omega_k) (\omega_\alpha - \omega_j - \omega_k - \bar{\omega})} + \frac{\lambda_{nak} \lambda_{jnk}^* a_k^* b_k^* \sigma_{aa}}{(\omega_\alpha - \omega_n - \bar{\omega}) (\omega_\alpha - \omega_j - \omega_k - \bar{\omega})} + \frac{\lambda_{jnk}^* \lambda_{nak} b_k^* a_k \sigma_{aa}}{(\omega_\alpha - \omega_n + \omega_k) (\omega_\alpha - \omega_j + \omega_k - \bar{\omega})} + \frac{\lambda_{nak} \lambda_{jnk}^* b_k^* a_k \sigma_{aa}}{(\omega_\alpha - \omega_n - \bar{\omega}) (\omega_\alpha - \omega_j + \omega_k - \bar{\omega})} + \frac{\lambda_{nak}^* \lambda_{jnk} a_k^* \sigma_{aa}}{(\omega_\alpha - \omega_j) (\omega_\alpha - \omega_n + \bar{\omega})} + \frac{\lambda_{jnk} a_k^* \lambda_{nak} \sigma_{aa}}{(\omega_\alpha - \omega_n - \omega_k) (\omega_\alpha - \omega_j - \omega_k + \bar{\omega})} + \frac{\lambda_{nak}^* \lambda_{jnk} a_k \sigma_{aa}}{(\omega_\alpha - \omega_n - \omega_k) (\omega_\alpha - \omega_j - \omega_k + \bar{\omega})} + \frac{\lambda_{jnk} \lambda_{nak} a_k \sigma_{aa}}{(\omega_\alpha - \omega_n - \omega_k) (\omega_\alpha - \omega_j - \omega_k + \bar{\omega})}.$$

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\[ A_{ij}^{22} = B(1,2), \quad (A.3) \]

where

\[ B(\alpha, \beta) = \frac{\lambda_{\beta\alpha} \lambda_{j\beta k} a_k a_k^\dagger \sigma_{\beta\beta}}{(\omega_\alpha - \omega_j)(\omega_\beta - \omega_j - \omega_k)} - \frac{\lambda_{j\beta k}^* \lambda_{\beta\alpha}^* a_k a_k^\dagger \sigma_{\beta\beta}}{(\omega_\alpha - \omega_j)(\omega_\beta - \omega_j - \omega_k)} \]

\[ - \frac{\lambda_{j\beta k}^* \lambda_{j\beta}^* b_j b_j^\dagger \sigma_{\beta\beta}}{(\omega_\beta - \omega_j - \omega_k)(\omega_\alpha - \omega_j - \omega_k - \omega)} \]

\[ - \frac{\lambda_{\beta\alpha} \lambda_{j\beta k} a_k a_k^\dagger \sigma_{\beta\beta}}{(\omega_\beta - \omega_j - \omega_k)(\omega_\alpha - \omega_j - \omega_k - \omega)} \]

\[ - \frac{\lambda_{j\beta k}^* \lambda_{j\beta}^* b_j b_j^\dagger \sigma_{\beta\beta}}{(\omega_\beta - \omega_j - \omega_k)(\omega_\alpha - \omega_j - \omega_k - \omega)} \]

\[ - \frac{\lambda_{\beta\alpha} \lambda_{j\beta k} a_k a_k^\dagger \sigma_{\beta\beta}}{(\omega_\beta - \omega_j - \omega_k)(\omega_\alpha - \omega_j - \omega_k - \omega)} \]

\[ - \frac{\lambda_{\beta\alpha} \lambda_{j\beta k} a_k a_k^\dagger \sigma_{\beta\beta}}{(\omega_\beta - \omega_j - \omega_k)(\omega_\alpha - \omega_j - \omega_k - \omega)} \]

\[ A_{ij}^{12}(n) = \frac{\lambda_{jnk}^* \lambda_{n2}^* b_j^\dagger a_k^\dagger \sigma_{12}}{(\omega_1 - \omega_j)(\omega_1 - \omega_n + \omega_k)} + \frac{\lambda_{jnk} \lambda_{n2}^* b_j^\dagger a_k^\dagger \sigma_{12}}{(\omega_1 - \omega_j)(\omega_1 - \omega_n + \omega_k)} \]

\[ + \frac{\lambda_{jn}^* \lambda_{n2}^* b_j b_j^\dagger \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2}^* b_j b_j^\dagger \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* a_k a_k^\dagger \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* a_k a_k^\dagger \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ \quad + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ \quad + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ \quad + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ \quad + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]

\[ \quad + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} + \frac{\lambda_{jn} \lambda_{n2} \lambda_{n2}^* \sigma_{12}}{(\omega_1 - \omega_n + \omega_k)(\omega_1 - \omega_j + \omega_k - \omega)} \]
Spontaneous-emission suppression via multiphoton quantum interference

\[ A_{2j}^{11} = B(2, 1), \]

\[ A_{2j}^{22}(n) = A(2, n), \]

\[ A_{2j}^{12} = \frac{\lambda_{12k}^* \lambda_{2j1}^* a_k^\dagger b^j \sigma_{21}}{(\omega_1 - \omega_j) (\omega_1 - \omega_j + \omega_k)} - \frac{\lambda_{12k} \lambda_{2j1}^* a_k b^j \sigma_{21}}{(\omega_1 - \omega_j - \omega_k)(\omega_2 - \omega_j - 2\omega_k)} \]

\[ - \frac{\lambda_{12k} \lambda_{2j1}^* a_k^\dagger b^j \sigma_{21}}{(\omega_1 - \omega_j - 2\omega_k)(\omega_2 - \omega_j - 2\omega_k)} - \frac{\lambda_{21k} \lambda_{1j1}^* a_k b^j \sigma_{21}}{(\omega_1 - \omega_j)(\omega_2 - \omega_j - \omega_k - 2\omega)} \]

\[ A_{2j}^{2j}(n) = \frac{\lambda_{jnk} \lambda_{n1} b a_k \sigma_{21}}{(\omega_2 - \omega_j)(\omega_2 - \omega_j - \omega_k)} + \frac{\lambda_{n1} \lambda_{jnk}^* b a_k^\dagger \sigma_{21}}{(\omega_2 - \omega_j - 2\omega_k)(\omega_2 - \omega_j - \omega_k)} \]

\[ + \frac{\lambda_{jn1}^* \lambda_{jn1}^* b^j b^j \sigma_{21}}{(\omega_2 - \omega_j - \omega_k - 3\omega)(\omega_2 - \omega_j - \omega_k - 2\omega)} + \frac{\lambda_{jn1} \lambda_{jn1}^* b^j a_k \sigma_{21}}{(\omega_2 - \omega_j - 2\omega)(\omega_2 - \omega_j - \omega_k - 2\omega)} \]

(A.6)

(A.7)

(A.8)
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