

Matrix perturbation theory for driven three-level systems with damping

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We investigate the dynamics of the Λ system driven by two resonant laser fields in presence of dissipation for coupling strengths where the rotating-wave approximation starts to break down. This regime is characterised by Rabi frequencies being approximately equal or smaller than the field frequencies. A systematic procedure to obtain an expansion for the solution of the Bloch evolution equations of the system is presented. The lowest contribution results to be the well-known rotating-wave approximation. The method is based on a semi-classical treatment of the problem, and its predictions are interpreted fully quantum mechanically. The theory is illustrated by a detailed study of the disappearance of coherent population trapping as the intensities of the fields increase.

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1 Introduction

In comparison to the two-level atom, the three-level atom exhibits a much richer variety of effects in its interaction with the electromagnetic field, with examples such as lasing without inversion, laser cooling, population transfer and loss-free pulse propagation. This broader range of processes arises as a result of coherence among the states, as induced by (classical) radiation as well as quantum interference. Typical examples are Λ systems, in which two closely spaced long-lived levels (“ground states”) are coupled by two nearly resonant laser fields to a third distant short-lived level (excited state). If the frequency difference of the laser fields is close to the atomic splitting of the two ground states (Raman resonance), the excited state spontaneously decays into a coherent superposition of the two closely spaced levels [1]. The system then cannot be excited by the applied light and remains in this *trapped* (or *dark*) state, which in fact is a *stationary* state of the dressed atom-plus-field system.

Most of these coherent phenomena can be adequately treated within the limits imposed by the rotating-wave approximation (RWA) [2]. This approximation gives a good description when the Rabi frequencies of each field are much smaller than their respective field frequencies, e.g. $\Omega_1, \Omega_2 \ll \omega_1, \omega_2$, the so-called weak-field regime. In this paper, we explore the response of the Λ system to the two laser fields beyond the RWA in a relatively strong-field regime defined by the Rabi frequencies being approximately equal or smaller than the field frequencies, e.g. $\Omega_1, \Omega_2 \lesssim \omega_1, \omega_2$. We show how the trapping effect gradually loses its definition up to its disappearance as the intensities of the fields increase.

Our primary motivation for this study is the extension of concepts from quantum optics into mesoscopic condensed matter physics. To be specific, various authors [3–7] have identified three-level systems as

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promising candidates to utilise coherent control [8] in solid state structures such as semiconductor quantum dots. In these systems, dark states and coherent population trapping can be combined with the tunnelling of quasi-particles (electrons, or electrons and holes) which, e.g., leads to the appearance of quantum optical coherent phenomena in electronic transport. In these situations, it is then not clear *a priori* that weak coupling approximations like the RWA always hold. Second, for strong coupling it has been known for a while that time-dependent fields can lead to coherent suppression of tunnelling, which is usually described in a (single or multi-mode) Floquet state picture [9]. This has led to an enormous activity in applications to systems driven by strong time-dependent fields, such as electron transport and tunnelling in semiconductor structures under AC radiation [10].

In three-level systems in intense bichromatic fields, non-RWA effects can be interpreted as a breaking of a Gell-Mann-type $SU(3)$ dynamical symmetry. Ho and Chu [11] have shown how to use the $SU(3)$ generators and many-mode Floquet theory in order to obtain perturbative results in absence of dissipation. Matisov, Mazets and Windholz [12] used a direct integration of the Schrödinger equation to discuss the coherent time-evolution under strong fields. Furthermore, coherent suppression of tunneling in coherent three-level systems was recently investigated by Unanyan, Guérin, and Jauslin [13].

We have developed a formalism for systematic expansions both around the weak-coupling (RWA) limit, and the strong-coupling regime of dissipative three-level systems. Since the technical treatment is quite different for these two cases, in the present paper we will only concentrate on a systematic perturbation theory in the weak-coupling regime and discuss the strong coupling case in a separate publication. The matrix perturbation scheme developed here will allow us to solve all stationary quantities and find, e.g., systematic corrections to the RWA for the excited state population profile.

The paper is organised as follows. In Sect. 2, we introduce the model and the equations of motion. In Sect. 3, we re-derive the RWA for the three-level system. In Sect. 4, we present the formalism for the expansion. Sect. 5 is devoted to the display and explanation of the results, and the extended appendices A-E contain detailed derivations of the equations.

2 Model and evolution equations

We start from the Hamiltonian

$$H(t) = H_0 + \mathbf{d} \cdot \sum_{j=1,2} \mathbf{e}_j \mathcal{E}_j \cos(\omega_j t), \quad (1)$$

where H_0 is the Hamiltonian of the free three-level system $\{|1\rangle, |2\rangle, |0\rangle\}$ of energies E_1, E_2, E_0 , and \mathbf{d} is the dipole moment operator (coupling transition 1–0 by the action of field 1 and transition 2–0 by the action of field 2). The total electric field containing the two main frequencies $\underline{\omega} = (\omega_1, \omega_2)$ is characterised by the unit vector \mathbf{e}_j , and real-valued amplitudes $\underline{\mathcal{E}} = (\mathcal{E}_1, \mathcal{E}_2)$. We consider a Λ -configuration, Fig. 1, with relaxation processes due to spontaneous emission at rates Γ_i^{sp} ($i = 0, 1, 2$), where $\Gamma_0 \approx \Gamma_0^{sp}$, $\Gamma_{0 \rightarrow 1} \approx \Gamma_{0 \rightarrow 2} \approx \Gamma_0^{sp}/2$ and $\Gamma_{01} = \Gamma_{01} \approx \Gamma_{02} = \Gamma_{02} \approx \Gamma_0^{sp}/2$. The detuning of the two fields (frequencies ω_1 and ω_2 , Rabi frequencies Ω_1 and Ω_2) with respect to their respective resonance condition are measured by the parameter $\delta_1 = \omega_1 - \omega_{01}$ and $\delta_2 = \omega_2 - \omega_{02}$ where $\omega_{0j} = (E_0 - E_j)/\hbar$ with $j = 1, 2$. The Raman detuning δ_R from the Raman two photon-resonance condition is defined as the relative detuning of both fields as $\delta_R = \delta_1 - \delta_2$. Specifically we investigate how the mean value of the occupation of the upper level, $\langle \rho_{00}(t \rightarrow \infty) \rangle$, when the system relaxes at $t \rightarrow \infty$, varies when the two field frequencies are scanned around the Raman resonance value, $\delta_R = 0$. We devise a perturbative method to estimate this physical quantity, where the RWA results to be the approximation of order zero.

The evolution of the system is governed by the optical Bloch evolution equations,

$$\frac{d\rho(t)}{dt} = \frac{1}{i\hbar} [H(t), \rho(t)] + \mathcal{R}\rho(t), \quad (2)$$

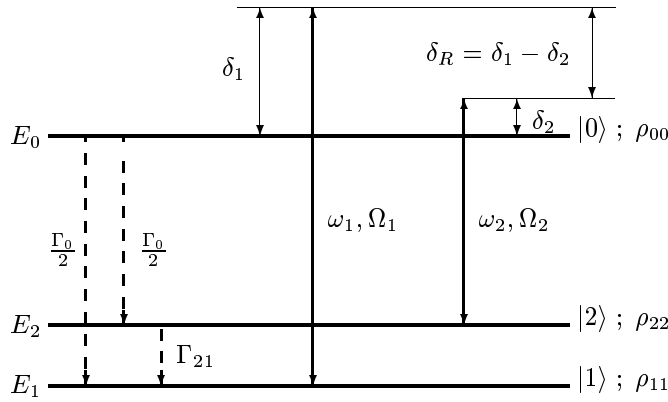


Fig. 1 The parameters involved in the calculation. The two monochromatic fields are defined by their respective frequencies ω_1 and ω_2 , and their respective intensities or Rabi frequencies Ω_1 and Ω_2 . The system is characterised by the three Bohr frequencies ω_{01} , ω_{02} and ω_{21} , and the rate of population decay Γ_0 from the excited-level and the decay rate $\Gamma_{12} = \Gamma_{21}$ for the coherences between the two lower levels. The optical detuning of the two fields from their respective optical resonances is given by $\delta_1 = \omega_1 - \omega_{01}$ and $\delta_2 = \omega_2 - \omega_{02}$ and their relative difference is the Raman detuning $\delta_R = \delta_1 - \delta_2$.

which explicitly read

$$\begin{aligned}
 \frac{d\rho_{00}}{dt} &= -\Gamma_0\rho_{00} + i\Omega_1 \cos(\omega_1 t)\rho_{01} + i\Omega_2 \cos(\omega_2 t)\rho_{02} - i\Omega_1 \cos(\omega_1 t)\rho_{10} - i\Omega_2 \cos(\omega_2 t)\rho_{20} \\
 \frac{d\rho_{11}}{dt} &= \frac{\Gamma_0}{2}\rho_{00} - i\Omega_1 \cos(\omega_1 t)\rho_{01} + i\Omega_1 \cos(\omega_1 t)\rho_{10} \\
 \frac{d\rho_{22}}{dt} &= \frac{\Gamma_0}{2}\rho_{00} - i\Omega_2 \cos(\omega_2 t)\rho_{02} + i\Omega_2 \cos(\omega_2 t)\rho_{20} \\
 \frac{d\rho_{01}}{dt} &= -\left(\frac{\Gamma_0}{2} + i\omega_{01}\right)\rho_{01} + i\Omega_1 \cos(\omega_1 t)(\rho_{00} - \rho_{11}) - i\Omega_2 \cos(\omega_2 t)\rho_{21} \\
 \frac{d\rho_{02}}{dt} &= -\left(\frac{\Gamma_0}{2} + i\omega_{02}\right)\rho_{02} + i\Omega_2 \cos(\omega_2 t)(\rho_{00} - \rho_{22}) - i\Omega_1 \cos(\omega_1 t)\rho_{12} \\
 \frac{d\rho_{21}}{dt} &= -(\Gamma_{21} + i\omega_{21})\rho_{21} + i\Omega_1 \cos(\omega_1 t)\rho_{20} - i\Omega_2 \cos(\omega_2 t)\rho_{01},
 \end{aligned} \tag{3}$$

with $\rho_{ij}^* = \rho_{ji}$ and where ω_{ij} are the transition frequencies of the system. The Rabi frequencies Ω_1 and Ω_2 , which are proportional to the intensity of each respective field, are defined as follows

$$\Omega_1 = \frac{-\mathbf{d}_{01} \cdot \mathbf{e}_1 \cdot \mathcal{E}_1}{\hbar}, \quad \Omega_2 = \frac{-\mathbf{d}_{02} \cdot \mathbf{e}_2 \cdot \mathcal{E}_2}{\hbar}, \tag{4}$$

where \mathbf{d}_{0j} are the matrix elements $\langle 0|\mathbf{d}|j\rangle$ of the dipole moment operator $\mathbf{d} = e\mathbf{r}$. There are no diagonal elements of \mathbf{d} because it is a vector operator and thus has odd parity and the states $|i\rangle$ are assumed to have definite parity. *A priori* the values of these Rabi frequencies are complex numbers. However, if the eigen-states $|i\rangle$ correspond, for example, to three different orbital states of same quantum magnetic number m , it can be shown that, by adjusting appropriately the phase of these eigen-states, eq. (4) yields real values for the Rabi frequencies. The consideration of only $\Delta m = 0$ transitions is consistent with the fact that the system is driven by linear polarised fields in this work.

3 Rotating-wave approximation

Let us first re-derive the Rotating-Wave Approximation (RWA) for the three-level system. Throughout this section, we assume that both the coupling to the external fields and the damping are small and that we are close to resonance, i.e.

$$\Omega_1, \Omega_2, \Gamma_0, \Gamma_{21}, |\omega_1 - \omega_{01}|, |\omega_2 - \omega_{02}| \ll \omega_1 \sim \omega_2. \tag{5}$$

The RWA consists of neglecting the non-resonant part of the fields and resolving the equations of motion in the *transformed picture*. The problem thus being simply the evolution of the free *dressed system* becomes stationary and can easily be back-transformed into the original picture ('Laboratory frame').

3.1 Reference frame transformation

For weak coupling ($\Omega_1 \ll \omega_{01}$ and $\Omega_2 \ll \omega_{02}$), the representation where the fields can be interpreted as a relatively weak non-resonant force, corresponds to the unitary operator

$$U(t) = \begin{bmatrix} 1 & 0 & 0 \\ 0 & e^{-i(\omega_1 - \omega_2)t} & 0 \\ 0 & 0 & e^{-i\omega_1 t} \end{bmatrix}, \quad (6)$$

which is written with respect to the eigen-state basis $\{|1\rangle, |2\rangle, |0\rangle\}$. This yields

$$\frac{d\tilde{\rho}(t)}{dt} = \frac{1}{i\hbar} [\tilde{H}(t), \tilde{\rho}(t)] + \tilde{\mathcal{R}}\tilde{\rho}(t), \quad (7)$$

where we introduced

$$\tilde{\rho} = U^+ \rho U, \quad \tilde{H}(t) = -i\hbar U^+ \frac{\partial U}{\partial t} + U^+ H(t) U, \quad (8)$$

and the relaxation operator remains unchanged, $\tilde{\mathcal{R}} = \mathcal{R}$. Note that in the case of a *two-level system* interacting with one monochromatic field, this transformation has a simple geometrical interpretation in terms of a 'rotating frame'. It is now easy to show that the explicit form of the transformed Hamiltonian $\tilde{H}(t)$ is (we set $E_1 = 0$ for convenience)

$$\tilde{H}(t) = \hbar \begin{bmatrix} 0 & 0 & \frac{\Omega_1}{2}(1 + e^{-i2\omega_1 t}) \\ 0 & -\delta_1 + \delta_2 & \frac{\Omega_2}{2}(1 + e^{-i2\omega_2 t}) \\ \frac{\Omega_1}{2}(1 + e^{i2\omega_1 t}) & \frac{\Omega_2}{2}(1 + e^{i2\omega_2 t}) & -\delta_1 \end{bmatrix},$$

where $\delta_j = \omega_j - \omega_{0j}$ for $j = 1, 2$ are the detuning of each field from their respective optical resonances.

