Optical absorption and emission in simple systems: Beyond the rotating-wave approximation

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Some model Hamiltonians of simple material systems in interaction with a radiation field are exactly solved by a new generalized recursion method. The linear and nonlinear optical responses are computed.

I. INTRODUCTION

In the past ten years highly idealized material models, namely, two-level atoms in a single electromagnetic mode and three-level atoms in two electromagnetic modes, have been the object of detailed theoretical investigations.[1-14] Fundamental effects observed in real systems, for instance, Rabi nutation, superradiant effects, and so on, are explained by these idealized models.

In optical experiments, if the material system is excited very close to an electronic or vibrational transition, the model Hamiltonian that describes its behavior is usually solved under the so-called rotating-wave approximation (RWA) and the dipole approximation completes the framework usually adopted.

Swein[2] in his pioneering paper of 1973 exactly solved the two-level atom model in a one-mode electromagnetic field (without using the RWA) by a continued fraction algorithm. In the same article the author underlined that the RWA is fully justified in the following two cases: (a) when electron-photon coupling is much lower than the photon energy, and (b) for short-time response of the system when the aperiodic nature of the continued fraction solution was not evident.

The recent development of Rydberg atom studies[15] has allowed the preparation of two-level atom systems strongly coupled to the radiation field. In these systems the coupling between the atom and the single electromagnetic mode becomes the dominant process in the time evolution. Moreover, when the photon energy is not very close to an energy transition of the material systems the effect of the counter-rotating part of the matter-radiation interaction would not be negligible.

Finally, I would emphasize that the RWA is a completely insufficient framework to explain the behavior of real systems in multiphoton spectroscopy and in nonlinear quantum optics.[16,17] Moreover, very recently,[17] the dependence of the photon absorption cross-section values of molecules from the gauge adopted for the radiation-matter interaction has been pointed out. All these considerations lead to the conclusion that exact solutions of model Hamiltonians are particularly interesting in quantum optics.

In this paper I will show that the excitation amplitude method can solve exactly a large number of model Hamiltonians well suited to quantum optic problems. The solution consists of two steps: (a) the excitation amplitudes of the model Hamiltonian are obtained as a system of recurrence equation, and (b) the solution is given in closed analytical form by a generalized operator. Since this operator is model dependent, I will show how to obtain this operator for a large class of model Hamiltonians recently studied in the literature under the RWA.[3-14]

In Sec. II the general outline of the excitation amplitude method will be given and in Sec. III the probability of optical absorption and emission will be computed in the present framework. In Sec. IV I will use the excitation amplitude method to obtain exact solutions for a set of model Hamiltonians well suited to quantum optic problems. In the same section I will underline which model Hamiltonians have an exact solution available in the literature and which models are solved for the first time. In Sec. V selected numerical examples will be discussed recovering well-known and new results; moreover, the convergence of the method will be addressed. The conclusions will be given in Sec. VI.

II. METHOD OF EXCITATION AMPLITUDES

Recently the method of excitation amplitude has been used to solve a large class of physical phenomena.[18-23] The method is able to solve exactly electron-boson interaction in closed electronic shells. Now, very briefly I will review the general procedure to obtain the recursion equations and the generalized operator. For a more exhaustive discussion of the method see Ref. 18.

Let me consider a total Hamiltonian of a material system in interaction with a radiation field,

$$H = H_0 + H_I \ , \tag{1}$$

where the unperturbed Hamiltonian H_0 is composed of material system Hamiltonian H_M and radiation Hamiltonian,

$$H_R = \sum_i \hbar\omega_i a_i^\dagger a_i \ , \tag{2}$$

where $\hbar\omega_i$ is the energy of the ith photon of the electromagnetic field and $a_i^\dagger$ its creation operator. The nonlinear interaction Hamiltonian in the dipole approximation is

$$H_I = \sum_i \sum_n (M_n a_i^\dagger + M_n^* a_i)^n L_i \ , \tag{3}$$

where M_n are the matrix elements of nonlinear susceptibility expansion and the dependence on electronic degrees of freedom is contained in the operators,

$$L_i = \sum_{\alpha\beta} \Lambda(i,\alpha,\beta) P_{\alpha\beta} , \tag{4}$$

where $|\alpha\rangle$ and $|\beta\rangle$ are the electronic configurations and $P_{\alpha\beta}=|\alpha\rangle\langle\beta|$. The quantities $\Lambda(i,\alpha,\beta)$ characterize the model of the material system.

According to the physical problem, we are interested in computing the following excitation amplitudes:

$$\Phi_{\alpha\beta}(E)=\langle P_{\alpha\alpha}(E-H)^{-1}P_{\beta\beta}\rangle , \tag{5}$$

where the angular brackets denote the average on a coherent photon state or an eigenstate of photon number.

The method allows the computation of the excitation amplitudes $\Phi_{\alpha\beta}$ by a system of recursive equations obtained by the combined use of two operator identities, namely,

$$(E-H)^{-1}=(E-H_0)^{-1}+(E-H_0)^{-1}H_I(E-H)^{-1} , \tag{6}$$

$$a_i^n H_0^m=(H_0+n\hbar\omega_i)^m a_i^n . \tag{7}$$

The last step of the method concerns the solution of recursive equations in closed form by a continued fraction or a more general mathematical algorithm. A more exhaustive presentation of the method, embodying the convergence criteria and the comparison with other recursive methods available in the literature is discussed in Ref. 18. In the present paper I prefer to discuss the convergence of the method through physical arguments, rather than resorting to rigorous mathematical theorems. Moreover, some review articles, which have recently appeared in the literature,[24,25] address the general problem of convergence in recursive methods.

III. OPTICAL ABSORPTION AND EMISSION

Let me consider a material system in its initial state $|i\rangle$ at time $t=0$. The transition probability of reaching the final state $|f\rangle$ at the time $t>0$ under laser irradiation will be

$$P_{i\to f}(t)=|\langle f|i(t)\rangle|^2 , \tag{8}$$

where,

$$\langle f|i(t)\rangle=\langle f|\exp(-iHt)|i\rangle = -\frac{1}{2\pi i}\oint dE\, G_{fi}(E)e^{-iEt} . \tag{9}$$

The integral of Eq. (9) can be computed by the theorem of residues (the path integral is above the real axis and encloses all the poles of the integral function[2]). The final state can be given as a general expansion in photon number $|f\rangle=\sum_n A_n|f,n\rangle$, where f characterizes the electronic state and n is the photon number, the function $G_{fi}(E)$ is

$$G_{fi}(E)=\langle f|(E-H)^{-1}|i\rangle=\sum_n A_n\Phi_{fi}^n(E) , \tag{10}$$

where the transition amplitudes are

$$\Phi_{fi}^n(E)=\langle f,n|(E-H)^{-1}|i\rangle , \tag{11}$$

and the initial state $|i\rangle$ can be a coherent state.

The present method can give exact solutions of model Hamiltonians recently published in the literature[3-14] in a unified mathematical scheme, as I will show in Sec. IV.

