

Field-dependent relaxation effects in a three-level system driven by a strong coherent field

Olga Kocharovskaya,^{1,2} Y. V. Radeonychev,² Paul Mandel,³ and M. O. Scully¹

¹*Department of Physics, Texas A&M University, College Station, Texas 77843
and Max-Planck Institut für Quantenoptik, D-85748 Garching, Germany*

²*Institute of Applied Physics, Russian Academy of Science, 46 Ulyanov str., 603600 Nizhny Novgorod, Russia*

³*Optique Nonlinéaire Théorique, Université Libre de Bruxelles, Campus Plaine, Code Postal 231, 1050 Bruxelles, Belgium*

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We describe the physical effects emerging in three-level atoms as a result of a drastic modification of the relaxation processes under the action of a strong coherent field when one of two dynamic Stark levels crosses a nearby atomic state. [S1050-2947(99)05309-3]

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I. INTRODUCTION

Recently much attention has been paid to the analysis of coherent effects in three-level systems driven by a strong resonant field and interacting with a reservoir. Lasing without inversion, electromagnetically induced transparency, enhancement of the refractive index, and other effects have been widely discussed [1]. The analysis of these effects is traditionally based on the master equations where the interaction of atoms with the reservoir is described by means of relaxation constants which are assumed to be independent of the driving field. However, it is well known that a strong coherent driving can modify the relaxation processes (see Refs. [2–9], and references therein). This modification was studied both theoretically and experimentally mainly for two-level systems, and was supposed to be connected with a violation of the Markov approximation [2–5].

Recently it was emphasized that field-dependent relaxation effects appear even within the Markov approximation made for the dressed atom [6–9]. For two-level atoms in free space, these effects are typically weak, since they are defined by a small parameter which is the Rabi frequency divided by the frequency of the resonant driven transition [9]. However, they may be strongly enhanced by placing atoms inside a frequency-selective cavity, whose density of modes is sharply changed on the scale of the Rabi frequency [2–9]. This was demonstrated experimentally [3,4].

In three-level atoms the drastic modification of the decay rates by the driving field may occur even in free space [7,10], as soon as one dynamic Stark sublevel crosses a neighboring unperturbed atomic energy state (Fig. 1). Very recently we predicted the phenomenon of coherent population trapping of atoms into the lower dynamic Stark sublevel in a scheme with ground state splitting [Fig. 1(a)] in the case of very strong and exactly resonant driving [7], and studied some other effects caused by level crossing for that scheme [10]. The origin of these phenomena is the appearance of spontaneous decay from the former ground state to the lower dynamic Stark sublevel, resulting from the crossing between these two states.

In this paper we systematically study field-dependent relaxation effects in driven three-level atoms at arbitrary intensity and detuning of the driving field for all possible level configurations [Figs. 1(a)–1(d)]. We base our treatment on

the set of generalized Maxwell-Bloch equations which were derived recently [6]. We show that the physical origin of the field-dependent relaxation is a dependence of the relaxation rates on the dressed atomic frequencies. Both the sign and magnitude of the relaxation rates are drastically modified in the case of level crossing.

We find an analytical solution of the generalized master equations at an arbitrary intensity and detuning of the driving field. On this basis we predict a strong breaking of the symmetry between the dressed-level populations with respect to the detuning of the driving field from the resonant transition caused by field-dependent relaxation, and leading under certain conditions to a population trapping into one or another dynamic Stark level. We emphasize the possibility of a large population inversion at the driven transition in the case of a negative detuning, and show that both absorption and disper-