In the RWA, one now neglects the oscillating part of this Hamiltonian which is replaced by

$$\tilde{H}_{rwa} = \hbar \begin{bmatrix} 0 & 0 & \frac{\Omega_1}{2} \\ 0 & -\delta_1 + \delta_2 & \frac{\Omega_2}{2} \\ \frac{\Omega_1}{2} & \frac{\Omega_2}{2} & -\delta_1 \end{bmatrix}. \quad (9)$$

This time-independent Hamiltonian \tilde{H}_{rwa} can be interpreted as the mathematical description of the system as seen from the transformed picture. The approximate evolution is dictated by eq. (7) with $\tilde{H}(t)$ replaced by \tilde{H}_{rwa} . Thus, the problem has been reduced to the evolution of the free *dressed system* with the same relaxation mechanisms. As a stationary problem, it can easily be solved analytically.

3.2 Validity of the RWA

In the Laboratory frame, the effect of the driving fields cannot be ignored although they constitute a weak perturbation there ($\Omega_1 \ll \omega_{01}$ and $\Omega_2 \ll \omega_{02}$). This is because they are at near resonance with the system ($\omega_1 \sim \omega_{01}$ and $\omega_2 \sim \omega_{02}$). In other words, the 'small force' is in phase with the natural oscillations of the system and it has an accumulative effect that can considerably change the state of the 'heavy system' with little effort.

However, this is in contrast with the situation as it is perceived in the transformed picture. There, for example, the natural oscillations of the dressed system are much slower. For example, at Raman resonance condition, $\delta_R = \delta_1 - \delta_2 = 0$, a simple diagonalisation of the Hamiltonian \tilde{H}_{rwa} shows that the values of the characteristic frequencies of the dressed system are $\lambda_1 = \sqrt{\delta_1^2 + \Omega_1^2 + \Omega_2^2}$ and $\lambda_{2,3} = (\sqrt{\delta_1^2 + \Omega_1^2 + \Omega_2^2} \pm \delta_1)/2$, which are much smaller than ω_{01} or ω_{02} in situations of near optical resonance ($\delta_1 \ll \omega_{01}$ and $\delta_2 \ll \omega_{02}$) and when the coupling is weak ($\Omega_1 \ll \omega_{01}$ and $\Omega_2 \ll \omega_{02}$). On the other hand, the frequencies of the varying perturbation discerned in the transformed picture are $2\omega_1$ and $2\omega_2$ as the oscillating part of the Hamiltonian $\tilde{H}(t)$ indicates. These two facts show that the driving force, which is also not too strong compared with the resistance of the dressed system to alter its own dynamics (the magnitudes of the oscillating and static parts of $\tilde{H}(t)$ are of the same order), is far out of resonance, e.g. $\lambda_{1,2,3} \ll 2\omega_{1,2}$ since $\omega_{1,2} \sim \omega_{01} \sim \omega_{02}$ at near optical resonance, and consequently it can be safely neglected in this approximation.

The stationary state $\tilde{\rho}^{asy}$ can be transformed back to the Laboratory frame,

$$\rho(t \rightarrow \infty) = U(t \rightarrow \infty) \tilde{\rho}^{asy} U^\dagger(t \rightarrow \infty), \quad (10)$$

which is particularly simple if we are interested in the population $\rho_{00}(t \rightarrow \infty)$. After straightforward algebra, one obtains

$$\begin{aligned} \langle \rho_{00}(t \rightarrow \infty) \rangle_{rwa} = & \Omega^2 [\Omega^2 \Gamma_{12} + \Gamma_0 (\Gamma_{12}^2 + \delta_R^2)] \\ & / \left\{ 2\Omega^2 \Gamma_0^2 \Gamma_{12} + \Omega^2 \Gamma_{12} [3\Omega^2 + (\delta_1 + \delta_2)^2] + \Gamma_0^3 (\Gamma_{12}^2 + \delta_R^2) \right. \\ & \left. + \Gamma_0 [\Omega^4 + 3\Omega^2 \Gamma_{12}^2 + 2\Gamma_{12}^2 (\delta_1^2 + \delta_2^2) - 2\Omega^2 \delta_R^2 + (3\Omega^2 + 2\delta_1^2 + 2\delta_2^2) \delta_R^2] \right\}, \end{aligned} \quad (11)$$

where we have chosen $\Omega_1 = \Omega_2 = \Omega$ for simplicity.

4 An expansion scheme for the RWA

In this section, we present a systematic expansion, which yields the rotating-wave approximation as the lowest order contribution. We notice that $\rho_{00} = 1 - \rho_{11} - \rho_{22}$, and perform the following manipulations explicitly only for ρ_{11} , ρ_{01} , and ρ_{21} . The other matrix elements are easily obtained by similar operations and change of indices.

We now consider $\rho_{ij}(t)$ in eqs. (3) as functions depending on t in an indirect way through two intermediate time variables $t_1(t)$ and $t_2(t)$, e.g. $\rho_{ij}(t) = \rho_{ij}[t_1(t), t_2(t)]$. If we choose t_1 and t_2 as $t_1 = t_1(t) = t$ and $t_2 = t_2(t) = t$, we find that the rate of change of a generic matrix element can be written as

$$\frac{d\rho_{ij}}{dt} = \left(\frac{\partial \rho_{ij}}{\partial t_1} + \frac{\partial \rho_{ij}}{\partial t_2} \right)_{t_1=t_2=t}, \quad (12)$$

where we have applied the chain rule and used the fact that $\partial t_1 / \partial t = \partial t_2 / \partial t = 1$.

This means, e.g.,

$$\frac{\partial \rho_{11}}{\partial t_1} + \frac{\partial \rho_{11}}{\partial t_2} = \frac{\Gamma_0}{2} (1 - \rho_{11} - \rho_{22}) - i\Omega_1 \cos(\omega_1 t_1) \rho_{01} + i\Omega_1 \cos(\omega_1 t_1) \rho_{10} \quad (13)$$

and similar for the other matrix elements. Equivalence to the original equations is achieved if and only if the constraint

$$t_1 = t_2 = t \quad (14)$$

holds. In the following, we consider the variables t_1 and t_2 as two independent variables, thus obtaining a set of *partial* differential equations, and only set $t_1 = t_2 = t$ at the end. Expanding each cosine, we find

$$\begin{aligned}
D\rho_{11} &= \underline{-\frac{i\Omega_1}{2}e^{-i\omega_1 t_1}\rho_{01}} + \underline{\frac{i\Omega_1}{2}e^{i\omega_1 t_1}\rho_{10}} + \frac{\Gamma_o}{2} - \frac{\Gamma_o}{2}\rho_{11} - \frac{\Gamma_o}{2}\rho_{22} - \frac{i\Omega_1}{2}e^{i\omega_1 t_1}\rho_{01} \\
&\quad + \frac{i\Omega_1}{2}e^{-i\omega_1 t_1}\rho_{10}, \\
D\rho_{01} &= \underline{-i\Omega_1 e^{i\omega_1 t_1}\rho_{11}} - \underline{\frac{i\Omega_1}{2}e^{i\omega_1 t_1}\rho_{22}} - \underline{\frac{i\Omega_2}{2}e^{i\omega_2 t_2}\rho_{21}} + \underline{\frac{i\Omega_1}{2}e^{i\omega_1 t_1}} - \left(\frac{\Gamma_o}{2} + i\omega_{01}\right)\rho_{01} \\
&\quad - i\Omega_1 e^{-i\omega_1 t_1}\rho_{11} - \underline{\frac{i\Omega_1}{2}e^{-i\omega_1 t_1}\rho_{22}} - \underline{\frac{i\Omega_2}{2}e^{-i\omega_2 t_2}\rho_{21}} + \frac{i\Omega_1}{2}e^{-i\omega_1 t_1}, \\
D\rho_{21} &= \underline{\frac{i\Omega_1}{2}e^{i\omega_1 t_1}\rho_{20}} - \underline{\frac{i\Omega_2}{2}e^{-i\omega_2 t_2}\rho_{01}} - (\Gamma_{21} + i\omega_{21})\rho_{21} + \frac{i\Omega_1}{2}e^{-i\omega_1 t_1}\rho_{20} \\
&\quad - \frac{i\Omega_2}{2}e^{i\omega_2 t_2}\rho_{01},
\end{aligned} \tag{15}$$

where the underlined terms are those neglected in the RWA and similar equations hold for the remaining matrix elements. The operator D is the differential operator $D = \partial/\partial t_1 + \partial/\partial t_2$.

4.1 Double Laplace transform

We now transform the partial differential eqs. (15) into ordinary algebraic equations by using the double Laplace transform,

$$\rho_{ij}(z_1, z_2) = \int_0^\infty dt_1 \int_0^\infty dt_2 e^{-z_1 t_1 - z_2 t_2} \rho_{ij}(t_1, t_2), \tag{16}$$

where we have distinguished the direct from the transformed functions by the use of the independent variables as their arguments. Let us abbreviate this double integral by using the symbol \mathcal{L} , and write $\rho_{ij}(z_1, z_2) = \mathcal{L}\rho_{ij}(t_1, t_2)$. On the left-hand side, the expression to transform is always of the form $D\rho_{ij} = \partial\rho_{ij}/\partial t_1 + \partial\rho_{ij}/\partial t_2$. Thus, recalling the relations between the Laplace transforms of derivatives of functions, we have

$$\mathcal{L}D\rho_{ij}(t_1, t_2) = z_1\rho_{ij}(z_1, z_2) + z_2\rho_{ij}(z_1, z_2) - \rho_{ij}(t_1 = 0, z_2) - \rho_{ij}(z_1, t_2 = 0) \tag{17}$$

where, for example, $\rho_{ij}(t_1 = 0, z_2)$ is the normal Laplace transform of the function $\rho_{ij}(t_1 = 0, t_2)$. This expression can be further simplified if the boundary conditions $\rho_{ij}(t_1 = 0, t_2) = 0$ and $\rho_{ij}(t_1, t_2 = 0) = 0$ are chosen. In that case, we have

$$\mathcal{L}D\rho_{ij} = z_1\rho_{ij}(z_1, z_2) + z_2\rho_{ij}(z_1, z_2). \tag{18}$$

We mention that there remains an arbitrariness in the choice of the boundary conditions when one is not interested in the transient behaviour of the system as is the case in this work. This is due to the fact that we are not interested in the function $\rho_{ij}(t_1, t_2)$ for all the values of t_1 and t_2 . The asymptotic behaviour of the system as $t \rightarrow \infty$, however, does not depend on this choice.

After shifting the independent variables z_1 and z_2 , we finally obtain the following equations,

$$\begin{aligned}
&\underline{\frac{i\Omega_1}{2}\tilde{\rho}_{01}(z_1 + i2\omega_1, z_2)} - \underline{\frac{i\Omega_1}{2}\tilde{\rho}_{10}(z_1 - i2\omega_1, z_2)} + \left(z_1 + z_2 + \frac{\Gamma_o}{2}\right)\tilde{\rho}_{11}(z_1, z_2) \\
&+ \frac{\Gamma_o}{2}\tilde{\rho}_{22}(z_1, z_2) + \frac{i\Omega_1}{2}\tilde{\rho}_{01}(z_1, z_2) - \frac{i\Omega_1}{2}\tilde{\rho}_{10}(z_1, z_2) = \frac{\Gamma_o}{2z_1 z_2},
\end{aligned}$$

$$\begin{aligned}
& \frac{i\Omega_1 \tilde{\rho}_{11}(z_1 - i2\omega_1, z_2) + \frac{i\Omega_1}{2} \tilde{\rho}_{22}(z_1 - i2\omega_1, z_2) + \frac{i\Omega_2}{2} \tilde{\rho}_{21}(z_1, z_2 - i2\omega_2)}{2} \\
& + \left(z_1 + z_2 + \frac{\Gamma_o}{2} - i\delta_1 \right) \tilde{\rho}_{01}(z_1, z_2) \\
& + i\Omega_1 \tilde{\rho}_{11}(z_1, z_2) + \frac{i\Omega_1}{2} \tilde{\rho}_{22}(z_1, z_2) + \frac{i\Omega_2}{2} \tilde{\rho}_{21}(z_1, z_2) = \frac{i\Omega_1}{2z_2} \frac{1}{(z_1 - i2\omega_1)} + \frac{i\Omega_1}{2z_2 z_2}, \\
& - \frac{i\Omega_1}{2} \tilde{\rho}_{20}(z_1 - i2\omega_1, z_2) + \frac{i\Omega_2}{2} \tilde{\rho}_{01}(z_1, z_2 + i2\omega_2) \\
& + (z_1 + z_2 + \Gamma_{21} - i\delta_R) \tilde{\rho}_{21}(z_1, z_2) - \frac{i\Omega_1}{2} \tilde{\rho}_{20}(z_1, z_2) + \frac{i\Omega_2}{2} \tilde{\rho}_{01}(z_1, z_2) = 0,
\end{aligned} \tag{19}$$

where we have also re-defined the dependent variables as $\tilde{\rho}_{jj}(z_1, z_2) = \rho_{jj}(z_1, z_2)$, $\tilde{\rho}_{01}(z_1, z_2) = \rho_{01}(z_1 - i\omega_1, z_2)$, $\tilde{\rho}_{10}(z_1, z_2) = \rho_{10}(z_1 + i\omega_1, z_2)$, $\tilde{\rho}_{21}(z_1, z_2) = \rho_{21}(z_1 - i\omega_1, z_2 + i\omega_2)$ and $\tilde{\rho}_{20}(z_1, z_2) = \rho_{20}(z_1, z_2 + i\omega_2)$. The parameter $\delta_1 = \omega_1 - \omega_{01}$ is the detuning of the laser 1 from its resonance, and $\delta_R = \delta_1 - \delta_2 = \omega_1 - \omega_2 - \omega_{01} + \omega_{02}$ is the Raman detuning from the two-photon resonance. Note that we have also arranged the unknowns to be on the left-hand side.