IV. THEORY

Schweber[1] in his pioneering paper of 1967 exactly solved the two-level system under one-mode laser irradiation. Some years later, Swain[2] exactly solved the same problem, by using the equation-of-motion approach to generate a continued fraction solution. Later on, the solution for three levels and one boson and three levels and two bosons under the RWA was published in the literature.[3-14] In a recent paper[18] the general problem of N levels and M bosons was addressed by the present author.

Now I will recover by the excitation amplitude method the exact solution for two levels and one mode, showing the equivalence with Swein's results and then I will give the exact solution for the following systems: three levels and one mode, two levels and two modes, three levels and two modes, and two levels under nonlinear photon interaction. These methods will be studied in order of increasing complexity of the mathematical algorithm involved in the exact solution.

A. Two levels and one mode

The simplest model for the interaction between radiation and matter is that in which a single two-level atom interacts with a single mode of the electromagnetic field and the atomic transition is electric dipole permitted. This system is modeled by the total Hamiltonian,

$$H=H_0+H_I , \tag{12}$$

$$H_0=\varepsilon_1 c_1^\dagger c_1+\varepsilon_2 c_2^\dagger c_2+\omega_0 a^\dagger a , \tag{13}$$

$$H_I=(M_0^* a+M_0 a^\dagger)(c_1^\dagger c_2+c_2^\dagger c_1) , \tag{14}$$

where M_0 is the matrix element of dipole moment, ε_1 and ε_2 are the energies of the unperturbed electron levels, and ω_0 is the one-mode laser energy (where $\hbar=1$).

Let me consider a general non-normalized initial state $|i\rangle=(a^\dagger)^{n_\nu}|\nu\rangle$, where the optical electron is at the $\nu=1$ or 2 unperturbed state in presence of n_ν photons of energy ω_0. From the two operatorial identities of Eqs. (6) and (7) and using the relation $\sum_{j=1,2}\sum_n |j,n\rangle\langle j,n|=1$ we obtain the recurrence equations,[18]

$$\begin{aligned}\Phi_2^n(\omega)&=G_2^0(\omega-n\omega_0)[\,n!\delta_{2\nu}\delta_{n\nu}+M_0^*\Phi_1^{n+1}(\omega)\\&\quad+nM_0\Phi_1^{n-1}(\omega)] ,\\ \Phi_1^n(\omega)&=G_1^0(\omega-n\omega_0)[\,n!\delta_{1\nu}\delta_{n\nu}+M_0^*\Phi_2^{n+1}(\omega)\\&\quad+nM_0\Phi_2^{n-1}(\omega)] ,\end{aligned} \tag{15}$$

where the zeroth-order Green functions are

$$G_j^0(\omega-n\omega_0)=\langle j,n|(\omega-\varepsilon_j-H_0)^{-1}|j,n\rangle$$
$$=\langle j|(\omega-\varepsilon_j-H_0-n\omega_0)^{-1}|j\rangle\ , \qquad (16)$$

and the excitation amplitudes are defined by

$$\Phi_j^n(\omega)=\langle j|a^n(\omega-H)^{-1}|i\rangle\ . \qquad (17)$$

Note that I have chosen non-normalized boson states to avoid tedious factorial terms. Finally we can obtain decoupled equations by inserting the second equation (15) into the first one,

$$\Phi_2^n(\omega)=D_2^0(\omega-n\omega_0)[\,n!\delta_{2\nu}\delta_{n\nu}+M_0^*G_1^0(\omega-(n+1)\omega_0)(n+1)!\delta_{1\nu}\delta_{n+1,\nu}$$
$$+nM_0G_1^0(\omega-(n-1)\omega_0)(n-1)!\delta_{1\nu}\delta_{n-1,\nu}$$
$$+(M_0^*)^2G_1^0(\omega-(n+1)\omega_0)\Phi_2^{n+2}(\omega)+n(n-1)M_0^2G_1^0(\omega-(n-1)\omega_0)\Phi_2^{n-2}(\omega)]\ , \qquad (18)$$

where the dressed zeroth-order Green function $D_2^0(\omega)$ is

$$D_2^0(\omega-n\omega_0)=G_2^0(\omega-n\omega_0)/\{1-|M_0|^2G_2^0(\omega-n\omega_0)[(n+1)G_1^0(\omega-(n+1)\omega_0)+nG_1^0(\omega-(n-1)\omega_0)]\} \qquad (19)$$

and analogously for $1\rightarrow2,2\rightarrow1$, due to the symmetry of Eqs. (15).

Equation (18) exactly solves the model; the three-diagonal form obtained gives the solution in closed form by an infinite continued fraction.[18]

Let me consider the system prepared at the time $t=0$ into the excited state $|2\rangle$ with zero photons present in the cavity (in this case I take $\varepsilon_2>\varepsilon_1=0$ and $|\nu\rangle=|2\rangle$); the probability that at $t>0$ the system still remains in the same state $|f(t)\rangle=|i(t)\rangle$ will be

$$P(t)=\left|-\frac{1}{2\pi i}\oint d\omega\,\Phi_2^0(\omega)\exp(-i\omega t)\right|^2\ , \qquad (20)$$

where the excitation amplitude $\Phi_2^0(\omega)$ is given by recursive equations

$$\Phi_2^0(\omega)=G_2^0(\omega)[1+M_0^*\Phi_1^1(\omega)]\ ,$$
$$\Phi_1^1(\omega)=G_1^0(\omega-\omega_0)[M_0^*\Phi_2^2(\omega)+M_0\Phi_2^0(\omega)]\ ,$$
$$\Phi_2^2(\omega)=G_2^0(\omega-2\omega_0)[M_0^*\Phi_1^3(\omega)+2M_0\Phi_1^1(\omega)]\ .$$

Finally, imposing the boundary conditions $n\rightarrow N>|M_0|^2/\omega_0^2$, for an N even integer number,

$$\Phi_2^N(\omega)=NG_2^0(\omega-N\omega_0)\Phi_1^{N-1}(\omega)\ ,$$

we obtain the exact solution in closed form,

$$\Phi_2^0(\omega)=\cfrac{G_2^0(\omega)}{1-\cfrac{|M_0|^2G_2^0(\omega)G_1^0(\omega-\omega_0)}{1-\cfrac{2|M_0|^2G_1^0(\omega-\omega_0)G_2^0(\omega-2\omega_0)}{1-\cfrac{3|M_0|^2G_2^0(\omega-2\omega_0)G_1^0(\omega-3\omega_0)}{1-\cdots}}}}\ . \qquad (21)$$

This formula coincides with Eq. (41) of Ref. 2. If we limit ourselves to the RWA we must consider only the first denominator of Eq. (21), in this case,

$$\Phi_2^0(q)=\frac{1}{q-\dfrac{|M_0|^2}{q-\Delta\omega}}\ , \qquad (22)$$

where $q=\omega-\varepsilon_2$ for $\varepsilon_1=0$ and $\Delta\omega=\omega_0-\varepsilon_2$ is the detuning between electron transition $\varepsilon_2-\varepsilon_1$ and photon energy ω_0. The poles of Eq. (22) are obtained by

$$q-\frac{|M_0|^2}{q-\Delta\omega}=0\ ,$$

as function of Rabi energy $\Omega=(\Delta\omega^2+4|M_0|^2)^{1/2}$; in fact, the poles are $q_\pm=(\Delta\omega\pm\Omega)/2$. Now we can compute the integral of Eq. (20) by the theorem of residues and finally, the probability is

$$P(t)=|[\Omega\cos(\Omega t)+i\Delta\omega\sin(\Omega t)]\exp(-i\Delta\omega t)/\Omega|^2\ .$$

Under resonant condition $\Delta\omega=0$ we obtain the well-known result,

$$P(t)=\cos^2(\Omega t)\ ,$$

where $\Omega=2|M_0|$. In Sec. V I will discuss the correctives to the RWA.