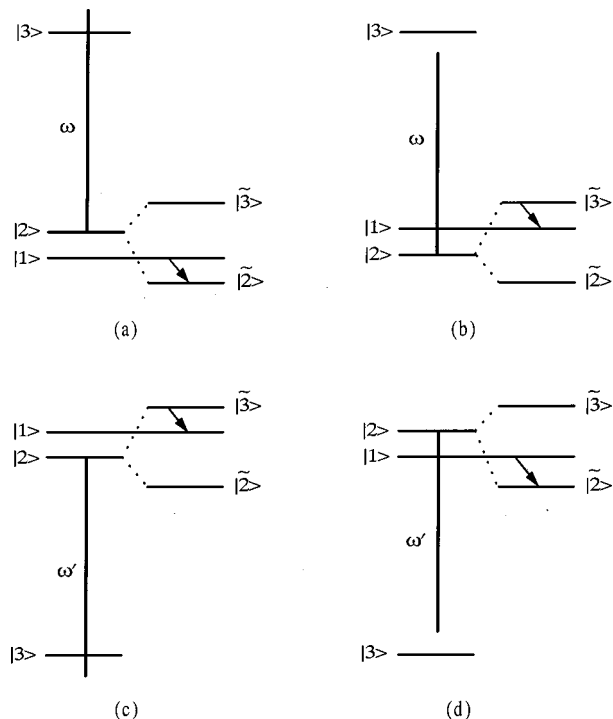


FIG. 1. Possible schemes of a three-level atomic system strongly driven by the monochromatic field at a $|2\rangle$ - $|3\rangle$ atomic transition $\omega' = -\omega$.

sion profiles are drastically modified if one of the dynamic Stark levels crosses a nearby atomic state. We also demonstrate a qualitative modification of the Mollow and Autler-Townes spectra in the case of level crossing, and on this basis predict a mechanism for the realization of a high refractive index and lasing without inversion.

II. GENERALIZED MASTER EQUATIONS FOR A MULTILEVEL SYSTEM

A. Bare-state description

We base our analysis on a set of generalized master equations for the density matrix of a multilevel system driven by a strong coherent field,

$$\mathbf{E} = \frac{1}{2} \sum_m (\vec{\mathcal{E}}_m e^{-i\omega_m t} + \text{c.c.}),$$

when at least one level in each pair of levels is either coupled with one component of the field or is not coupled with the field at all. These equations were derived in Refs. [6] and [10] in Born-Markov approximations for the dressed atom, and have a form which is similar to that of traditional master equations with the field-independent relaxation

$$\frac{d}{dt} \rho_{m'm} + i\omega_{m'm} \rho_{m'm} + \frac{i}{\hbar} [V(t), \rho]_{m'm} = \sum_{n'n} R_{m'mn'n} \rho_{n'n}, \quad (1)$$

where

$$V(t) = -\frac{1}{2} \sum_m (\vec{\mu}_{m'm} \vec{\mathcal{E}}_m e^{-i\omega_m t} |m'\rangle \langle m| + \text{H.c.})$$

is the Hamiltonian describing the interaction of the atoms with a coherent field in the dipole and rotating-wave approximations. $\vec{\mu}_{m'm} = \langle m' | \vec{\mu} | m \rangle$ is the matrix element of the dipole moment of the corresponding atomic transition,

$$\rho_{m'm} = \langle m' | \rho | m \rangle,$$

$$H_0 |m\rangle = E_m |m\rangle,$$

$$\omega_{m'm} = (E_{m'} - E_m) / \hbar,$$

and H_0 is the Hamiltonian of the unperturbed atomic system. However, the matrix elements of the relaxation supermatrix $R_{m'mn'n}$ are functions of the driving field. They are expressed via the well-known relaxation constants $\Gamma_{m'mn'n}^{(0)}$ of the unperturbed atomic system:

$$R_{m'mn'n} = \Gamma_{nm m'n'} + \Gamma_{n'm'mn}^* - \sum_k (\delta_{mn} \Gamma_{m'kkn'} + \delta_{m'n'} \Gamma_{mkkn}^*), \quad (2)$$

$$\Gamma_{nm m'n'} = \sum_{kk' ll'} (\zeta_{m'}^k)^* \zeta_{k'}^k (\zeta_{l'}^l)^* \zeta_{l'}^l \times \Gamma_{nm k'l'}^{(0)} (\tilde{\omega}_{kl} - \epsilon_{k'} \omega_{k'} + \epsilon_{l'} \omega_{l'}), \quad (3)$$

$$\Gamma_{nm m'n'}^{(0)}(\omega) = \frac{1}{\hbar^2} \int_0^\infty d\tau \exp(-i\omega\tau) \times \text{Tr} \left\{ W_{nm} \exp\left(-\frac{i}{\hbar} H_r \tau\right) W_{m'n'} \times \exp\left(\frac{i}{\hbar} H_r \tau\right) \rho_r(0) \right\}. \quad (4)$$