This shift in the Laplace domain corresponds to the *transformed picture* introduced in the previous section when working in real space. In fact, eqs. (19) are just the double Laplace transform of the matrix elements of the operator eq. (7) after introducing the times t_1 and t_2 . Thus the dependent variables in eqs. (19) are simply $\tilde{\rho}(z_1, z_2) = \mathcal{L}\tilde{\rho}(t_1, t_2)$ where $\tilde{\rho}(t) = \tilde{\rho}(t_1 = t, t_2 = t)$.

4.2 Matrix perturbation theory

The full set of equations for the density operator in the Laplace domain can be written as

$$\begin{aligned}
& \bar{\alpha}_1 \bar{\rho}(z_1 - i2\omega_1, z_2) + \bar{\alpha}_2 \bar{\rho}(z_1, z_2 - i2\omega_2) \\
& + \bar{\pi}(z_1, z_2) \bar{\rho}(z_1, z_2) + \bar{\beta}_1 \bar{\rho}(z_1 + i2\omega_1, z_2) + \bar{\beta}_2 \bar{\rho}(z_1, z_2 + i2\omega_2) = \bar{\chi}(z_1, z_2),
\end{aligned} \tag{20}$$

where $\bar{\alpha}_1$, $\bar{\alpha}_2$, $\bar{\beta}_1$, $\bar{\beta}_2$, $\bar{\rho}(z_1, z_2)$ and $\bar{\chi}(z_1, z_2)$ are the matrices defined in Appendix A, and $\bar{\pi}(z_1, z_2)$ is the square matrix in eq. (21) below. Eq. (20) is our main result; in fact it is a short-hand notation for an infinite number of coupled equations for the Laplace components of the density operator.

4.2.1 Lowest order truncation: RWA

The two first terms and the last two on the left-hand side of (20) are made up solely of the underlined terms of eqs. (19), which are neglected in the RWA. Therefore, since the matrices $\bar{\alpha}_j$, $\bar{\beta}_j$ and $\bar{\pi}(z_1, z_2)$ are of the same order of magnitude, the RWA neglects $\bar{\rho}(z_1 \pm i2\omega_1, z_2)$ and $\bar{\rho}(z_1, z_2 \pm i2\omega_2)$ when compared with $\bar{\rho}(z_1, z_2)$ so that eq. (20) becomes immediately solvable. This suggests that the function $|\bar{\rho}(z_1, z_2)|$ is centred around the point $(z_1 = 0, z_2 = 0)$. Note that we are only interested in $\bar{\rho}(z_1, z_2)$ for $z_1 \rightarrow 0$ and $z_2 \rightarrow 0$ for the stationary behaviour of the system. Then, in this zeroth-order approximation, we consider $|\bar{\rho}(z_1 \pm i2\omega_1, z_2)|$, $|\bar{\rho}(z_1, z_2 \pm i2\omega_2)| \ll |\bar{\rho}(z_1, z_2)|$, and assumed that $\bar{\rho}(z_1 \pm i2\omega_1, z_2) \sim \bar{\rho}(z_1, z_2 \pm i2\omega_2) \sim 0$. With these

simplifications, the equation to solve becomes

$$\begin{bmatrix} (z_1+z_2+\frac{\Gamma_o}{2}) & \frac{\Gamma_o}{2} & \frac{i\Omega_1}{2} & 0 & 0 & \frac{-i\Omega_1}{2} & 0 & 0 \\ \frac{\Gamma_o}{2} & (z_1+z_2+\frac{\Gamma_o}{2}) & 0 & \frac{i\Omega_2}{2} & 0 & 0 & \frac{-i\Omega_2}{2} & 0 \\ i\Omega_1 & \frac{i\Omega_1}{2} & (z_1+z_2+\frac{\Gamma_o}{2}-i\delta_1) & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 \\ \frac{i\Omega}{2} & i\Omega_2 & 0 & (z_1+z_2+\frac{\Gamma_o}{2}-i\delta_R) & 0 & 0 & 0 & \frac{i\Omega_1}{2} \\ 0 & 0 & \frac{i\Omega_2}{2} & 0 & (z_1+z_2+\Gamma_{21}-i\delta_R) & 0 & \frac{-i\Omega_1}{2} & 0 \\ -i\Omega_1 & -\frac{i\Omega_1}{2} & 0 & 0 & 0 & (z_1+z_2+\frac{\Gamma_o}{2}+i\delta_1) & 0 & -\frac{i\Omega_2}{2} \\ -\frac{i\Omega_2}{2} & -i\Omega_2 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 & (z_1+z_2+\frac{\Gamma_o}{2}+i\delta_2) & 0 \\ 0 & 0 & 0 & \frac{i\Omega_1}{2} & 0 & -\frac{i\Omega_2}{2} & 0 & (z_1+z_2+\Gamma_{21}+i\delta_R) \end{bmatrix} \times \begin{bmatrix} \tilde{\rho}_{11}(z_1, z_2) \\ \tilde{\rho}_{22}(z_1, z_2) \\ \tilde{\rho}_{01}(z_1, z_2) \\ \tilde{\rho}_{02}(z_1, z_2) \\ \tilde{\rho}_{21}(z_1, z_2) \\ \tilde{\rho}_{10}(z_1, z_2) \\ \tilde{\rho}_{20}(z_1, z_2) \\ \tilde{\rho}_{12}(z_1, z_2) \end{bmatrix} = \frac{1}{z_1 z_2} \begin{bmatrix} \frac{\Gamma_o}{2} \\ \frac{\Gamma_o}{2} \\ \frac{i\Omega_1}{2} \\ \frac{i\Omega_2}{2} \\ 0 \\ -\frac{i\Omega_1}{2} \\ -\frac{i\Omega_2}{2} \\ 0 \end{bmatrix}. \quad (21)$$

which is the RWA equation in Laplace space.

4.2.2 Beyond the RWA

We now can go beyond the RWA in a systematic truncation scheme which we explain in the following. For example, in the first order approximation beyond RWA, we will not longer neglect $\bar{\rho}(z_1 \pm i2\omega_1, z_2)$ and $\bar{\rho}(z_1, z_2 \pm i2\omega_2)$ when compared with $\bar{\rho}(z_1, z_2)$ in eq. (20). We can obtain the additional equations for these variables by shifting the independent variables $z_1 \rightarrow z_1 \pm i2\omega_1$ and $z_2 \rightarrow z_2 \pm i2\omega_2$ in the main eq. (20). For example, the shift $z_1 \rightarrow z_1 - i2\omega_1$, $z_2 \rightarrow z_2$ yields the following additional equation

$$\begin{aligned} \bar{\alpha}_1 \bar{\rho}(z_1 - i4\omega_1, z_2) + \bar{\alpha}_2 \bar{\rho}(z_1 - i2\omega_1, z_2 - i2\omega_2) + \bar{\pi}(z_1 - i2\omega_1, z_2) \bar{\rho}(z_1 - i2\omega_1, z_2) \\ + \bar{\beta}_1 \bar{\rho}(z_1, z_2) + \bar{\beta}_2 \bar{\rho}(z_1 - i2\omega_1, z_2 + i2\omega_2) = \bar{\chi}(z_1 - i2\omega_1, z_2). \end{aligned} \quad (22)$$

We see that the price paid is the introduction of new unknowns such as $\bar{\rho}(z_1 - i4\omega_1, z_2)$, $\bar{\rho}(z_1 - i2\omega_1, z_2 - i2\omega_2)$ and $\bar{\rho}(z_1 - i2\omega_1, z_2 + i2\omega_2)$. The last two new unknowns should be of the same order of magnitude as the old unknowns $\bar{\rho}(z_1 \pm i2\omega_1, z_2)$ and $\bar{\rho}(z_1, z_2 \pm i2\omega_2)$. However, $\bar{\rho}(z_1 - i4\omega_1, z_2)$ should be considered an order of magnitude smaller under the assumption of the function $|\bar{\rho}(z_1, z_2)|$ having a bell-like shape. Therefore, we can neglect the term containing $\bar{\rho}(z_1 - i4\omega_1, z_2)$ in eq. (22) quite in the same way as we did before for the terms containing the variables $\bar{\rho}(z_1 \pm i2\omega_1, z_2)$ and $\bar{\rho}(z_1, z_2 \pm i2\omega_2)$ in eq. (20) when compared to the variable $\bar{\rho}(z_1, z_2)$ in the RWA. Thus eq. (22) becomes

$$\begin{aligned} \bar{\alpha}_2 \bar{\rho}(z_1 - i2\omega_1, z_2 - i2\omega_2) + \bar{\pi}(z_1 - i2\omega_1, z_2) \bar{\rho}(z_1 - i2\omega_1, z_2) \\ + \bar{\beta}_1 \bar{\rho}(z_1, z_2) + \bar{\beta}_2 \bar{\rho}(z_1 - i2\omega_1, z_2 + i2\omega_2) = \bar{\chi}(z_1 - i2\omega_1, z_2). \end{aligned} \quad (23)$$

Having two equations whose unknowns have the same order of magnitude, we still require additional equations: we choose shifts of the form $z_1 \rightarrow z_1 + i2p\omega_1$, $z_2 \rightarrow z_2 + i2q\omega_2$ with $p, q = \{-1, 0, 1\}$ and repeat the process above, again neglecting $\bar{\rho}(z_1 \pm i4\omega_1, z_2)$ or $\bar{\rho}(z_1, z_2 \pm i4\omega_2)$ when compared with $\bar{\rho}(z_1 \pm i2\omega_1, z_2)$ or $\bar{\rho}(z_1, z_2 \pm i2\omega_2)$.

The general scheme for the n th-order approximation therefore is as follows. The variables to be neglected are $\bar{\rho}(z_1 \pm i2(n+1)\omega_1, z_2)$ and $\bar{\rho}(z_1, z_2 \pm i2(n+1)\omega_2)$, with the set of $(2n+1)^2$ shifts of the independent variables z_1 and z_2 necessary to yield a complete set of equations from the main eq. (20) given by $z_1 \rightarrow z_1 + i2p\omega_1, z_2 \rightarrow z_2 + i2q\omega_2$ with $p, q = \{-n, -n+1, \dots, 0, \dots, n-1, n\}$.

Since we are only interested in *average* quantities in our approximation scheme here, one can furthermore neglect all terms like

$$\frac{1}{z_1 \pm ip2\omega_1} \quad \text{or} \quad \frac{1}{z_2 \pm iq2\omega_2}, \quad (24)$$

with $p, q = \{-n, \dots, -1, 1, \dots, n\}$ and $p, q \neq 0$ in the n th-order approximation. These terms appear in the shifted functions $\bar{\chi}(z_1 + i2p\omega_1, z_2 + i2q\omega_2)$ and generate oscillating contributions around the average.

As a result, we end up with $(2n+1)^2$ matrix equations of the type of eq. (20) or eq. (23) for $(2n+1)^2$ unknowns of the form $\bar{\rho}(z_1 + i2p\omega_1, z_2 + i2q\omega_2)$. Equivalently, recalling that each of these matrix equations represents actually a set of eight elementary equations, the ultimate set of equations to be solved consists of $8(2n+1)^2$ elementary equations in the n th-order approximation. This can be written symbolically, again using matrix notation, as

$$\left[\begin{array}{c} \boxed{\text{diagonal}} \\ 8(2n+1)^2 \times 8(2n+1)^2 \end{array} \right] \left[\begin{array}{c} \boxed{\rho} \\ \text{diagonal} \end{array} \right] = \frac{1}{z_1 z_2} \left[\begin{array}{c} \boxed{\chi} \\ \text{diagonal} \end{array} \right], \quad (25)$$

where the $8(2n+1)^2 \times 8(2n+1)^2$ matrix can be thought of being made up of sub-blocks consisting of the 8×8 matrices $\bar{\alpha}_i, \bar{\beta}_j$ and $\bar{\pi}(z_1 + i2p\omega_1, z_2 + i2q\omega_2)$ with $i, j = \{1, 2\}$ and $p, q = \{-n, \dots, 0, \dots, n\}$. Thus the solution of eq. (25) is essentially the n th-order approximation. Note the similarity between eq. (25) and eq. (21) for the RWA. In Appendix A-C, we give a systematic and straight-forward procedure to form the matrices of eq. (25) from the main eq. (20).

4.3 Analytical solutions from determinants

4.3.1 RWA

The population $\tilde{\rho}_{11}(z_1, z_2)$ in the RWA can be written as

$$\tilde{\rho}_{11}(z_1, z_2) = \rho_{11}(z_1, z_2) = \frac{1}{z_1 z_2} \frac{||\bar{\pi}_1(z_1, z_2)||}{||\bar{\pi}(z_1, z_2)||}, \quad (26)$$

where $\bar{\pi}(z_1, z_2)$ is the eight-by-eight matrix of eq. (21) and $\bar{\pi}_1(z_1, z_2)$ is the matrix obtained from $\bar{\pi}(z_1, z_2)$ by replacing its first column by the matrix column on the right-hand side of eq. (21). Here, double bars indicate the determinant of the matrix. Again, in the stationary limit this simplifies to

$$\langle \rho_{11}(t \rightarrow \infty) \rangle = \mathcal{L}^{-1} \left(\frac{1}{z_1 z_2} \frac{||\bar{\pi}_1(0, 0)||}{||\bar{\pi}(0, 0)||} \right) = \frac{||\bar{\pi}_1(0, 0)||}{||\bar{\pi}(0, 0)||}, \quad (27)$$

a rigorous proof for the case of the two-level system is given in Appendix D.