B. Three levels and one mode

The generalization of the two levels to N levels and one boson can be given in closed form as shown in Ref. 18, but the recursive vectorial equations obtained for $N>4$ [see Ref. 18 Eq. (5.14), p. 223] cannot decouple with respect to electronic levels and the numerical solutions become rapidly cumbersome.

The total Hamiltonian of three-level atoms, with different electronic symmetries, in interaction with one-mode electromagnetic field is

$$H=H_0+H_I\ , \qquad (23)$$
$$H_0=\sum_{j=1,3}\varepsilon_jc_j^\dagger c_j+\omega_0a^\dagger a\ , \qquad (24)$$
$$H_I=(M_0^*a+M_0a^\dagger)(c_1^\dagger c_2+c_2^\dagger c_1)$$
$$+(M_1^*a+M_1a^\dagger)(c_2^\dagger c_3+c_3^\dagger c_2)\ , \qquad (25)$$

where M_0 and M_1 are the matrix dipole moments for $1\rightarrow2$ and $2\rightarrow3$ electronic transitions, respectively. Following the same procedure as in the two-level case, we obtain a system of recurrence equations for the excitation amplitudes $\Phi_j^n(\omega)$, where $j=1,2,3$,

$$\Phi_1^n(\omega)=G_1^0(\omega-n\omega_0)[n!\delta_{1\nu}\delta_{n\nu}+M_0^*\Phi_2^{n+1}(\omega)+nM_0\Phi_2^{n-1}(\omega)] ,$$
$$\Phi_2^n(\omega)=G_2^0(\omega-n\omega_0)[n!\delta_{2\nu}\delta_{n\nu}+M_0^*\Phi_1^{n+1}(\omega)+M_1^*\Phi_3^{n+1}(\omega)+n(M_0\Phi_1^{n-1}(\omega)+M_1\Phi_3^{n-1}(\omega))] , \quad (26)$$
$$\Phi_3^n(\omega)=G_3^0(\omega-n\omega_0)[n!\delta_{3\nu}\delta_{n\nu}+M_1^*\Phi_2^{n+1}(\omega)+nM_1\Phi_2^{n-1}(\omega)] .$$

Finally the decoupled equation is

$$\begin{aligned}\Phi_2^n(\omega)=D_2^0(\omega-n\omega_0)\{ & n!\delta_{2\nu}\delta_{n\nu}+M_0^*(n+1)!G_1^0(\omega-(n+1)\omega_0)\delta_{1\nu}\delta_{n+1,\nu}\\ & +M_1^*(n+1)!G_3^0(\omega-(n+1)\omega_0)\delta_{3\nu}\delta_{n+1,\nu}\\ & +nM_0(n-1)!G_1^0(\omega-(n-1)\omega_0)\delta_{1\nu}\delta_{n-1,\nu}+nM_1(n-1)!G_3^0(\omega-(n-1)\omega_0)\\ & +\Phi_2^{n+2}(\omega)[M_0^2G_1^0(\omega-(n+1)\omega_0)+M_1^2G_3^0(\omega-(n+1)\omega_0)]\\ & +n(n-1)\Phi_2^{n-2}(\omega)[M_0^{*2}G_1^0(\omega-(n-1)\omega_0)+M_1^{*2}G_3^0(\omega-(n-1)\omega_0)]\} ,\end{aligned} \quad (27)$$

and the dressed zeroth-order Green function $D_2^0(\omega-n\omega_0)$ is

$$\begin{aligned}D_2^0(\omega-n\omega_0)=G_2^0(\omega-n\omega_0)/\{1-G_2^0(\omega-n\omega_0)[& (n+1)|M_0|^2G_2^0(\omega-(n+1)\omega_0)+(n+1)|M_1|^2G_3^0(\omega-(n+1)\omega_0)\\ & +n|M_0|^2G_1^0(\omega-(n-1)\omega_0)+n|M_1|^2G_3^0(\omega-(n-1)\omega_0)]\} .\end{aligned} \quad (28)$$

The solution is again in the three-diagonal shape and we can compute $\Phi_2^n(\omega)$ by the continued fraction algorithm. Finally, the quantities $\Phi_1^n(\omega)$ and $\Phi_3^n(\omega)$ are computed by Eqs. (26).

C. Two levels and two modes

A simple material system with two bosons involved in its dynamic is the so-called vibrating molecule in the one-mode laser cavity. This model under resonant Raman spectroscopy was studied by the present author,[20,21] in the case of harmonic and anharmonic vibration potential. The Frank-Condon effect was also discussed. The model Hamiltonian is

$$H=H_0+H_I , \quad (29)$$
$$H=\varepsilon_1c_1^\dagger c_1+\varepsilon_2c_2^\dagger c_2+\omega_0a^\dagger a+\Omega_0b^\dagger b , \quad (30)$$
$$H_I=gc_2^\dagger c_2(b^\dagger+b)+(M_0^*a+M_0a^\dagger)(c_2^\dagger c_1+c_1^\dagger c_2) . \quad (31)$$

where Ω_0 is the interatomic vibration energy, ω_0 the photon energy, g the electron-vibration coupling, and M_0 the matrix dipole moment of the electron transition. The solution of the model is given by the following recurrence relations:

$$\begin{aligned}\Phi_2^{n,m}(\omega)&=G_2^0(\omega-n\omega_0-m\Omega_0)\{n!m!\delta_{2\nu}\delta_{n\nu}\delta_{m\nu}+g[\Phi_2^{n,m+1}(\omega)+m\Phi_2^{n,m-1}(\omega)]+M_0^*\Phi_1^{n+1,m}(\omega)+nM_0\Phi_1^{n-1,m}(\omega)\} ,\\ \Phi_1^{n,m}(\omega)&=G_1^0(\omega-n\omega_0-m\Omega_0)[n!m!\delta_{1\nu}\delta_{n\nu}\delta_{m\nu}+M_0^*\Phi_2^{n+1,m}(\omega)+nM_0\Phi_2^{n-1,m}(\omega)] .\end{aligned} \quad (32)$$