Here $\zeta_n^m = \langle \tilde{m} | n \rangle$ are time-dependent elements of the unitary matrix connecting the bare states $|n\rangle$ and rotating semiclassical dressed states $|\tilde{m}\rangle = U|\bar{m}\rangle$ where time-independent dressed states $|\bar{m}\rangle$ diagonalize a time-independent Hamiltonian:

$$\bar{H} = U^{-1} H U - i\hbar U^{-1} \frac{dU}{dt},$$

$$H = H_0 + V(t),$$

$$U = \sum_k \exp(i\epsilon_k \omega_k t) |k\rangle \langle k|,$$

$$\bar{H} |\bar{m}\rangle = \bar{E}_m |\bar{m}\rangle,$$

$$\tilde{\omega}_{nm} = (\bar{E}_n - \bar{E}_m) / \hbar.$$

$\epsilon_k = 1$ if level k interacts with only one component of the field, and it is the lower level in a pair of levels coupled to this field component; $\epsilon_k = -1$ if level k interacts with only one component of the field, and is the upper level in a pair of levels coupled to this component; $\epsilon_k = 0$ otherwise. The trace in Eq. (4) is taken over the reservoir variables described by the unperturbed density matrix $\rho_r(0)$ in the Born approximation, H_r is the reservoir Hamiltonian, and W_{nm} is the matrix element of the atom-reservoir interaction Hamiltonian.

B. Dressed-state description

In the semiclassical dressed-state basis which is often helpful for clarifying the physical picture, the set of equations (1) takes the forms:

$$\frac{d}{dt} \tilde{\rho}_{m'm} + i\tilde{\omega}_{m'm} \tilde{\rho}_{m'm} = \sum_{n'n} \tilde{R}_{m'mn'n} \tilde{\rho}_{n'n},$$

$$\tilde{\rho}_{m'm} = \langle \tilde{m}' | \rho | \tilde{m} \rangle, \quad (5)$$

$$\tilde{R}_{m'mn'n} = \tilde{\Gamma}_{nm m'n'} + \tilde{\Gamma}_{n'm'mn}^* - \sum_k (\delta_{mn} \tilde{\Gamma}_{m'kkn'} + \delta_{m'n'} \tilde{\Gamma}_{mkkn}^*), \quad (6)$$

$$\tilde{\Gamma}_{nm m'n'} = \sum_{kk' ll'} \zeta_k^n (\zeta_{k'}^m)^* \zeta_l^{m'} (\zeta_{l'}^{n'})^* \times \Gamma_{nm k'l'}^{(0)} (\tilde{\omega}_{m'n'} - \epsilon_l \omega_l + \epsilon_{l'} \omega_{l'}). \quad (7)$$

In this picture we get rid of the reversible Hamiltonian terms and deal only with relaxation processes between dressed levels.

The generalized master equations (1) and (5) can be used for analyzing a wide class of physical problems. We shall use them below to study the behavior of the three-level system coupled to a field reservoir and driven by a strong monochromatic field at one of the atomic transitions.

III. GENERALIZED MASTER EQUATIONS FOR A THREE-LEVEL SYSTEM

A. Bare-state description

For a three-level system driven by the monochromatic field $\mathbf{E} = \frac{1}{2}(\vec{\mathcal{E}}e^{-i\omega t} + \text{c.c.})$ at the transition $|3\rangle - |2\rangle$, equations (1) in the general case take the forms