Using $\rho_{00} = 1 - \rho_{11} - \rho_{22}$, Eq. (27), and the corresponding relation for the population ρ_{22} , one furthermore finds

$$\langle \rho_{00}(t \rightarrow \infty) \rangle = \frac{||\bar{\pi}(0, 0) - \bar{\pi}_1(0, 0) - \bar{\pi}_2(0, 0)||}{||\bar{\pi}(0, 0)||}. \quad (28)$$

The three terms in the numerator can be combined into a single determinant by using the property of the determinant of being a multi-linear function of its columns. Thus we have

$$\langle \rho_{00}(t \rightarrow \infty) \rangle = \frac{||\bar{\bar{\pi}}_{12}(0, 0)||}{||\bar{\bar{\pi}}(0, 0)||}, \quad (29)$$

where $\bar{\bar{\pi}}_{12}(0, 0)$ results to be the matrix obtained from $\bar{\bar{\pi}}(0, 0)$ by subtracting the matrix column on the right-hand side of eq. (21) from the first column and subtracting the first column from the second column. Note that eqs. (27) to (29) again correspond to average values of the asymptotic approximate solution. For the special case of $\Omega_1 = \Omega_2 = \Omega$, this calculation yields exactly the result already found in eq. (11).

4.3.2 n -th order truncation

For the n th-order approximation, the columns corresponding to the unknowns $\tilde{\rho}_{11}(z_1, z_2)$ and $\tilde{\rho}_{22}(z_1, z_2)$ of the $8(2n+1)^2 \times 8(2n+1)^2$ matrix of the eq. (25) are no longer, in general, respectively the first and second columns as in the RWA. As shown in Appendix A-C, after re-arranging the variables one finds

$$\langle \rho_{00}(t \rightarrow \infty) \rangle = \frac{||\overbrace{\pi \cdot \bar{\cdot} \cdot \pi}^{n+1} \pi_{16n(n+1)+1, 16n(n+1)+2}(0, 0)||}{||\underbrace{\pi \cdot \bar{\cdot} \cdot \pi}_{n+1}(0, 0)||}, \quad (30)$$

where $\pi \cdot \bar{\cdot} \cdot \pi(0, 0)$ is now the $8(2n+1)^2 \times 8(2n+1)^2$ matrix of eq. (25) with $z_1 = z_2 = 0$ and $\overbrace{\pi \cdot \bar{\cdot} \cdot \pi}^{n+1} \pi_{16n(n+1)+1, 16n(n+1)+2}(0, 0)$ results to be the matrix obtained from $\pi \cdot \bar{\cdot} \cdot \pi(0, 0)$ by subtracting the matrix column on the right-hand side of eq. (25) from its $16n(n+1)+1$ -th column and subtracting its $16n(n+1)+1$ -th column from its $16n(n+1)+2$ -th column. Here, the derivation of the n th-order approximation of eq. (30) has been based on the symbolic eq. (25). This matrix eq. (25) represents actually a set of $(2n+1)$ equations of the form of, for example, eq. (20) or eq. (23). There are many ways to arrange these shifted equations in a single matrix equation which, however, all describe the same physics within a given order of approximation.

In Appendix A-C, we present a complete derivation of eq. (25) or eq. (30) for the case of the first order approximation, $n = 1$, and a straight-forward procedure to obtain the relevant matrices from the sub-blocks consisting of the more elementary matrices $\bar{\alpha}_i, \bar{\beta}_i, \bar{\pi}(z_1 + i2p\omega_1, z_2 + i2q\omega_2), \bar{\chi}(z_1 + i2p\omega_1, z_2 + i2q\omega_2)$ and $\bar{\rho}(z_1 + i2p\omega_1, z_2 + i2q\omega_2)$ for any order of approximation. Analytical results to any given order n are then easily obtained by using standard algebraic software packages.

4.4 Physical interpretation

As indicated before, the main eq. (20) of this theory is the double Laplace transform of the equation of the density matrix operator in the *transformed picture*, where the values of the field frequencies are perceived as $2\omega_1$ and $2\omega_2$ rather than ω_1 and ω_2 , e.g. $\tilde{\rho}_{ij}(z_1, z_2) = \mathcal{L}\tilde{\rho}_{ij}(t_1, t_2)$. We can therefore call this a mathematical description of a new three-level system, the *dressed system*, interacting with two (fictitious) photonic fields of frequency $2\omega_1$ and $2\omega_2$.

On the other hand, we have seen that the exact solution of (20) corresponding to the state of the system in steady-state situation as $t \rightarrow \infty$, requires the knowledge of all the density matrix elements $\tilde{\rho}_{ij}(z_1 + ip\omega_1, z_2 + iq\omega_2)$ with $p, q = \{-\infty, \dots, +\infty\}$ for $z_1 \rightarrow 0$ and $z_2 \rightarrow 0$. Therefore, before giving any interpretation to this variables, let us write them again using a different notation as $\hat{\rho}_{ij}(p, q)$, where the integers $p = 0, \pm 1, \dots$ and $q = 0, \pm 1, \dots$ are now an abbreviated way to indicate actually the values $ip2\omega_1$ and $iq2\omega_2$ respectively. Then this extended density matrix operator $\tilde{\rho}_{ij}(n, m)$ can be interpreted as the density matrix operator describing the state of the system plus the state of the two photonic fields. In this interpretation, the indices i, j are the quantum numbers of the different states of the system whereas the indices p, q become the quantum numbers corresponding to the different states of the two photonic fields.

Then, for example, $\tilde{\rho}_{ij}(-1, 2)$ indicates, regarding only to the state of the fields, the probability that the field 1 has one photon of frequency $2\omega_1$ less and the field 2 has two photons of frequency $2\omega_2$ more than the photon averages of the respective field.

In this interpretation, the RWA describes processes without exchange of photons with the *dressed system*. The $n = 1$ truncation then describes processes involving the exchange of one photon of frequency $2\omega_1$ and one of frequency $2\omega_2$ with the dressed system. In general, the n -th truncation describes processes involving the exchange of n photons from each field with the dressed system.

Now we have to consider that the dressed system is actually the bare system exchanging one real photon of frequency ω_1 or ω_2 from each real field, and that the ‘fictitious’ photons of frequencies $2\omega_1$ and $2\omega_2$ used in the discussion above manifest in reality as two real photons of frequency ω_1 and ω_2 , respectively. Then, the physical interpretation in the Laboratory frame is as follows: the solution of order zero (RWA) describes processes involving the exchange of one real photon of frequencies ω_1 or ω_2 from each field with the bare system. In general, the solution of order n describes processes involving the exchange of $2n + 1$ real photons from each real field with the real system.

5 Results

Before we apply and evaluate our method for three-level systems, we present results for two-level systems [14] both as a consistency check and as a test of the method itself. In this case, the physical interpretation remains essentially the same. The only difference is that now there is only one photonic field of frequency ω and one transition frequency ω_0 .

5.1 Check: results for two-level systems

Here, the standard quantity plotted is the mean value of the inversion, $\langle w(t) \rangle = \langle \rho_{22}(t) - \rho_{11}(t) \rangle$, when the system relaxes ideally at $t \rightarrow \infty$, as a function of the field frequency ω . The coupling of the field is given as usual by the only Rabi frequency Ω , and the relaxation coefficients are the population decay, Γ_0 , from the upper-level and the dephasing rate of the two levels, $\Gamma_{12} = \Gamma_0/2$.

In Fig. 2, we compare the numerical solution with the RWA and the first-order approximation. The first-order approximation and the numerical solution practically coincide, and their values start to differ from those predicted by the RWA. Also one recognises that the maximum in the inversion, is achieved not exactly at $\omega = \omega_0 = 1$, as the RWA predicts, indicating a shift in the true resonance frequency. This is the famous Bloch-Siegert shift [15, 16]. Overall, the physics here is dominated by processes involving the exchange of one photon of frequency $\omega = \omega_0 = 1$ (peak at $\omega = 1$).

Furthermore, there is a new, although less pronounced, peak at field frequency $\omega = \omega_0/3 = 1/3$, which indicates processes involving the exchange of three photons of frequency $\omega = 1/3$. A fact that passes totally unnoticed in the RWA but not in the first-order approximation where a peak also appear there. Again, this shows the correctness of the physical interpretation of the perturbative method.

5.1.1 Strong field limit

In Fig. 3, we compare the numerical solution with the first, second, third and fourth order of approximation for an intensity of the field equal to $\Omega = 0.99$. Apart from the main resonance at $\omega = \omega_0 = 1$, the numerical solution shows four extra peaks. On the other hand, the first-order approximation has only one extra peak, the second-order approximation has two extra peaks, the third-order approximation has three extra peaks and the fourth-order approximation has fourth extra peaks. Under our previous physical interpretation of each order of approximation, this means that these four extra peaks of the numerical solution are simply three-photon, five-photon, seven-photon and nine-photon resonances. The fact that they are not centred exactly at $\omega = 1/3$, $\omega = 1/5$, $\omega = 1/7$ and $\omega = 1/9$ is due to the anti-resonant terms that have a large effect at this high intensity of the field. This selection rule of odd number of photons for the multi-photon

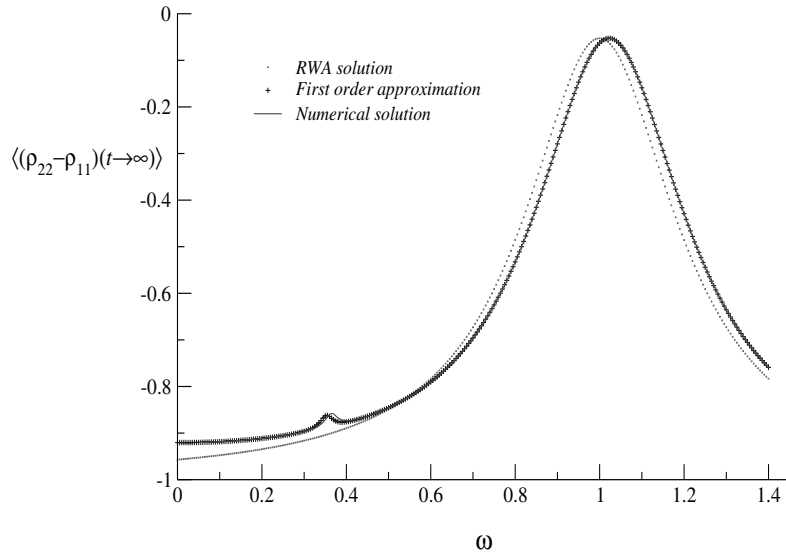


Fig. 2 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, RWA indicated by the dotted line and first order approximation indicated by the plus signs for the steady-state inversion $\langle\rho_{22}(t \rightarrow \infty) - \rho_{11}(t \rightarrow \infty)\rangle$ for the two-level system as a function of the field frequency ω . The intensity of the field is so strong that there also appears a second peak in the numerical solution near $\omega = 1/3$ indicating a three-photon resonance as well. Parameters: $\Omega = 0.3$, $\Gamma_0 = 0.1$, $\Gamma_{12} = \Gamma_0/2$.

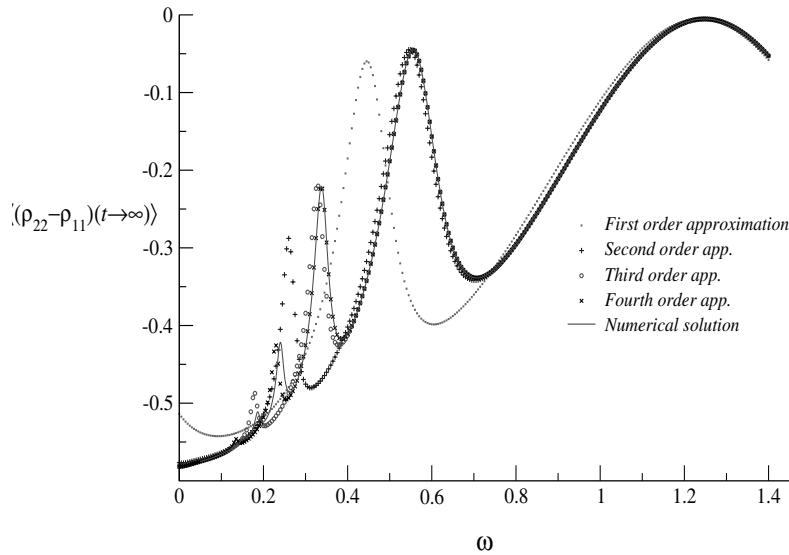


Fig. 3 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, first order approximation indicated by the dotted line, second order approximation indicated by the plus signs, third order approximation indicated by the circles and fourth order approximation indicated by the crosses for the steady-state inversion $\langle\rho_{22}(t \rightarrow \infty) - \rho_{11}(t \rightarrow \infty)\rangle$ for the two-level system as a function of the field frequency ω . The intensity of the field has been increased even more so that there now appear four extra peaks in the numerical solution indicating a three-photon, five-photon, seven-photon and nine-photon resonances as well. Parameters: $\Omega = 0.99$, $\Gamma_0 = 0.1$, $\Gamma_{12} = \Gamma_0/2$.

resonances of the two-level system has already been deduced by Shirley [9] in his seminal paper on Floquet theory in an undamped system, which is reassuring again since it agrees perfectly well with the interpretation of the theory here.