Now inserting the second equation into the first one, we obtain the decoupled recurrence equation,

$$\begin{aligned}\Phi_2^{n,m}(\omega)=D_2^0(\omega-n\omega_0-m\Omega_0)\{ & n!m!\delta_{2\nu}\delta_{n\nu}\delta_{m\nu}+M_0^*(n+1)!m!G_1^0(\omega-(n+1)\omega_0-m\Omega_0)\delta_{1\nu}\delta_{n+1,\nu}\delta_{m\nu}\\ & +nM_0(n-1)!m!G_1^0(\omega-(n-1)\omega_0-m\Omega_0)\delta_{1\nu}\delta_{n-1,\nu}\delta_{m\nu}\\ & +g[\Phi_2^{n,m+1}(\omega)+m\Phi_2^{n,m-1}(\omega)]+M_0^{*2}G_1^0(\omega-(n+1)\omega_0-m\Omega_0)\Phi_2^{n+2,m}(\omega)\\ & +n(n-1)M_0^2G_1^0(\omega-(n-1)\omega_0-m\Omega_0)\Phi_2^{n-2,m}(\omega)\} ,\end{aligned} \quad (33)$$

and the zeroth-order Green function $D_2^0(\omega-n\omega_0-m\Omega_0)$ is

$$\begin{aligned}D_2^0(\omega-n\omega_0-m\Omega_0)=G_2^0(\omega-n\omega_0-m\Omega_0)/\{1-G_2^0(\omega-n\omega_0-m\Omega_0) & \\ \times[(n+1)|M_0|^2G_1^0(\omega-(n+1)\omega_0-m\Omega_0) & \\ +n|M_0|^2G_1^0(\omega-(n-1)\omega_0-m\Omega_0)]\} . & \end{aligned} \quad (34)$$

The solution of Eq. (33) is given in closed form by a matrix recursive algorithm. In fact, let me consider for the sake of simplicity the initial state $|i\rangle=|2\rangle$ with zero vibrations and zero photons; in this case Eq. (33) can be written in the form

$$\sum_p \{\delta_{np} - D_2^0(\omega - n\omega_0 - m\Omega_0)[M_0^{*2} G_1^0(\omega - (n+1)\omega_0 - m\Omega_0)\delta_{p,n+2} + n(n-1)M_0^2 G_1^0(\omega - (n-1)\omega_0 - m\Omega_0)\delta_{p,n-2}]\}\Phi_2^{pm}(\omega) - \sum_p gD_2^0(\omega - n\omega_0 - m\Omega_0)\delta_{pn}\Phi_2^{p,m+1}(\omega) - \sum_p mgD_2^0(\omega - n\omega_0 - m\Omega_0)\delta_{pn}\Phi_2^{p,m-1}(\omega) = n!m!\delta_{n0}\delta_{m0}D_2^0(\omega - n\omega_0 - m\Omega_0) . \quad (35)$$

Equation (35) is three diagonal with respect to the m index, but the quantities are matrices of dimension $N \times N$ with respect to the indices p and n (if the recurrence equation has converged for $n \to N$) and the transition amplitudes are column vectors of dimension N with respect to the p index. In this case Eq. (35) is

$$\overleftrightarrow{A}(m)\Phi(m) - \overleftrightarrow{B}(m)\Phi(m+1) - \overleftrightarrow{C}(m)\Phi(m-1) = \mathbf{E}(m) , \quad (36)$$

where the matrices and the vector quantities are given by

$$\begin{aligned}
&\overleftrightarrow{A} \equiv (A_{np}(m)) = \delta_{np} - D_2^0(\omega - n\omega_0 - m\Omega_0)[M_0^{*2} G_1^0(\omega - (n+1)\omega_0 - m\Omega_0)\delta_{p,n+2} \\
&\qquad + n(n-1)M_0^2 G_1^0(\omega - (n-1)\omega_0 - m\Omega_0)\delta_{p,n-2}] , \\
&\overleftrightarrow{B} \equiv (B_{pn}(m)) = gD_2^0(\omega - n\omega_0 - m\Omega_0)\delta_{pn} , \\
&\overleftrightarrow{C} \equiv (C_{pn}(m)) = mgD_2^0(\omega - n\omega_0 - m\Omega_0)\delta_{pn} , \\
&\Phi(m) \equiv (\Phi_2^{pm}(\omega)) ,
\end{aligned} \quad (37)$$

and

$$\mathbf{E}(m) \equiv n!m!D_2^0(\omega - n\omega_0 - m\Omega_0)\delta_{n0}\delta_{m0} .$$

Finally, the solution in closed form is given as continued fraction of matrices, namely,

$$\begin{aligned}
&\Phi(0) = [\overleftrightarrow{F}(0)]^{-1}\mathbf{E}(0) , \\
&\overleftrightarrow{F}(0) = \overleftrightarrow{A}(0) - \overleftrightarrow{B}(0)\{\overleftrightarrow{A}(1) - \overleftrightarrow{B}(1)[\overleftrightarrow{A}(2) - \overleftrightarrow{B}(2)\overleftrightarrow{F}(3)^{-1}\overleftrightarrow{C}(3)]^{-1}\overleftrightarrow{C}(2)\}^{-1}\overleftrightarrow{C}(1) .
\end{aligned} \quad (38)$$

Note that for reasonable values of parameter, g^2/Ω_0^2 the size of matrices $N \times N$ is not too large and the numerical convergence will be reached sufficiently fast by a medium size computer as shown in Ref. 26.

D. Three levels and two modes

A three-level atom in interaction with two electromagnetic modes is a very useful model in quantum optics and it has been given a lot of attention in literature.[7-14] Two-photon absorption and emission and the coherent Raman scattering are spectroscopies that need three levels and two modes as a minimal model. The total Hamiltonian is

$$H = H_0 + H_I , \quad (39)$$

$$H_0 = \sum_{j=1,3} \varepsilon_j c_j^\dagger c_j + \sum_{i=1,2} \omega_i a_i^\dagger a_i , \quad (40)$$

$$H_I = (M_1 a_1^\dagger + M_1^* a_1 + N_2 a_2^\dagger + N_2^* a_2)(c_1^\dagger c_2 + c_2^\dagger c_1) + (M_2 a_2^\dagger + M_2^* a_2 + N_1 a_1^\dagger + N_1^* a_1)(c_2^\dagger c_3 + c_3^\dagger c_2) , \quad (41)$$

where M_i and N_i for $i = 1,2$ are the dipole matrix elements of electron transitions. In the simple case of resonance, where the photon energy ω_1 (ω_2) is very close to the $1 \to 2$ ($2 \to 3$) electron transition, we can assume $N_i \to 0$ for $i = 1,2$ and the solution is obtained by the operatorial identities (6) and (7) as coupled recurrence equations,

$$\begin{aligned}
&\Phi_1^{m,n}(\omega) = G_1^0(\omega - m\omega_1 - n\omega_2)[n!m!\delta_{1\nu}\delta_{m\nu}\delta_{n\nu} + M_1^* \Phi_2^{m+1,n}(\omega) + mM_1\Phi_2^{m-1,n}(\omega)] , \\
&\Phi_2^{m,n}(\omega) = G_2^0(\omega - m\omega_1 - n\omega_2)[n!m!\delta_{2\nu}\delta_{m\nu}\delta_{n\nu} + M_1^* \Phi_1^{m+1,n}(\omega) + mM_1\Phi_1^{m-1,n}(\omega) + M_2^* \Phi_3^{m,n+1}(\omega) \\
&\qquad + nM_2\Phi_3^{m,n-1}(\omega)] , \\
&\Phi_3^{m,n}(\omega) = G_3^0(\omega - m\omega_1 - n\omega_2)[n!m!\delta_{3\nu}\delta_{m\nu}\delta_{n\nu} + M_2^* \Phi_2^{m,n+1}(\omega) + nM_2\Phi_2^{m,n-1}(\omega)] .
\end{aligned} \quad (42)$$