$$\begin{aligned}
 \dot{\rho}_{11} &= \sum_n R_{11nn} \rho_{nn} + 2 \operatorname{Re}(R_{1121} \sigma_{21}) \\
 &\quad + 2 \operatorname{Re}(R_{1131} \sigma_{31}) + 2 \operatorname{Re}(R_{1132} \sigma_{32}), \\
 \dot{\rho}_{22} + 2 \operatorname{Im}(\beta^* \sigma_{32}) &= \sum_n R_{22nn} \rho_{nn} + 2 \operatorname{Re}(R_{2221} \sigma_{21}) \\
 &\quad + 2 \operatorname{Re}(R_{2231} \sigma_{31}) + 2 \operatorname{Re}(R_{2232} \sigma_{32}), \\
 \dot{\rho}_{33} - 2 \operatorname{Im}(\beta^* \sigma_{32}) &= \sum_n R_{33nn} \rho_{nn} + 2 \operatorname{Re}(R_{3321} \sigma_{21}) \\
 &\quad + 2 \operatorname{Re}(R_{3331} \sigma_{31}) + 2 \operatorname{Re}(R_{3332} \sigma_{32}), \\
 \dot{\sigma}_{32} + i \delta \sigma_{32} - i \beta n_{23} &= \sum_n R_{32nn} \rho_{nn} + \sum_{n \neq m} R_{32nm} \sigma_{nm}, \\
 \dot{\sigma}_{31} + i(\omega_{21} + \delta) \sigma_{31} - i \beta \sigma_{21} &= \sum_n R_{31nn} \rho_{nn} \\
 &\quad + \sum_{n \neq m} R_{31nm} \sigma_{nm}, \\
 \dot{\sigma}_{21} + i \omega_{21} \sigma_{21} - i \beta^* \sigma_{31} &= \sum_n R_{21nn} \rho_{nn} + \sum_{n \neq m} R_{21nm} \sigma_{nm},
 \end{aligned} \tag{8}$$

where

$$\begin{aligned}
 \rho_{32} &= \sigma_{32} e^{-i\omega t}, \\
 \rho_{31} &= \sigma_{31} e^{-i\omega t}, \quad \rho_{21} = \sigma_{21}, \\
 n_{23} &= \rho_{22} - \rho_{33}, \quad \delta = \omega_{32} - \omega, \\
 \beta &= \frac{\vec{\mu}_{32} \vec{\mathcal{E}}}{2\hbar} = |\beta| e^{-i\varphi}.
 \end{aligned} \tag{9}$$

We assume here that even in the case of sufficiently strong driving ($|\beta| \gg \omega_{21}$), the monochromatic field interacts with only one transition due, for example, to specific selection rules or polarization properties of the field. For the schemes shown in Fig. 1, both the Rabi frequency β of the driving field and its detuning δ may be greater than the frequency ω_{21} . One can calculate elements of the relaxation matrix $R_{mnm'n'}$ (see Appendix A) according to Eqs. (2)–(4), taking into account such possibility. Calculating $R_{m'mn'n}$, we neglected all the terms above order zero of $|R_{m'mn'n}|/\omega$ (the secular approximation with respect to the frequency ω of the driving field). We consider the interaction of the atomic system with a field reservoir. Then the unperturbed relaxation constants $\Gamma_{m'mn'n}^{(0)}$ in Eq. (4) are well known [11]. We keep only the real parts of $\Gamma_{nmk'l'}^{(0)}$, neglecting by the frequency shifts of the levels due to the coupling to the reservoir.

In principle, the radiation shifts of atomic levels in the case of a strongly driven system are the radiation shifts of dressed levels in the same manner as radiation widths of dressed levels in case of strong driving are the radiation widths of the dressed levels (see Sec. IV). Hence they are field dependent too. However, in the case of well-resolved dressed states (when the Rabi frequency of the driving field is greater than the radiation widths of the dressed levels), one can neglect by radiation shifts of the dressed levels in the same manner as one can neglect the radiation shifts of the nondegenerate (well-resolved) bare-energy states. Indeed, it can be seen directly from Eqs. (25) and (50) that in this case one can neglect imaginary parts of field-modified relaxation rates.