5.1.2 Bloch-Siegert shift

Another check of the validity of the perturbative method is to re-derive the Bloch-Siegert shift: The corresponding maximum of the inversion is not centred exactly at $\omega = \omega_0 = 1$. The Bloch-Siegert shift for this frequency, $\delta_{B-S} = \omega_{B-S} - \omega_0$, can be estimated by calculating the position of this maximum in an analytical solution. Here, the RWA (which predicts a maximum at $\omega_{max} = \omega_0$) predicts $\delta_{B-S} = 0$. Setting the relaxation parameter $\Gamma_o \rightarrow 0$ and solving a simple extreme value problem for the analytical solution of order four, we find

$$\omega_{B-S} = \omega_0 + \delta_{B-S} = \omega_0 + \frac{\Omega^2}{4\omega_0} + \frac{\Omega^4}{64\omega_0^3} - \frac{53}{2048} \frac{\Omega^6}{\omega_0^5} + \frac{103}{32768} \frac{\Omega^8}{\omega_0^7} + \dots, \quad (31)$$

which agrees perfectly with the expression obtained by Shirley [9] if we take into account that the definition of his Rabi frequency differs from ours just in a factor of 2, e.g. $b = \Omega/2$. He obtained this expansion by using the Floquet theory of an isolated system.

5.2 Results: three-level system

In our numerical results, we have chosen $\omega_{21} = 176 \times 2^{-12} \omega_{01}$ for numerical convenience (see appendix E).

Fig. 4 shows a comparison between the RWA, the $n = 1$, and the exact solution for the occupation of the upper level. All the parameters are measured in units of the transition frequency ω_{01} , which has here been chosen to be equal to $\omega_{01} = 1$. Therefore, the condition $\Omega_1, \Omega_2 \ll \omega_{01} \sim \omega_{02}$ for the RWA to be a good approximation does not apply now, for these large values of the Rabi frequencies, $\Omega_1 = \Omega_2 = \omega_{01}/4$. This is effectively corroborated in Fig. 4. The dotted curve, representing the values assumed by the RWA, passes approximately 10% above the continuous curve (the numerical result), on the left of $\delta_1 = 0$ around $\delta_1 = -2\Gamma_0$ and 10% below on the right around $\delta_1 = 2\Gamma_0$, showing a considerable discrepancy.

The other important aspect is the fact that coherent population trapping is distorted for these large values of the Rabi frequencies. The usually very narrow dip in the occupation of the $|0\rangle$ upper level around the two-photon Raman resonance condition now broadens drastically. Furthermore, the splitting of the two maxima away from each other is also accompanied by a gradual loss of definition of the Raman resonance trapped state, which corresponds to the rise of the minimum as the intensities of the fields are increased. Another feature is the loss of symmetry around $\delta_R = 0$, as compared with the RWA solution.

5.2.1 Beyond the RWA

Fig. 4 also demonstrates that the $n = 1$ st order approximation of our scheme works already nearly perfect at $\Omega_1 = \Omega_2 = 0.25$.

In Figs. 5 to 9, we compare the different truncation levels of our scheme for Rabi frequencies as increased from $\Omega = \Omega_1 = \Omega_2 = 0.3$ to $\Omega = \Omega_1 = \Omega_2 = 0.8$. In Fig. 5, one recognises that at $\Omega = 0.3$, the first-order approximation starts to fail slightly. The second-order approximation is very good upto Rabi frequency $\Omega = 0.4$, and only at $\Omega = 0.5$ there is a very slight difference between this curve and the numerical solution, as Fig. 6 and 7 corroborate. Fig. 8 shows that, if the Rabi frequency is increased even more upto $\Omega = 0.6$, the third-order approximation performs well. The threshold beyond which this approximation start to fail is around $\Omega = 0.8$, cf. Fig. 9.

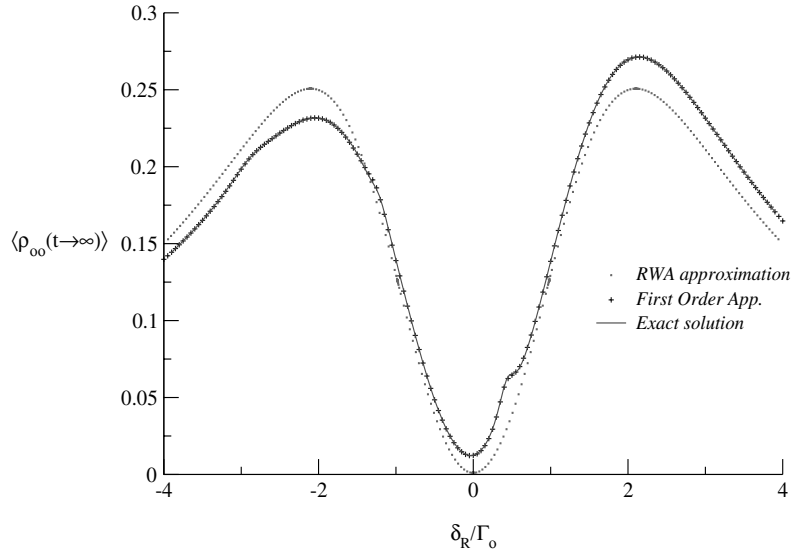


Fig. 4 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, RWA indicated by the dotted line and first order approximation indicated by the blue plus signs for the steady-state excited-state population $\langle \rho_{00}(t \rightarrow \infty) \rangle$ for Λ system as a function of the Raman detuning $\delta_R = \delta_1$. Parameters: $\delta_2 = 0$, $\Omega_1 = \Omega_2 = 0.25$, $\Gamma_0 = 0.1$, $\Gamma_{12} = 0.0001$.

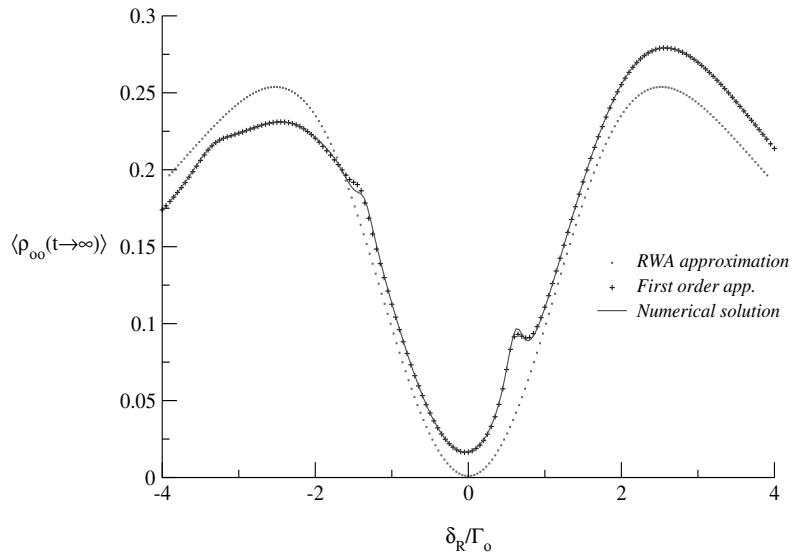


Fig. 5 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, RWA indicated by the dotted line and first order approximation indicated by the plus signs for the steady-state excited-state population $\langle \rho_{00}(t \rightarrow \infty) \rangle$ for Λ system as a function of the Raman detuning $\delta_R = \delta_1$. Parameters: $\delta_2 = 0$, $\Omega_1 = \Omega_2 = 0.3$, $\Gamma_0 = 0.1$, $\Gamma_{12} = 0.0001$.

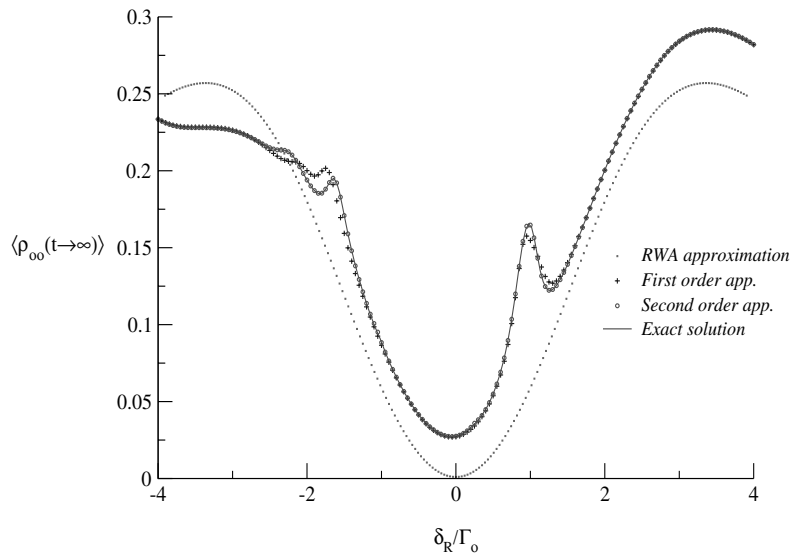


Fig. 6 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, RWA indicated by the dotted line, first order approximation indicated by the plus signs and second order approximation indicated by the circles for the steady-state excited-state population $\langle \rho_{00}(t \rightarrow \infty) \rangle$ for Λ system as a function of the Raman detuning $\delta_R = \delta_1$. Parameters: $\delta_2 = 0$, $\Omega_1 = \Omega_2 = 0.4$, $\Gamma_0 = 0.1$, $\Gamma_{12} = 0.0001$.

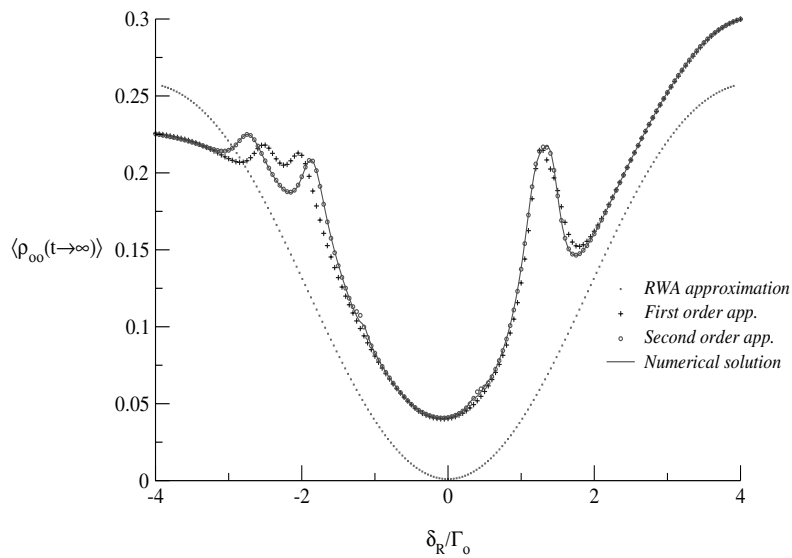


Fig. 7 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, RWA indicated by the dotted line, first order approximation indicated by the plus signs and second order approximation indicated by the circles for the steady-state excited-state population $\langle \rho_{00}(t \rightarrow \infty) \rangle$ for Λ system as a function of the Raman detuning $\delta_R = \delta_1$. Parameters: $\delta_2 = 0$, $\Omega_1 = \Omega_2 = 0.5$, $\Gamma_0 = 0.1$, $\Gamma_{12} = 0.0001$.

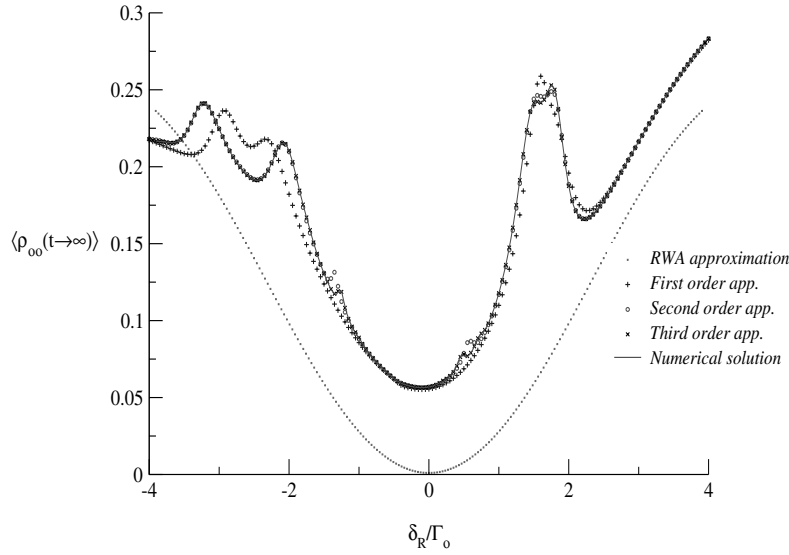


Fig. 8 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, RWA indicated by the dotted line, first order approximation indicated by the plus signs, second order approximation indicated by the circles and third order approximation indicated by the crosses for the steady-state excited-state population $\langle \rho_{00}(t \rightarrow \infty) \rangle$ for Λ system as a function of the Raman detuning $\delta_R = \delta_1$. Parameters: $\delta_2 = 0$, $\Omega_1 = \Omega_2 = 0.6$, $\Gamma_0 = 0.1$, $\Gamma_{12} = 0.0001$.