The general solution is obtained by the decoupled recurrence equation,

$$\Phi_2^{mn}(\omega)=D_2^0(\omega-m\omega_1-n\omega_2)[\,n!m!\delta_{2\nu}\delta_{m\nu}\delta_{n\nu}+M_1^* n!(m+1)!G_1^0(\omega-(m+1)\omega_1-n\omega_2)\delta_{1\nu}\delta_{m+1,\nu}\delta_{n\nu}$$
$$+mM_1 n!(m-1)!G_1^0(\omega-(m-1)\omega_1-n\omega_2)\delta_{1\nu}\delta_{m-1,\nu}\delta_{n\nu}$$
$$+M_2^* m!(n+1)!G_3^0(\omega-m\omega_1-(n+1)\omega_2)\delta_{3\nu}\delta_{n+1,\nu}\delta_{m\nu}$$
$$+nM_2 m!(n-1)!G_3^0(\omega-m\omega_1-(n-1)\omega_2)\delta_{3\nu}\delta_{n-1,\nu}\delta_{m\nu}$$
$$+M_1^{*2}\Phi_2^{m+2,n}(\omega)G_1^0(\omega-(m+1)\omega_1-n\omega_2)$$
$$+m(m-1)M_1^2\Phi_2^{m-2,n}(\omega)G_1^0(\omega-(m-1)\omega_1-n\omega_2)$$
$$+M_2^{*2}G_3^0(\omega-m\omega_1-(n+1)\omega_2)\Phi_2^{m,n+2}(\omega)$$
$$+n(n-1)M_2^2G_3^0(\omega-m\omega_1-(n-1)\omega_2)\Phi_2^{m,n-2}(\omega)]\ , \tag{43}$$

and the zeroth-order Green function is

$$D_2^0(\omega-m\omega_1-n\omega_2)=G_2^0(\omega-m\omega_1-n\omega_2)/\{1-G_2^0(\omega-m\omega_1-n\omega_2)[\,|M_1|^2(m+1)G_1^0(\omega-(m+1)\omega_1-n\omega_2)$$
$$+|M_1|^2mG_2^0(\omega-(m-1)\omega_1-n\omega_2)$$
$$+|M_2|^2(n+1)G_2^0(\omega-m\omega_1-(n+1)\omega_2)$$
$$+n|M_2|^2G_2^0(\omega-m\omega_1-(n-1)\omega_2)]\}\ . \tag{44}$$

Finally, the solution in closed form for the vector $\Phi(n)\equiv[\Phi_2^{pn}(\omega)]$ is given by the recursive equation (36) using the same procedure as in Sec. IV C to obtain the matrices $\overleftrightarrow{A}$, $\overleftrightarrow{B}$, and $\overleftrightarrow{C}$. The quantities $\Phi_1^{mn}(\omega)$ and $\Phi_3^{mn}(\omega)$ are computed by Eq. (42).

If the system is out of resonance we cannot associate each photon to a particular electron transition and we have to solve the full interaction Hamiltonian as given in Eq. (41). The exact solution is more involved than that of Eqs. (43) and (44) and the explicit form is given for the first time in the Appendix.

E. Nonlinear optic in two-level system

The potential of the method is completely developed by solving exactly nonlinear optic models. In these cases the RWA fails to give sensible results.

Let me consider the simplest model, namely, a two-level atom interacting with a one-mode electromagnetic field. The interaction embodies linear and quadratic electric fields. The model Hamiltonian is

$$H=H_0+H_I\ , \tag{45}$$
$$H_0=\varepsilon_1 c_1^\dagger c_1+\varepsilon_2 c_2^\dagger c_2+\omega_0 a^\dagger a\ , \tag{46}$$
$$H_I=[(M_0 a^\dagger+M_0^* a)+(M_1 a^\dagger+M_1^* a)^2](c_1^\dagger c_2+c_2^\dagger c_1)\ , \tag{47}$$

where M_0 and M_1 are correlated to the diagonal part of linear and nonlinear susceptibilities. In this case the two operator identities of Eqs. (6) and (7) give symmetric recursive equations,

$$\Phi_2^n(\omega)=A_2^n\Phi_1^{n-2}(\omega)+B_2^n\Phi_1^{n-1}(\omega)+C_2^n\Phi_1^n(\omega)+D_2^n\Phi_1^{n+1}(\omega)+E_2^n\Phi_1^{n+2}(\omega)+F_2^n\ ,$$
$$\Phi_1^n(\omega)=A_1^n\Phi_2^{n-2}(\omega)+B_1^n\Phi_2^{n-1}(\omega)+C_1^n\Phi_2^n(\omega)+D_1^n\Phi_2^{n+1}(\omega)+E_1^n\Phi_2^{n+2}(\omega)+F_1^n\ , \tag{48}$$

where the coefficients for $i=1,2$ are

$$A_i^n=M_1^2 n(n-1)G_i^0(\omega-n\omega_0)\ ,$$
$$B_i^n=M_0 nG_i^0(\omega-n\omega_0)\ ,$$
$$C_i^n=(2n+1)|M_1|^2G_i^0(\omega-n\omega_0)\ ,$$
$$D_i^n=M_0^*G_i^0(\omega-n\omega_0)\ , \tag{49}$$
$$E_i^n=M_1^{*2}G_i^0(\omega-n\omega_0)\ ,$$
$$F_i^n=n!\delta_{i\nu}\delta_{n\nu}G_i^0(\omega-n\omega_0)\ .$$

Inserting one recurrence equation into the other, we obtain the decoupled recurrence equations for $i=1$ or 2,

$$\Phi_i^n=\alpha_i^n+\beta_i^n\Phi_i^{n-4}+\gamma_i^n\Phi_i^{n-3}+\delta_i^n\Phi_i^{n-2}+\varepsilon_i^n\Phi_i^{n-1}+\zeta_i^n\Phi_i^{n+1}+\eta_i^n\Phi_i^{n+2}+\theta_i^n\Phi_i^{n+3}+\kappa_i^n\Phi_i^{n+4}\ , \tag{50}$$

where the coefficients for $i=2$ are

$$\alpha_2^n=[\,F_2^n+(A_2^nF_1^{n-2}+B_2^nF_1^{n-1}+C_2^nF_1^n+D_2^nF_1^{n+1}+E_2^nF_1^{n+2})]/P_2^n\ ,$$
$$\beta_2^n=A_2^nA_1^{n-2}/P_2^n\ ,$$
$$\gamma_2^n=(A_2^nB_1^{n-2}+B_2^nA_1^{n-1})/P_2^n\ ,$$
$$\delta_2^n=(A_2^nC_1^{n-2}+B_2^nB_1^{n-1}+C_2^nA_1^n)/P_2^n\ ,$$
$$\varepsilon_2^n=(A_2^nD_1^{n-2}+B_2^nC_1^{n-1}+C_2^nB_1^n+D_2^nA_1^{n+1})/P_2^n\ , \tag{51}$$
$$\zeta_2^n=(B_2^nE_1^{n-1}+C_2^nD_1^n+D_2^nC_1^{n+1}+E_2^nB_1^{n+2})/P_2^n\ ,$$
$$\eta_2^n=(C_2^nE_1^n+D_2^nD_1^{n+1}+E_2^nC_1^{n+2})/P_2^n\ ,$$
$$\theta_2^n=(D_2^nE_1^{n+1}+E_2^nD_1^{n+2})/P_2^n\ ,$$
$$\kappa_2^n=E_2^nE_1^{n+2}/P_2^n\ ,$$

and

$$P_2^n=[1-(A_2^nE_1^{n-2}+B_2^nD_1^{n-1}+C_2^nC_1^n+D_2^nB_1^{n+1}+E_2^nA_1^{n+2})] .$$

Note that the coefficients of Eq. (50) for $i=1$ are obtained from Eqs. (51) changing $1\rightarrow 2$ and $2\rightarrow 1$.

The recurrence equation (50) falls into a class of equations that can give the exact solution by using an algorithm more general than the continued fraction; in fact, we have to diagonalize a non-Hermitian nine-diagonal matrix.

To proceed we cast Eq. (50) in the following simpler form:

$$\Phi_i^n=\alpha_i^n+a_i^n\Phi_i^{n-4}+b_i^n\Phi_i^{n-3}+c_i^n\Phi_i^{n-2}+d_i^n\Phi_i^{n-1} , \quad (52)$$

where the coefficients a_i, b_i, c_i, and d_i are

$$\begin{aligned} a_i^n&=\beta_i^n/D_i^n ,\\ b_i^n&=(\gamma_i^n+E_i^na_i^{n+1})/D_i^n ,\\ c_i^n&=(\delta_i^n+F_i^na_i^{n+2}+E_i^nb_i^{n+1})/D_i^n ,\\ d_i^n&=(\varepsilon_i^n+G_i^na_i^{n+3}+F_i^nb_i^{n+2}+E_i^nc_i^{n+1})/D_i^n , \end{aligned} \quad (53)$$

and the functions D_i^n, E_i^n, F_i^n, and G_i^n are

$$\begin{aligned} D_i^n&=1-\kappa_i^na_i^{n+4}-G_i^nb_i^{n+3}-F_i^nc_i^{n+2}-E_i^nd_i^{n+1} ,\\ E_i^n&=\xi_i^n+\kappa_i^nb_i^{n+4}+G_i^nc_i^{n+3}+F_i^nd_i^{n+2} ,\\ F_i^n&=\eta_i^n+\kappa_i^nc_i^{n+4}+G_i^nd_i^{n+3} ,\\ G_i^n&=\theta_i^n+\kappa_i^nd_i^{n+4} . \end{aligned} \quad (54)$$

Equations (52)–(54) completely solve the model Hamiltonian of Eqs. (45)–(47), taking into account the boundary conditions for $n\rightarrow N\rightarrow\infty$,

$$a_i^N=\beta_i^N,\quad b_i^N=\gamma_i^N,\quad c_i^N=\delta_i^N,\quad d_i^N=\varepsilon_i^N . \quad (55)$$

V. DISCUSSION

The first point to emphasize is that the present method can solve a large class of electron-boson interaction Hamiltonians and I have shown only a few well-known models. For example, we can solve the Hamiltonians of Sec. IV when two- or three-level atoms are in nonradiative interaction with a nonmetallic solid surface (closed bands) as shown in Refs. 19–21 or a vibrating molecule in a two-mode laser field.

The second, more formal point concerns the mathematical method. While obtaining the recurrence system of equations from the model Hamiltonian is always possible using the operator identities of Eqs. (6) and (7), finding the exact solution in closed form is not always possible. In particular cases, the number of unknowns in the recurrence equations can diverge increasing n and we have to introduce a cutoff in order to numerically solve the problem.

Finally, under resonance conditions we can easily recover the solutions obtained by the RWA, as shown in Sec. IV A. Moreover, as pointed out by Swein, while the probability $P(t)$ computed by the RWA [Eq. (22)] is a periodic function of the time, the exact solution of Eq. (21) describes an almost periodic function and this different time behavior would be evident in the long-time response even for very small electron-photon coupling.

The absorption and emission probabilities $P(t)$ of Eqs. (8) and (9) are computed by the two following steps: (a) to find the poles of excitation amplitude $\Phi(\omega)$, and (b) to compute the probability $P(t)$ by the residues theorem. Point (a) can be performed numerically and usually the convergence of the method is very fast for "screening" energy $|M_0|^2/\omega_0$ not much greater than ω_0. Note that we can perform the pole calculations analytically for the continued fraction solution. In fact, imposing the boundary conditions (for $n=N$), the excitation amplitude $\Phi_2^{(0)}(\omega)$ is the Nth Padé approximant[25] of the continued fraction of Eq. (21) and the poles are roots of an Nth-order equation, obtained as product of Padé approximants from zero to N,

$$F(\omega)=[\Phi_2^{(0)}(\omega)\Phi_1^1(\omega)\Phi_2^2(\omega)\Phi_1^3(\omega)\cdots\Phi_2^N(\omega)]^{-1} , \quad (56)$$

where N is an even integer number. Moreover, the analytical solution can be obtained by using a computer with a good symbolic mathematical software that can perform algebraic operations.

The convergence of the method is given by physical considerations. In fact, increasing the number of bosons $n\rightarrow N$, the energy shift $-N\omega_0$ of the zeroth-order Green function moves the pole far from the characteristic energies of the system ε_i for $i=1,2,\ldots$; thus the effect of the $(N+1)$ term on the response must be negligible. In this case we can bound the Hilbert space to the Nth dimension, making a very small error.

Now let me consider more closely the two levels in a one-mode electromagnetic field. The first correction to the RWA is given by Eq. (21) with $N=2$. The poles are roots of the following equation:

$$q^3-q^2(2\omega_0+\Delta\omega)+q(2\omega_0\Delta\omega-3|M_0|^2)+2\omega_0|M_0|^2=0 , \quad (57)$$

where $q=\omega-\varepsilon$ and $\Delta\omega=\varepsilon-\omega_0$ is the energy detuning. Under condition $|M_0|^2/\omega_0<\omega_0$ and not far from resonance ($\Delta\omega\ll\omega_0$), the probability of radiative decay at $t>0$ is $1-P(t)$ and

$$P(t)=\left|\frac{q'_+-\Delta\omega'}{\Omega'+|M_0|^2q'_+/\omega_0}\exp(-iq'_+t)+\frac{q'_--\Delta\omega'}{-\Omega'+|M_0|^2q'_-/\omega_0}\exp(-iq'_-t)-\frac{2|M_0|^2}{(2\omega_0-q'_+)(2\omega_0-q'_-)}\exp(-2i\omega_0t)\right|^2 , \quad (58)$$

where $\Delta\omega'=\Delta\omega-|M_0|^2/\omega_0$, $q'_\pm=(\Delta\omega'\pm\Omega')^{1/2}$. The new Rabi energy is $\Omega'=[\Delta\omega'+4|M_0|^2]^{1/2}$.

Finally, I would underline that the first correction to the RWA for two levels in one mode gives an energy shift of $|M_0|^2/\omega_0$ (Bloch-Siegert shift) and a new term appears in the optical response at the $2\omega_0$ energy.