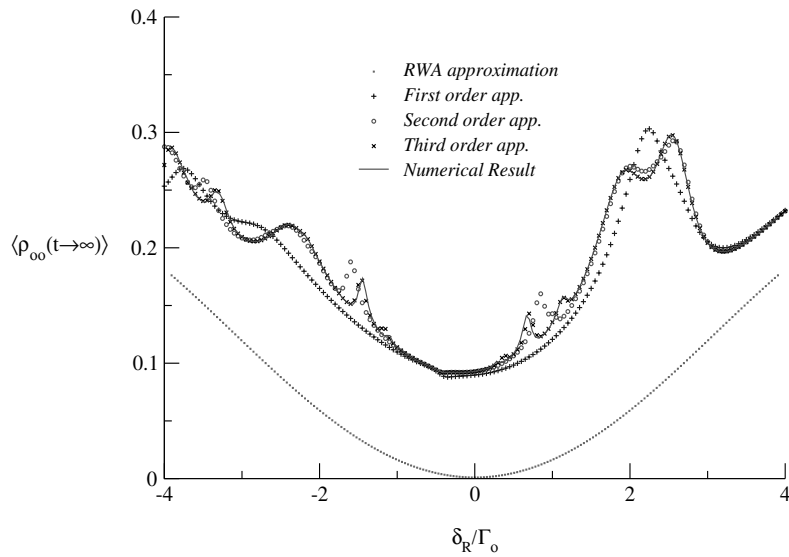


Fig. 9 (online colour at: www.ann-phys.org) Numerical solution indicated by the continuous line, RWA indicated by the dotted line, first order approximation indicated by the plus signs, second order approximation indicated by the circles and third order approximation indicated by the crosses for the steady-state excited-state population $\langle \rho_{00}(t \rightarrow \infty) \rangle$ for Λ system as a function of the Raman detuning $\delta_R = \delta_1$. Parameters: $\delta_2 = 0$, $\Omega_1 = \Omega_2 = 0.8$, $\Gamma_0 = 0.1$, $\Gamma_{12} = 0.0001$.

5.2.2 Efficiency of the method

Figs. 4 to 9 also reveal the rapid convergence of the perturbative method. When the dimensionless coupling constant $g = \Omega/\omega_{01} = \Omega$, is as large as $\Omega = 0.8$, the analytical solution of third order is already a good approximation. This, combined with the speed of the actual calculation, makes this theory an efficient method to estimate physical quantities, in this case the occupation of the excited-level.

The first-order approximation, which is essentially the solution of the matrix eq. (38), is much faster to calculate than the numerical solution in normal practical situations. Indeed, the mathematical operation to perform is the ratio of two 72×72 determinants for this case. We calculated this determinant quotient 160 times (once for each value of the Raman detuning ($\delta_R = \delta_1$) plotted in any of Figs. 4 to 9), using *Mathematica*, which yields the whole curve for this order of approximation in 8 seconds. For the curves corresponding to the second and third order of approximation, these times increase to 1:55 minutes and 13:10 minutes, respectively while the sizes of the determinants involved in the operation become 200×200 for one case and 392×392 for the other. The ratios of these times of calculation of one order to the previous one ($1:55/0:08 = 14.375$, $13:10/1:55 = 6.87$) show that this length of time increases but with smaller proportion as superior order are considered. A tendency that is also followed in the size of the determinants involved in each order of calculation.

The essential point here is that the calculation times in each order of approximation are fixed and do not depend on the actual values of the parameters introduced in Fig. 1. By contrast, the numerical calculation of the solution in Figs. 4 to 9 are proportional to the relaxation time of the system $1/\Gamma_0$. This is because each point of these curves are obtained essentially by giving an arbitrary initial condition to the evolution eqs. (3) at $t = 0$ and propagating the solution far beyond the relaxation time $1/\Gamma_0$. The actual specific desired value in each propagation is the mean value of the excited-level occupation when the system relaxes, $\langle \rho_{00}(t \rightarrow \infty) \rangle$ ideally at $t \rightarrow \infty$. We performed this propagation by using a Runge-Kutta method with adaptive step-size control. The actual routine used yields the whole numerical curves in Figs. 4 to 9 in only 14:17 minutes because the relaxation coefficient Γ_0 has been chosen here unrealistically large, e.g. $\Gamma_0 = 0.1$, for convenience.

6 Discussion

In real experimental situations, the value of the excited state lifetime ($1/\Gamma_0$) is much longer than we assumed in our model calculations here. This makes the duration of standard numerical calculations increase proportional to $1/\Gamma_0$. As an example in optical resonance, sodium vapour possesses the well-known D lines at $\omega_{01} = 3198770$ GHz (5890 Å) and $\omega_{02} = 3195520$ GHz (5896 Å) with an absorption line-width of $\delta\omega_I \sim 10$ GHz at ordinary temperature and pressure [17, 18]. In this case, the relaxation parameter Γ_0 measured in units of the transition frequency ω_{01} or ω_{02} can be estimated to be $\Gamma_0^{op} \sim \delta\omega_I/\omega_{01} \sim \delta\omega_I/\omega_{02} \sim 0.000003$. This makes the numerical calculation based on the Runge-Kutta method very inconvenient.

In the case of the photon-assisted-tunnelling problem mentioned before, this superiority in calculation speed of the perturbative method is crucial because of the need to consider analytical solutions of order superior to the RWA. In this practical application the inelastic rate Γ_0 at which the quantum dots in GaAs emit phonons was estimated to be between $\Gamma_0 = 7.8 \times 10^{-4}$ to $\Gamma_0 = 1.3 \times 10^{-3}$ times the characteristic transition frequencies $\omega_0 = 7.6 \times 10^2$ GHz and $\omega_0 = 1.52 \times 10^3$ GHz respectively [6]. These longer values of the excited-state lifetime $1/\Gamma_0$ makes the numerical calculation, which before lasted 14:17 minutes for $\Gamma_0 = 0.1$, now last 18:19:49 hours for the most favourable case of $\Gamma_0 = 1.3 \times 10^{-3}$.

In conclusion, the perturbative method presented in this paper is an elegant theory, easy to use and constitutes a powerful tool to estimate physical quantities where the RWA is a poor approximation, e.g. $\Omega_1 \leq \omega_{01}$ and $\Omega_2 \leq \omega_{02}$. Although in this paper, we have only discussed the 'isolated' three-level system physics, in principle the method is applicable to more general situations (collisions, other decay channels) and where the optical system is combined with, e.g., transport of electrons in semiconductor structures [6, 7].

A The expression for the first order approximation

In this Appendix, we review the procedure outlined around the explanation of eqs. (22) to (25) leading to the expression of the n th-order approximation of eq. (30). Let us re-write (20) as

$$\begin{aligned} & \bar{\alpha}_1 \bar{\rho}(z_1 - i2\omega_1, z_2) + \bar{\alpha}_2 \bar{\rho}(z_1, z_2 - i2\omega_2) \\ & + \bar{\pi}(z_1, z_2) \bar{\rho}(z_1, z_2) + \bar{\beta}_1 \bar{\rho}(z_1 + i2\omega_1, z_2) + \bar{\beta}_2 \bar{\rho}(z_1, z_2 + i2\omega_2) = \bar{\chi}(z_1, z_2), \end{aligned} \quad (32)$$

where

$$\begin{aligned} \bar{\alpha}_1 &= \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ i\Omega_1 & \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{i\Omega_1}{2} \\ 0 & 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}, & \bar{\alpha}_2 &= \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_2}{2} & 0 \\ 0 & 0 & 0 & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 \\ \frac{i\Omega_2}{2} & i\Omega_2 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_2}{2} & 0 & 0 \end{bmatrix}, \\ \bar{\beta}_1 &= \begin{bmatrix} 0 & 0 & \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ -i\Omega_1 - \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 \end{bmatrix}, & \bar{\beta}_2 &= \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_2}{2} \\ -\frac{i\Omega_2}{2} & -i\Omega_2 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}, \quad (33) \\ \bar{\rho}(z_1, z_2) &= \begin{bmatrix} \tilde{\rho}_{11}(z_1, z_2) \\ \tilde{\rho}_{22}(z_1, z_2) \\ \tilde{\rho}_{01}(z_1, z_2) \\ \tilde{\rho}_{02}(z_1, z_2) \\ \tilde{\rho}_{21}(z_1, z_2) \\ \tilde{\rho}_{10}(z_1, z_2) \\ \tilde{\rho}_{20}(z_1, z_2) \\ \tilde{\rho}_{12}(z_1, z_2) \end{bmatrix}, & \bar{\chi}(z_1, z_2) &= \begin{bmatrix} \frac{\Gamma_o}{2z_1 z_2} \\ \frac{\Gamma_o}{2z_1 z_2} \\ \frac{i\Omega_1}{2z_2} \frac{1}{z_1 - i2\omega_1} + \frac{i\Omega_1}{2z_1 z_2} \\ \frac{i\Omega_2}{2z_1} \frac{1}{z_2 - i2\omega_2} + \frac{i\Omega_2}{2z_1 z_2} \\ 0 \\ -\frac{i\Omega_1}{2z_2} \frac{1}{z_1 + i2\omega_1} - \frac{i\Omega_1}{2z_1 z_2} \\ -\frac{i\Omega_2}{2z_1} \frac{1}{z_2 + i2\omega_2} - \frac{i\Omega_2}{2z_1 z_2} \\ 0 \end{bmatrix} \end{aligned}$$

and $\bar{\pi}(z_1, z_2)$ being the eight-by-eight matrix in eq. (21).

In the RWA, we neglect $\bar{\rho}(z_1 \pm i2\omega_1, z_2)$ and $\bar{\rho}(z_1, z_2 \pm i2\omega_2)$ when compared to $\bar{\rho}(z_1, z_2)$ so that eq. (32) becomes immediately solvable. This neglect is based on the assumption that $|\bar{\rho}(z_1, z_2)|$ has a bell-like shape centred around the point $(z_1 = 0, z_2 = 0)$ as explained before. To obtain better approximations, we need more equation, for example in the first order approximation, for the variables $\bar{\rho}(z_1 + i2p\omega_1, z_2 + i2q\omega_2)$ with $p, q = \{-1, 0, 1\}$. These equations are obtained from eq. (32) by just performing the shifts $z_1 \rightarrow z_1 + i2p\omega_1$, $z_2 \rightarrow z_2 + i2q\omega_2$ with $p, q = \{-1, 0, 1\}$ in the independent variables z_1 and z_2 , and neglecting the generated terms containing the variables $\bar{\rho}(z_1 \pm i4\omega_1, z_2)$ and $\bar{\rho}(z_1, z_2 \pm i4\omega_2)$, which appear as by-products of this operation. For example, eq. (23) is obtained by replacing $z_1 \rightarrow z_1 - i2\omega_1$ and $z_2 \rightarrow z_2$ in eq. (32), and neglecting the term containing the variable $\bar{\rho}(z_1 - i4\omega_1, z_2)$. In synthesis, arranging together all the shifted equations, we end up with the following complete set of nine equations in this order of approximation

$$\begin{bmatrix}
\bar{\pi}(z_1^{-1}, z_2^{-1}) & \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\
\bar{\alpha}_2 & \bar{\pi}(z_1^{-1}, z_2^0) & \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\
\bar{0} & \bar{\alpha}_2 & \bar{\pi}(z_1^{-1}, z_2^{+1}) & \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} \\
\bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 & \bar{\pi}(z_1^0, z_2^{-1}) & \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} \\
\bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 & \bar{\pi}(z_1^0, z_2^0) & \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} \\
\bar{0} & \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 & \bar{\pi}(z_1^0, z_2^{+1}) & \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 \\
\bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 & \bar{\pi}(z_1^{+1}, z_2^{-1}) & \bar{\beta}_2 & \bar{0} \\
\bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 & \bar{\pi}(z_1^{+1}, z_2^0) & \bar{\beta}_2 \\
\bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 & \bar{\pi}(z_1^{+1}, z_2^{+1})
\end{bmatrix}
\begin{bmatrix}
\bar{\rho}(z_1^{-1}, z_2^{-1}) \\
\bar{\rho}(z_1^{-1}, z_2^0) \\
\bar{\rho}(z_1^{-1}, z_2^{+1}) \\
\bar{\rho}(z_1^0, z_2^{-1}) \\
\bar{\rho}(z_1^0, z_2^0) \\
\bar{\rho}(z_1^0, z_2^{+1}) \\
\bar{\rho}(z_1^{+1}, z_2^{-1}) \\
\bar{\rho}(z_1^{+1}, z_2^0) \\
\bar{\rho}(z_1^{+1}, z_2^{+1})
\end{bmatrix}
=
\begin{bmatrix}
\bar{\chi}(z_1^{-1}, z_2^{-1}) \\
\bar{\chi}(z_1^{-1}, z_2^0) \\
\bar{\chi}(z_1^{-1}, z_2^{+1}) \\
\bar{\chi}(z_1^0, z_2^{-1}) \\
\bar{\chi}(z_1^0, z_2^0) \\
\bar{\chi}(z_1^0, z_2^{+1}) \\
\bar{\chi}(z_1^{+1}, z_2^{-1}) \\
\bar{\chi}(z_1^{+1}, z_2^0) \\
\bar{\chi}(z_1^{+1}, z_2^{+1})
\end{bmatrix},
\tag{34}$$

where z_1^p and z_2^q with $p, q = \{-1, 0, 1\}$ are an abbreviated way to express actually $z_1 + i2p\omega_1$ and $z_2 + i2q\omega_2$ and $\bar{0}$ is the eight-by-eight matrix zero. Note that we have again used matrix notation to express this new set of equations. Let us label each row or column of the square matrix in eq. (34) with the number $3p + q$ where p and q are the superscripts of the arguments of the function $\bar{\pi}(z_1^p, z_2^q)$ in the row or column considered. Then we see that the main eq. (32) is represented by the row number zero, the central row, of this square matrix. The other rows represent the other shifted equations. For example, the row number $-3 = 3(-1) + 0$, the row located three rows above the central row, corresponds to the shift $z_1 \rightarrow z_1 - i2\omega_1, z_2 \rightarrow z_2$. This set of nine equations for the nine vector unknowns $\bar{\rho}(z_1 + i2p\omega_1, z_2 + i2q\omega_2)$ with $p, q = \{-1, 0, 1\}$ is actually a set of 72 equations for the 72 elementary unknowns since each element of the nine-by-nine matrix in eq. (34) represents, in turn, an eight-by-eight sub-matrix.