Now let me consider the three-level case in one electromagnetic mode with initial state $(t=0)|\nu\rangle=|\varepsilon_3,n=0\rangle$. The system of Eqs. (26) under the RWA is reduced to

$$\Phi_3^0(\omega)=G_3^0(\omega)[1+M_1^*\Phi_2^1(\omega)]\ ,$$
$$\Phi_2^1(\omega)=G_2^0(\omega-\omega_0)[M_0^*\Phi_1^2(\omega)+M_1\Phi_3^0(\omega)]\ , \quad (59)$$
$$\Phi_1^2(\omega)=2M_0G_1^0(\omega-2\omega_0)\Phi_2^1(\omega)\ .$$

If the unperturbed electron energies are $\varepsilon_2=0$, $\varepsilon_3=-\varepsilon_1=\varepsilon$ and the detuning is $\Delta\omega=\omega_0-\varepsilon$, the probability at time $t>0$ that the system will be in the state $|v\rangle$ is given by the excitation amplitude,

$$\Phi_3^0(z)=\frac{z^2-3\Delta\omega z+2\Delta\omega^2-2|M_0|^2}{z^3-3\Delta\omega z^2+(2\Delta\omega^2-2|M_0|^2-|M_1|^2)z+2\Delta\omega|M_1|^2}\ , \quad (60)$$

where $z=\omega-\varepsilon$. The poles of Eq. (60) are roots of the third-order equation,

$$Z^3-3\Delta\omega z^2+(2\Delta\omega^2-2|M_0|^2-|M_1|^2)z+2\Delta\omega|M_1|^2=0\ .$$

Under resonance condition $\Delta\omega=0$ and with dipole moment $|M_0|=|M_1|=|M|$, the poles of $\Phi_3^0(\omega)$ are $\omega_1=\varepsilon-|M|\sqrt{3}$, $\omega_2=\varepsilon$, and $\omega_3=\varepsilon+|M|\sqrt{3}$. The probability of radiative decay for three levels and one mode is $1-P(t)$, where

$$P(t)=|[\exp(-i\omega_1 t)+4\exp(-i\omega_2 t)+\exp(-i\omega_3 t)]/6|^2\ . \quad (61)$$

Finally, I would emphasize that the present method easily recovers approximated results in quantum optic systems recently published in the literature[3-14] and can give a quantitative estimation of terms neglected by using the RWA.

VI. CONCLUSIONS

A set of model Hamiltonians, well suited by computing optical stimulated absorption and emission in molecular and atomic systems, are exactly solved by a new recursive mathematical method. The general problem of N-atomic levels in M-electromagnetic modes (for N and M integer numbers) can be addressed by the present method and the exact solution obtained is free of the RWA. Moreover, nonlinear optic effects are computed by the present method for two-level atoms under one-mode laser irradiation and the exact result is given for the first time in closed analytical form.

APPENDIX

I will compute the exact solution of the model Hamiltonian of Eqs. (39)–(41) out of the resonance case $N_i\neq 0$ with $i=1,2$. This solution is given by a generalized five-diagonal matrix. The numerical solution of this algorithm is not time consuming for a medium-sized computer when the system is not very far from the resonance.

The two operatorial identities of Eqs. (6) and (7) give the following recursive system of equations:

$$\Phi_1^{mn}(\omega)=G_1^0(\omega-m\omega_1-n\omega_2)[\delta_{1v}\delta_{mv}\delta_{nv}n!m!+M_1^*\Phi_2^{m+1,n}(\omega)+mM_1\Phi_2^{m-1,n}(\omega)+nN_2\Phi_2^{m,n-1}(\omega)+N_2^*\Phi_2^{m,n+1}(\omega)]\ , \quad (A1)$$

$$\Phi_2^{mn}(\omega)=G_2^0(\omega-m\omega_1-n\omega_2)[\delta_{2v}\delta_{mv}\delta_{nv}n!m!+M_1^*\Phi_1^{m+1,n}(\omega)+mM_1\Phi_1^{m-1,n}(\omega)+M_2^*\Phi_3^{m,n+1}(\omega)+nM_2\Phi_3^{m,n-1}(\omega)+N_2^*\Phi_1^{m,n+1}(\omega)+nN_2\Phi_1^{m,n-1}(\omega)+N_1^*\Phi_3^{m+1,n}(\omega)+mN_1\Phi_3^{m-1,n}(\omega)]\ , \quad (A2)$$

$$\Phi_3^{mn}(\omega)=G_3^0(\omega-m\omega_1-n\omega_2)[\delta_{3v}\delta_{mv}\delta_{nv}n!m!+M_2^*\Phi_2^{m,n+1}(\omega)+nM_2\Phi_2^{m,n-1}(\omega)+N_1^*\Phi_2^{m+1,n}(\omega)+mN_1\Phi_2^{m-1,n}(\omega)]\ , \quad (A3)$$

where $G_i^0(\omega)$ with $i=1,2,3$ are bare Green functions of electronic states. The transition amplitudes are defined by $\Phi_i^{mn}(\omega)=\langle i|a_1^m a_2^n(\omega-H)^{-1}|v\rangle$, where $|v\rangle$ is the initial state of the system and $i=1,2,3$ are allowed final states.

Let me define the column vectors $\Phi(n)=(\Phi_2^{pn}(\omega))$ of dimension $m\to M\to\infty$. The decoupled matrix equation is

$$\vec{\vec{A}}(n)\Phi_2(n)-\vec{\vec{B}}(n)\Phi_2(n+2)-\vec{\vec{C}}(n)\Phi_2(n+1)-\vec{\vec{D}}(n)\Phi_2(n-1)-\vec{\vec{E}}(n)\Phi_2(n-2)=\mathbf{F}(n)\ , \quad (A4)$$

where the components of the matrices $\vec{\vec{A}}$, $\vec{\vec{B}}$, $\vec{\vec{C}}$, $\vec{\vec{D}}$, and $\vec{\vec{E}}$ are

$$\begin{aligned}\overleftrightarrow{A}(n)=A_{pm}(n)=\delta_{pm}\{1-G_2^0(\omega-m\omega_1-n\omega_2)\\ \times[|M_1|^2(m+1)G_1^0(\omega-(m+1)\omega_1-n\omega_2)+|M_1|^2mG_1^0(\omega-(m-1)\omega_1-n\omega_2)\\ +|M_2|^2(n+1)G_3^0(\omega-m\omega_1-(n+1)\omega_2)+|M_2|^2nG_3^0(\omega-m\omega_1-(n-1)\omega_2)\\ +|N_2|^2(n+1)G_1^0(\omega-m\omega_1-(n+1)\omega_2)+|N_2|^2nG_1^0(\omega-m\omega_1-(n-1)\omega_2)\\ +|N_1|^2(m+1)G_3^0(\omega-(m+1)\omega_1-n\omega_2)+|N_1|^2mG_3^0(\omega-(m-1)\omega_1-n\omega_2)]\}\\ -\delta_{p,m+2}[M_1^{*2}G_1^0(\omega-(m+1)\omega_1-n\omega_2)+N_1^{*2}G_3^0(\omega-(m+1)\omega_1-n\omega_2)]\\ -(m-1)\delta_{p,m-2}[M_1^2mG_1^0(\omega-(m-1)\omega_1-n\omega_2)+N_1^2mG_3^0(\omega-(m-1)\omega_1-n\omega_2)]\ ,\end{aligned}\tag{A5}$$