Note also that the set of equations represented by the single matrix eq. (34) is arranged so that one row of the square matrix there is just the previous row shifted one matrix element. This happens only when the ordering of the unknown quantities in the column matrix is the one chosen in eq. (34). In general, the arrangement of the shifted equations for the n th-order approximation that yields the same simplification is such that the unknown quantities $\bar{\rho}(z_1^p, z_2^q)$ with $z_1^p = z_1 + i2p\omega_1$ and $z_2^q = z_2 + i2q\omega_2$ have the same ordering as the integer $o = (2n + 1)p + q$ with the integers $p, q = \{-n, -n + 1, \dots, n - 1, n\}$. For example, in the case of the second order of approximation where $n = 2$, the minimum value of this ordering variable is $o = -12$ corresponding to the values $p = -2$ and $q = -2$, the next value is $o = -11$ corresponding to the values $p = -2$ and $q = -1$ and so on. This means, for example, that, for this order of approximation, $\bar{\rho}(z_1^{-2}, z_2^{-2})$ is the first unknown in the unknown column, $\bar{\rho}(z_1^{-2}, z_2^{-1})$, the second and so on. Thus with this arrangement of the shifted equations, the only row that we have to remember is the one corresponding to the main eq. (32), which, for example, in the first order of approximation looks like this

$$\bar{0}; \bar{\alpha}_1; \bar{0}; \bar{\alpha}_2; \bar{\pi}(z_1, z_2); \bar{\beta}_2; \bar{0}; \bar{\beta}_1; \bar{0}.
\tag{35}$$

Then the main matrix is obtained by shifting this row to the left and to the right and discarding the matrix elements that fall outside the square frame as appears, for example, in eq. (34) for the first order of approximation.

Finally it is worth mentioning that this straight-forward way to obtain the square matrix in eq. (34), or in general in eq. (30), from the main eq. (32), facilitates enormously the task to enter this matrix in, for example, the commercial software *Mathematica* when trying to calculate its determinant. This is because the problem has now been reduced essentially to enter only one row. Furthermore the blocks represented by the sub-matrices $\bar{\alpha}_1, \bar{\alpha}_2, \bar{\beta}_1, \bar{\beta}_2$ and $\bar{\pi}(z_1, z_2)$ can be entered only once in *Mathematica*, and then treated as single objects there when forming the square matrix.

B Analytical solution

We can neglect the terms containing the factors

$$\frac{1}{z_1 \pm ip2\omega_1} \quad \text{or} \quad \frac{1}{z_2 \pm iq2\omega_2}, \quad (36)$$

with $p, q = \{-1, 1\}$, in the matrix column on the right-hand side of eq. (34). This terms generate oscillatory contributions around the average. Then, for example, the vector $\bar{\chi}(z_1, z_2)$ becomes

$$\bar{\chi}(z_1, z_2) \rightarrow \frac{1}{z_1 z_2} \begin{bmatrix} \frac{\Gamma_o}{2} \\ \frac{\Gamma_o}{2} \\ i\frac{\Omega_1}{2} \\ i\frac{\Omega_2}{2} \\ 0 \\ -i\frac{\Omega_1}{2} \\ -i\frac{\Omega_2}{2} \\ 0 \\ 0 \end{bmatrix}. \quad (37)$$

If we perform the same simplification to the other elements of the matrix column on the right-hand side of eq. (34), these set of 72 equations for 72 unknowns becomes

$$\begin{bmatrix} \bar{\pi}(z_1^{-1}, z_2^{-1}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\ \bar{\alpha}_2 \bar{\pi}(z_1^{-1}, z_2^0) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\ \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1^{-1}, z_2^{+1}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\ \bar{\alpha}_1 \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1^0, z_2^{-1}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\ \bar{0} & \bar{\alpha}_1 \bar{0} & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1^0, z_2^0) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} \\ \bar{0} & \bar{0} & \bar{\alpha}_1 \bar{0} & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1^0, z_2^{+1}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} \\ \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 \bar{0} & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1^{+1}, z_2^{-1}) \bar{\beta}_2 & \bar{0} & \bar{0} & \bar{0} \\ \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 \bar{0} & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1^{+1}, z_2^0) \bar{\beta}_2 & \bar{0} & \bar{0} \\ \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 \bar{0} & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1^{+1}, z_2^{+1}) \bar{\beta}_2 & \bar{0} \end{bmatrix} \begin{bmatrix} \bar{\rho}(z_1^{-1}, z_2^{-1}) \\ \bar{\rho}(z_1^{-1}, z_2^0) \\ \bar{\rho}(z_1^{-1}, z_2^{+1}) \\ \bar{\rho}(z_1^0, z_2^{-1}) \\ \bar{\rho}(z_1^0, z_2^0) \\ \bar{\rho}(z_1^0, z_2^{+1}) \\ \bar{\rho}(z_1^{+1}, z_2^{-1}) \\ \bar{\rho}(z_1^{+1}, z_2^0) \\ \bar{\rho}(z_1^{+1}, z_2^{+1}) \end{bmatrix} = \frac{1}{z_1 z_2} \begin{bmatrix} \bar{\chi}_1 \\ \bar{\chi}_2 \\ \bar{\chi}_3 \\ \bar{\chi}_4 \\ \bar{\chi}_5 \\ \bar{\chi}_6 \\ \bar{\chi}_7 \\ \bar{\chi}_8 \\ \bar{\chi}_9 \end{bmatrix}, \quad (38)$$

where the constant vectors $\bar{\chi}_1, \bar{\chi}_2, \dots, \bar{\chi}_9$, on the right-hand side, are

$$\bar{\chi}_1 = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \bar{\chi}_2 = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ -\frac{i\Omega_1}{2} \\ 0 \\ 0 \end{bmatrix}, \quad \bar{\chi}_3 = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \bar{\chi}_4 = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ -\frac{i\Omega_2}{2} \\ 0 \end{bmatrix}, \quad \bar{\chi}_5 = \begin{bmatrix} \frac{\Gamma_o}{2} \\ \frac{\Gamma_o}{2} \\ i\frac{\Omega_1}{2} \\ i\frac{\Omega_2}{2} \\ 0 \\ -i\frac{\Omega_1}{2} \\ -i\frac{\Omega_2}{2} \\ 0 \\ 0 \end{bmatrix},$$

$$\bar{\chi}_6 = \begin{bmatrix} 0 \\ 0 \\ 0 \\ \frac{i\Omega_2}{2} \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \bar{\chi}_7 = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \bar{\chi}_8 = \begin{bmatrix} 0 \\ 0 \\ \frac{i\Omega_1}{2} \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \bar{\chi}_9 = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}. \quad (39)$$

Note the similarity between eq. (38) and eq. (21). The major difference is that we now have to resolve a set of 72 equations rather than the set of eight equations of the RWA. Another difference is that now the column related to the unknowns $\tilde{\rho}_{11}(z_1, z_2)$ and $\tilde{\rho}_{22}(z_1, z_2)$ are column 33 and column 34 respectively. Therefore, for example, the value of the population $\tilde{\rho}_{11}(z_1, z_2)$ can be written, in this order of approximation, as

$$\tilde{\rho}_{11}(z_1, z_2) = \frac{1}{z_1 z_2} \frac{\|\bar{\pi}\bar{\pi}_{33}(z_1, z_2)\|}{\|\bar{\pi}\bar{\pi}(z_1, z_2)\|}, \quad (40)$$

where $\bar{\pi}\bar{\pi}(z_1, z_2)$ is now the 72×72 matrix of eq. (38) and $\bar{\pi}\bar{\pi}_{33}(z_1, z_2)$ is the matrix obtained from $\bar{\pi}\bar{\pi}(z_1, z_2)$ by replacing its 33th column by the column matrix on the right-hand side of eq. (38). The double bars indicate, as usual, the determinant of the matrix standing between them.

To obtain the inverse-Laplace transform of eq. (40) when $z_1 \rightarrow 0, z_2 \rightarrow 0$, we proceed in the same way as we did before from eq. (26). Thus we find

$$\langle \rho_{11}(t \rightarrow \infty) \rangle = \frac{\|\bar{\pi}\bar{\pi}_{33}(0, 0)\|}{\|\bar{\pi}\bar{\pi}(0, 0)\|}, \quad (41)$$

where now this inverse-Laplace transform gives directly the mean value of the asymptotic solution for the population $\rho_{11}(t \rightarrow \infty)$. Remember that we have neglected the oscillatory contribution indicated in eq. (36).

To find the mean asymptotic value of the population of the upper level ρ_{00} , we essentially repeat the same reasoning yielding eq. (29) from eq. (27). Thus we find a similar formula

$$\langle \rho_{00}(t \rightarrow \infty) \rangle = \frac{\|\bar{\pi}\bar{\pi}_{33-34}(0, 0)\|}{\|\bar{\pi}\bar{\pi}(0, 0)\|}, \quad (42)$$

where $\bar{\pi}\bar{\pi}_{33-34}(0, 0)$ results to be the matrix obtained from $\bar{\pi}\bar{\pi}(0, 0)$ by subtracting the matrix column on the right-hand side of eq. (38) from its 33th column and subtracting its 33th column from its 34th column.

The generalisation of this procedure to obtain better and better approximations is straight-forward. For example, for the n th-order approximation, we end up resolving the set of $8(2n+1)^2$ equations for $8(2n+1)^2$ unknowns already introduced in eq. (25). This set of equations are obtained from eq. (32) by performing the $(2n+1)^2$ possible pairs of combinations of the following shifts of the independent variables $z_1 \rightarrow z_1 + i2p\omega_1, z_2 \rightarrow z_2 + i2q\omega_2$ with $p, q = \{-n, -n+1, \dots, n-1, n\}$ and neglecting the terms of these equations that contain the unknowns $\bar{\rho}(z_1 \pm i(n+1)2\omega_1, z_2)$ or $\bar{\rho}(z_1, z_2 \pm i(n+1)2\omega_2)$ as indicated before. And the expression for the the mean value of the asymptotic solution of the population of the upper level ρ_{00} for this order of approximation has already been given in eq. (30).

C Final simplification

Finally it is important to mention that the procedure described above for obtaining the matrix

$\overbrace{\pi \dots \pi}^{n+1} \pi_{16n(n+1)+1, 16n(n+1)+2}(0, 0)$ in the numerator in eq. (30) yields systematically a matrix that has

exactly the same structure as the one in the denominator, i.e., $\underbrace{\bar{\pi} \dots \bar{\pi}}_{n+1}(0, 0)$, irrespective of the order of approximation considered. For example, for the analytical solution of order one in eq. (42), the matrix in the numerator is equal to

$$\bar{\pi} \bar{\pi}_{33-34}(0, 0) = \begin{bmatrix} \bar{\pi}(z_{1-}, z_{2-}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\ \bar{\alpha}_2 \bar{\pi}(z_{1-}, z_2) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1^* & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\ \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_{1-}, z_{2+}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} & \bar{0} \\ \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1, z_{2-}) \bar{\beta}_2^* & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} & \bar{0} \\ \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 \bar{\pi}^*(z_1, z_2) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} & \bar{0} \\ \bar{0} & \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2^* \bar{\pi}(z_1, z_{2-}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 & \bar{0} \\ \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1, z_{2-}) \bar{\beta}_2 & \bar{0} & \bar{\beta}_1 \\ \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1^* & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1, z_2) \bar{\beta}_2 & \bar{0} \\ \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{0} & \bar{\alpha}_1 & \bar{0} & \bar{\alpha}_2 \bar{\pi}(z_1, z_2) \bar{\beta}_2 \end{bmatrix} \begin{matrix} z_1 = 0 \\ z_2 = 0 \end{matrix} \quad (43)$$

which looks exactly the same as the square matrix in eq. (38) with the only difference that the sub-blocks $\bar{\alpha}_1$, $\bar{\alpha}_2$, $\bar{\beta}_1$, $\bar{\beta}_2$ and $\bar{\pi}(z_1, z_2)$ in the central column have been replaced respectively by the following new sub-blocks

$$\begin{aligned} \bar{\alpha}_1^* &= \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \frac{i\Omega_1}{2} - \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{i\Omega_1}{2} \\ 0 & 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}, & \bar{\alpha}_2^* &= \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_2}{2} \\ 0 & 0 & 0 & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 \\ 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_2}{2} & 0 & 0 \end{bmatrix}, \\ \bar{\beta}_1^* &= \begin{bmatrix} 0 & 0 & \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ -\frac{i\Omega_1}{2} & \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{i\Omega_1}{2} & 0 & 0 & 0 & 0 \end{bmatrix}, & \bar{\beta}_2^* &= \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_2}{2} \\ 0 & -\frac{i\Omega_2}{2} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}, \quad (44) \\ \bar{\pi}^*(z_1, z_2) &= \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & -\frac{i\Omega_1}{2} & 0 & 0 \\ 0 & 0 & 0 & \frac{i\Omega_1}{2} & 0 & 0 & -\frac{i\Omega_2}{2} & 0 \\ \frac{i\Omega_1}{2} & -\frac{i\Omega_1}{2} (z_1+z_2+\frac{\Gamma_\rho}{2}-i\delta_1) & 0 & \frac{i\Omega_2}{2} & 0 & 0 & 0 & 0 \\ 0 & \frac{i\Omega_2}{2} & 0 & (z_1+z_2+\frac{\Gamma_\rho}{2}-i\delta_R) & 0 & 0 & 0 & \frac{i\Omega_1}{2} \\ 0 & 0 & \frac{i\Omega_2}{2} & 0 & (z_1+z_2+\Gamma_{21}-i\delta_R) & 0 & -\frac{i\Omega_1}{2} & 0 \\ -\frac{i\Omega_1}{2} & \frac{i\Omega_1}{2} & 0 & 0 & (z_1+z_2+\frac{\Gamma_\rho}{2}+i\delta_1) & 0 & -\frac{i\Omega_2}{2} & 0 \\ 0 & -\frac{i\Omega_2}{2} & 0 & 0 & -\frac{i\Omega_1}{2} & 0 & (z_1+z_2+\frac{\Gamma_\rho}{2}+i\delta_2) & 0 \\ 0 & 0 & 0 & \frac{i\Omega_1}{2} & 0 & -\frac{i\Omega_2}{2} & 0 & (z_1+z_2+\Gamma_{21}+i\delta_R) \end{bmatrix}. \end{aligned}$$

The generalisation of this method to obtain the matrix in the numerator from the one in the denominator in eq. (30) to the n th-order approximation is straight-forward. We only have to break the $8(2n+1)^2 \times 8(2n+1)^2$ matrix of the eq. (25) into its more elementary 8×8 sub-blocks $\bar{\alpha}_j$, $\bar{\beta}_j$ and $\bar{\pi}(z_1 + ip2\omega_1, z_2 + iq2\omega_2)$ and perform the replacement $\bar{\alpha}_j \rightarrow \bar{\alpha}_j^*$, $\bar{\beta}_j \rightarrow \bar{\beta}_j^*$ and $\bar{\pi}(z_1, z_2) \rightarrow \bar{\pi}(z_1, z_2)^*$ only in its central column.

The result of this operation is just the desired matrix $\overbrace{\pi \cdot \dots \cdot \pi}^{n+1}_{16n(n+1)+1-16n(n+1)+2}(0, 0)$. This represents a further great simplification since the obtainment of the expression of n th-order analytical solution is thus reduced essentially to the determination of a single matrix, i.e., $\underbrace{\pi \cdot \dots \cdot \pi}_{n+1}(0, 0)$, rather than two.

D The mean value of the asymptotic solution for the two-level system

The perturbative method introduced in this paper to analyse the response to the three-level system driven by two laser fields has a straight-forward generalisation to the case of a system of $n + 1$ levels driven by n laser fields. The main difference being, apart from the fact that the Bloch evolution equations are different for each case, the need to introduce n time variables, e.g. t_1, t_2, \dots, t_n , each for each laser field, and the use of a multiple-Laplace transform with n Laplace independent variables, e.g. z_1, z_2, \dots, z_n . The main equation of this generic case will thus have the following form

$$\bar{\alpha}_1 \bar{\rho}(z_1 - i2\omega_1, z_2, \dots, z_n) + \dots + \bar{\alpha}_n \bar{\rho}(z_1, z_2, \dots, z_n - i2\omega_n) + \bar{\pi}(z_1, z_2, \dots, z_n) \bar{\rho}(z_1, z_2, \dots, z_n) + \bar{\beta}_1 \bar{\rho}(z_1 + i2\omega_1, z_2, \dots, z_n) + \dots + \bar{\beta}_n \bar{\rho}(z_1, z_2, \dots, z_n + i2\omega_n) = \bar{\chi}(z_1, z_2, \dots, z_n), \quad (45)$$

where $\omega_1, \omega_2, \dots, \omega_n$ are the angular frequencies of each respective laser field driving each corresponding transition. From this point on, the theory will follow similar steps.

A most interesting case to study for its simplicity is the two-level system driven by one laser field. This system has already been thoroughly studied in the literature and can also be used to check old results. For this case, there is no need to introduce extra time variables rather than the real time t . And the main equation obtained after performing a simple Laplace transform \mathcal{L} has the following form

$$\bar{\alpha} \bar{\rho}(z - i2\omega) + \bar{\pi}(z) \bar{\rho}(z) + \bar{\beta} \bar{\rho}(z + i2\omega) = \bar{\chi}(z), \quad (46)$$

where ω is the angular frequency of the laser field. Thus, for example, in the RWA, the equation to solve is

$$\bar{\pi}(z) \bar{\rho}(z) = \bar{\chi}(z), \quad (47)$$

or considering the explicit form of these matrices for this case

$$\begin{bmatrix} (z + \Gamma) & i\Omega/2 & -i\Omega/2 \\ i\Omega & (z + \gamma - i\delta) & 0 \\ -i\Omega & 0 & (z + \gamma + i\delta) \end{bmatrix} \begin{bmatrix} \tilde{\rho}_{11}(z) \\ \tilde{\rho}_{21}(z) \\ \tilde{\rho}_{12}(z) \end{bmatrix} = \begin{bmatrix} \frac{\Gamma}{z} \\ +i\frac{\Omega}{2} \frac{1}{z} + i\frac{\Omega}{2} \frac{1}{z-i2\omega} \\ -i\frac{\Omega}{2} \frac{1}{z} - i\frac{\Omega}{2} \frac{1}{z+i2\omega} \end{bmatrix}, \quad (48)$$

where the relaxation parameters Γ and $\gamma = \Gamma/2$ are the population decay of the upper level $|2\rangle$ and the decay rate of the coherence between the two levels $|2\rangle$ and $|1\rangle$. The parameters $\delta = \omega - \omega_0$ and Ω are respectively the detuning of the laser field from its resonance and its Rabi frequency. Also note that $\tilde{\rho}_{ij}(z) = \mathcal{L}\tilde{\rho}_{ij}(t)$, where $\tilde{\rho}_{ij}(t)$ are the matrix elements of the density operator expressed in the “rotating frame”. In the Laboratory frame the corresponding matrix elements of this operator are $\rho_{ij}(t)$.

Then let us use this simple case of the two-level system in its lowest order of approximation, and whose mathematical description is given by eq. (48), to show that, for example, terms containing the factors

$$\frac{1}{z \pm i2\omega} \quad (49)$$

in the column matrix on the right-hand side of eq. (48) generate oscillatory contribution to the solution around the average. Let us start by showing, for example, how to equate and inverse-transform the unknown $\tilde{\rho}_{11}(z)$ of this set of simultaneous equations. Essentially, we solve a ratio of two determinants. In the denominator of this quotient, we have $\|\bar{\pi}(z)\|$, where the double bars indicate, as usual, determinant of the matrix standing between them. The matrix to use in the numerator is obtained from $\bar{\pi}(z)$ by replacing its first column by the column matrix on the right-hand side of eq. (48). Thus expanding the determinant in the numerator in a linear combination of the coefficients of its first column, we have

$$\tilde{\rho}_{11}(z) = \dots + \left(\frac{i\Omega}{2} \frac{1}{z} + \frac{i\Omega}{2} \frac{1}{z - i2\omega} \right) \frac{\begin{vmatrix} i\Omega/2 & -i\Omega/2 \\ 0 & z + \gamma + i\delta \end{vmatrix}}{\|\bar{\pi}(z)\|} + \dots, \quad (50)$$

where the factor between parenthesis is one of the coefficients of the column matrix on the right-hand side of eq. (48). The other terms missing in this expression have the same form as the one shown there, and thus their explicit consideration would result redundant in this analysis.

On the other hand, we have that the inverse-Laplace transform function of $\tilde{\rho}_{11}(z)$ is

$$\rho_{11}(t) = \mathcal{L}^{-1}\{\rho_{11}(z)\} = \frac{1}{2\pi i} \int_{-i\infty + \tau_o}^{+i\infty + \tau_o} \tilde{\rho}_{11}(z) e^{zt} dz, \quad (51)$$

where τ_o is a positive real number such that $\tilde{\rho}(z)$ with $z = x + iy$ is analytical in the strip $0 < x < \tau_o$. This integral may be evaluated by the calculus of residues. For $t > 0$, we may chose the contour by a semicircle in the left half of the complex plane. Thus we can write

$$\rho_{11}(t) = \sum_{\text{poles}} \text{Res} \{ \tilde{\rho}_{11}(z) e^{zt} \}, \quad (52)$$

where the only poles considered in this sum are those with their real part not positive, e.g. $\text{Re}(z_p) \leq 0$. Now, if we substitute eq. (50) into eq. (52), we find

$$\rho_{11}(t) = \dots + \sum_{\text{poles}} \text{Res} \left\{ \frac{i\Omega}{2} \frac{e^{zt}}{z} \frac{\|\bar{a}(z)\|}{\|\bar{\pi}(z)\|} \right\} + \sum_{\text{poles}} \text{Res} \left\{ \frac{i\Omega}{2} \frac{e^{zt}}{z - i2\omega} \frac{\|\bar{a}(z)\|}{\|\bar{\pi}(z)\|} \right\} + \dots, \quad (53)$$

where we have also distributed $\|\bar{a}(z)\|/\|\bar{\pi}(z)\|$ inside the terms between parenthesis in eq. (50). The matrix $\bar{a}(z)$ is the one between double bars in the numerator in the only term shown in eq. (50).

Now we proceed to show that the mean value $\langle \rho_{11}(t \rightarrow \infty) \rangle$ of the asymptotic solution is obtained from eq. (53) by only considering the poles $z_p = 0$ in each calculation of residue there. This is readily apparent by a simple inspection of this equation. It is evident that poles $z_p \neq 0$ and with its real part equal to zero, for example $z_p = i2\omega$ in the second term of eq. (53), generate oscillatory contributions that disappear when averaging over a cycle. On the other hand, the poles located in the left part of the complex plane, e.g. $\text{Re}(z_p) < 0$, yield oscillatory contribution that decay to zero as $t \rightarrow \infty$. The other poles remain outside the integration contour. Thus remaining with only the poles at $z_p = 0$, we write

$$\begin{aligned} \langle \rho_{11}(t \rightarrow \infty) \rangle &= \dots + \sum_{z_p=0} \text{Res} \left\{ \frac{i\Omega}{2} \frac{e^{zt}}{z} \frac{\|\bar{a}(z)\|}{\|\bar{\pi}(z)\|} \right\} \\ &+ \sum_{z_p=0} \text{Res} \left\{ \frac{i\Omega}{2} \frac{e^{zt}}{z - i2\omega} \frac{\|\bar{a}(z)\|}{\|\bar{\pi}(z)\|} \right\} + \dots \end{aligned} \quad (54)$$

But since the determinant

$$||\bar{\pi}(z)|| = (z + \Gamma) ((z + \gamma)^2 + \delta^2) + (z + \gamma)\Omega^2, \quad (55)$$

where the parameters $\Gamma, \gamma, \Omega > 0$, has not any zero with value $z_p = 0$, the second calculation of residue in eq. (54) vanishes for not having poles to evaluate. Thus we finally find

$$\langle \rho_{11}(t \rightarrow \infty) \rangle = \sum_{z_p=0} \text{Res} \{ \tilde{\rho}_{11}(z) e^{zt} \} = \dots + \frac{i\Omega}{2} \frac{||\bar{\alpha}(0)||}{||\bar{\pi}(0)||} + \dots, \quad (56)$$

where we have also computed the residue left.

This analysis shows that the consideration of only the poles at $z_p = 0$ in the calculation of the residues of the function $\tilde{\rho}_{ij}(z)e^{zt}$ is equivalent to evaluate the matrix $\bar{\pi}(z)$ at $z = 0$ and neglect terms containing the factor

$$\frac{1}{z \pm i2\omega} \quad (57)$$

in the column matrix on the right-hand side of eq. (48). Thus with this simplification, the equation to solve becomes

$$\begin{bmatrix} (z + \Gamma) & i\Omega/2 & -i\Omega/2 \\ i\Omega & (z + \gamma - i\delta) & 0 \\ -i\Omega & 0 & (z + \gamma + i\delta) \end{bmatrix}_{z=0} \begin{bmatrix} \tilde{\rho}_{11}(z) \\ \tilde{\rho}_{21}(z) \\ \tilde{\rho}_{12}(z) \end{bmatrix} = \frac{1}{2z} \begin{bmatrix} 2\Gamma \\ +i\Omega \\ -i\Omega \end{bmatrix}. \quad (58)$$

The solution of this matrix equation gives directly, as its inverse-Laplace transform, the mean value of the state of the system as $t \rightarrow \infty$ in the RWA. For example, the population $\langle \rho_{11}(t \rightarrow \infty) \rangle$ can be written as

$$\langle \rho_{11}(t \rightarrow \infty) \rangle = \mathcal{L}^{-1} \left(\frac{1}{z} \frac{||\bar{\pi}_1(0)||}{||\bar{\pi}(0)||} \right) = \frac{||\bar{\pi}(0)||}{||\bar{\pi}(0)||}, \quad (59)$$

where the matrix $\bar{\pi}(0)$ is the three-by-three matrix of eq. (58) with $z = 0$ and $\bar{\pi}_1(0)$ is the matrix obtained from $\bar{\pi}(0)$ by replacing its first column by the column matrix on the right-hand side of eq. (58). Note the similarity between eq. (59) for this case and eq. (27) for the case of the three-level system.

E Choice of ω_{21}

In Sect. 5.2, the reason for the specific choice of $0.04296875 = 176 \times 2^{-12}$ for the difference of Bohr frequencies is merely a computational question. The number 2^{-12} is the step increment in a *for-loop* used to change the frequency of laser 1, e.g. ω_1 . Computationally, the number 2^{-12} , together with its multiples, can be represented exactly in base 2 by a finite number of digits, e.g. $(2^{-12})_{10} = 0.00000000001_2$. This means that they can be exactly stored in a double-precision variable, and consequently the variation of ω_1 can be realised in precise increments in the *for-loop*. This is essential to ensure that the whole signal generated by both lasers together remains always a periodic function for each iteration of the *for-loop*. For example, at optical resonance condition $\omega_1 = \omega_{01} = 1 = n 2^{-12}$ and $\omega_2 = \omega_{02} = 1 - 0.4296875 = m 2^{-12}$ where n and m are respectively the following integers $n = 2^{12} = 4096$ and $m = 3920$. Therefore the ratio of the laser frequencies in this case is a rational number, e.g. $\omega_1/\omega_2 = n/m$, guaranteeing the periodicity of the whole signal. This property is preserved as long as the frequencies of the laser fields are varied in multiples of 2^{-12} . The need for a periodic signal is related to the fact that the ultimate information needed is the mean value of the asymptotic solution.

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