$$\overleftrightarrow{B}(n)=B_{pm}(n)=\{\delta_{pn}[M_2^{*2}G_3^0(\omega-m\omega_1-(n+1)\omega_2)+N_2^{*2}G_1^0(\omega-m\omega_1-(n+1)\omega_2)]\}G_2^0(\omega-m\omega_1-n\omega_2)\ ,\tag{A6}$$

$$\begin{aligned}\overleftrightarrow{C}(n)=C_{pm}(n)=\{\ \delta_{p,m+1}[N_2^*M_1^*G_1^0(\omega-(m+1)\omega_1-n\omega_2)+N_1^*M_2^*G_3^0(\omega-m\omega_1-(n+1)\omega_2)\\ +M_1^*N_2^*G_1^0(\omega-m\omega_1-(n+1)\omega_2)+M_2^*N_1^*G_3^0(\omega-(m+1)\omega_1-n\omega_2)]\\ +m\delta_{p,m-1}[\ N_2^*M_1G_1^0(\omega-(m-1)\omega_1-n\omega_2)+N_1M_2^*G_3^0(\omega-m\omega_1-(n+1)\omega_2)\\ +M_1N_2^*G_1^0(\omega-m\omega_1-(n+1)\omega_2)+M_2^*N_1G_3^0(\omega-(m-1)\omega_1-m\omega_2)]\}\\ \times G_2^0(\omega-m\omega_1-n\omega_2)\ ,\end{aligned}\tag{A7}$$

$$\begin{aligned}\overleftrightarrow{D}(n)=D_{pm}(n)=\{\ n\delta_{p,m+1}[N_2M_1^*G_1^0(\omega-(m+1)\omega_1-n\omega_2)+N_1^*M_2G_3^0(\omega-m\omega_1-(n-1)\omega_2)\\ +N_2M_1^*G_1^0(\omega-m\omega_1-(n-1)\omega_2)+M_2N_1^*G_3^0(\omega-(m+1)\omega_1-n\omega_2)]\\ +nm\delta_{p,m-1}[\ N_2M_1G_1^0(\omega-(m-1)\omega_1-n\omega_2)+N_1M_2G_3^0(\omega-m\omega_1-(n-1)\omega_2)\\ +M_1N_2G_1^0(\omega-m\omega_1-(n-1)\omega_2)\\ +M_2N_1G_3^0(\omega-(m-1)\omega_1-n\omega_2)]\}G_2^0(\omega-m\omega_1-n\omega_2)\ ,\end{aligned}\tag{A8}$$

$$\begin{aligned}\overleftrightarrow{E}(n)=E_{pm}(n)=\{n(n-1)\delta_{p,m}[\ M_2^2G_3^0(\omega-m\omega_1-(n-1)\omega_2)\\ +N_2^2G_1^0(\omega-m\omega_1-(n-1)\omega_2)]\}G_2^0(\omega-m\omega_1-n\omega_2)\ ,\end{aligned}\tag{A9}$$

and the known term of Eq. (A4) is

$$\begin{aligned}\overleftrightarrow{F}(n)=G_2^0(\omega-m\omega_1-n\omega_2)[\ \delta_{2\nu}\delta_{m\nu}\delta_{n\nu}n!m!+M_1^*n!(m+1)!\delta_{1\nu}\delta_{m+1,\nu}\delta_{n\nu}G_1^0(\omega-(m+1)\omega_1-n\omega_2)\\ +M_1n!m!\delta_{1\nu}\delta_{m-1,\nu}\delta_{n\nu}G_1^0(\omega-(m-1)\omega_1-n\omega_2)\\ +M_2^*(n+1)!m!\delta_{3\nu}\delta_{m\nu}\delta_{n+1,\nu}G_3^0(\omega-m\omega_1-(n+1)\omega_2)\\ +M_2n!m!\delta_{3\nu}\delta_{m\nu}\delta_{n-1,\nu}G_3^0(\omega-m\omega_1-(n-1)\omega_2)\\ +N_2^*(n+1)!m!\delta_{1\nu}\delta_{m\nu}\delta_{n+1,\nu}G_1^0(\omega-m\omega_1-(n+1)\omega_2)\\ +N_2n!m!\delta_{1\nu}\delta_{m\nu}\delta_{n-1,\nu}G_1^0(\omega-m\omega_1-(n-1)\omega_2)\\ +N_1^*n!(m+1)!\delta_{3\nu}\delta_{m+1,\nu}\delta_{n\nu}G_3^0(\omega-(m+1)\omega_1-n\omega_2)\\ +N_1n!m!\delta_{3\nu}\delta_{m-1,\nu}\delta_{n\nu}G_3^0(\omega-(m-1)\omega_1-n\omega_2)]\ .\end{aligned}\tag{A10}$$

Finally I can reduce the recursive equation (A4) in a simpler form

$$\Phi(n)=\overleftrightarrow{\alpha}(n)\Phi(n-1)+\overleftrightarrow{\beta}(n)\Phi(n-2)\ ,\tag{A11}$$

where the $\overleftrightarrow{\alpha}$ and $\overleftrightarrow{\beta}$ matrices are

$$\begin{aligned}\overleftrightarrow{\alpha}(n)=(\overleftrightarrow{A}(n)-\{[\overleftrightarrow{B}(n)\overleftrightarrow{\alpha}(n+2)+\overleftrightarrow{C}(n)]\overleftrightarrow{\alpha}(n+1)+\overleftrightarrow{B}(n)\overleftrightarrow{\beta}(n+2)\})^{-1}\\ \times\{[\overleftrightarrow{B}(n)\overleftrightarrow{\alpha}(n+2)+\overleftrightarrow{C}(n)]\overleftrightarrow{\beta}(n+1)+\overleftrightarrow{D}(n)\}\ ,\end{aligned}\tag{A12}$$

$$\overleftrightarrow{\beta}(n)=(\overleftrightarrow{A}(n)-\{[\overleftrightarrow{B}(n)\overleftrightarrow{\alpha}(n+2)+\overleftrightarrow{C}(n)]\overleftrightarrow{\alpha}(n+1)+\overleftrightarrow{B}(n)\overleftrightarrow{\beta}(n+1)\})^{-1}\overleftrightarrow{E}(n)\ .\tag{A13}$$

Now the model Hamiltonian of Eqs. (39)–(41) is completely solved if we impose the boundary conditions $n \to N \to \infty$,

$$\vec{\alpha}(N) = \vec{\mathrm{A}}(N)^{-1}\vec{\mathrm{D}}(N) , \tag{A14}$$

$$\vec{\beta}(N) = \vec{\mathrm{A}}(N)^{-1}\vec{\mathrm{E}}(N) . \tag{A15}$$

Equations (A11)–(A15) with $N_i = 0$ and $i = 1,2$ recover the solution obtained in Sec. IV D of the text as a particular case. Finally, I expect new and interesting phenomena when photons ω_1 and ω_2 are in resonance with the same electronic transition.

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