Then the ‘‘perturbed’’ relaxation terms $R_{m'mn'n}$ are expressed via the generalized transition rates

$$w_{mkl n}(\omega) = \operatorname{Re}(\Gamma_{mkl n}^{(0)}(\omega)) = \frac{2}{3\hbar c_0^3} \vec{\mu}_{mk} \vec{\mu}_{ln} \times \begin{cases} \omega^3 \eta(\omega) N(\omega), & \omega > 0 \\ (-\omega^3) \eta(-\omega) (N(-\omega) + 1), & \omega < 0 \end{cases} \tag{10}$$

and

$$w_{mk}(\omega) = 2w_{mkkm}(\omega), \tag{11}$$

where $N(\omega)$ is a mean number of reservoir photons, $\eta(\omega)$ is a dimensionless parameter characterizing the reservoir mode density [in vacuum $\eta(\omega) = 1$], $\vec{\mu}_{mk}$ is the dipole moment at the $|m\rangle - |k\rangle$ transition, and c_0 is the light velocity. The choice of the unitary transformation

$$U = |1\rangle\langle 1| + |2\rangle\langle 2| + e^{-i\omega t} |3\rangle\langle 3| \tag{12}$$

defines, in Eq. (3), the coefficients ζ_n^m connecting the bare and semiclassical dressed states

$$|\tilde{1}\rangle = |1\rangle,$$

$$|\tilde{2}\rangle = c|2\rangle + s e^{-i(\varphi + \omega t)} |3\rangle,$$

$$\begin{aligned}
|\tilde{3}\rangle &= -s e^{i\varphi}|2\rangle + e^{-i\omega t}c|3\rangle, \\
s &= \frac{|\beta|}{\sqrt{\theta^2 + |\beta|^2}}, \\
c &= \frac{\theta}{\sqrt{\theta^2 + |\beta|^2}}, \\
\theta &= (\delta + \Omega)/2, \\
\Omega &= \sqrt{\delta^2 + 4|\beta|^2},
\end{aligned} \tag{13}$$

where the dressed level $|\tilde{n}\rangle$ has energies

$$\begin{aligned}
\tilde{E}_1 &= E_1, \\
\tilde{E}_2 &= E_2 + \hbar(\delta - \Omega)/2, \\
\tilde{E}_3 &= E_2 + \hbar(\delta + \Omega)/2.
\end{aligned} \tag{14}$$

In general the relaxation matrix given in Appendix A has quite a complicated form.

The assumption that the driving field is coupled only to the $|3\rangle$ - $|2\rangle$ transition can be valid either when

$$|\vec{\mu}_{31}| = 0 \quad \text{or} \quad \vec{\mu}_{31} \perp \vec{\mu}_{32}. \tag{15}$$

In both cases, as follows from Eqs. (10), one can neglect by those cross-relaxation terms which form the (A2) of Appendix A. Then the set of equations (8) is divided into two independent blocks. The first block consists of three equations for the populations plus one equation for the coherence σ_{32} of the driven transition. The second block consists of a set of two homogeneous equations for the coherences σ_{31} and σ_{21} . Hence the driving field does not excite these coherences ($\sigma_{31} = 0$ and $\sigma_{21} = 0$), and the evolution of the atomic system is described by the first block of equations (8), which takes the following forms:

$$\begin{aligned}
\dot{\rho}_{11} &= R_{1111}\rho_{11} + R_{1122}\rho_{22} + R_{1133}\rho_{33} + 2 \operatorname{Re}(R_{1123}\sigma_{23}), \\
\dot{\rho}_{22} + 2 \operatorname{Im}(\beta^* \sigma_{32}) &= R_{2211}\rho_{11} + R_{2222}\rho_{22} + R_{2233}\rho_{33} \\
&\quad + 2 \operatorname{Re}(R_{2223}\sigma_{23}), \\
\dot{\rho}_{33} - 2 \operatorname{Im}(\beta^* \sigma_{32}) &= R_{3311}\rho_{11} + R_{3322}\rho_{22} + R_{3333}\rho_{33} \\
&\quad + 2 \operatorname{Re}(R_{3323}\sigma_{23}), \\
\dot{\sigma}_{32} + i\delta\sigma_{32} - i\beta n_{23} &= R_{3232}\sigma_{32} + R_{3223}\sigma_{23} + R_{3211}\rho_{11} \\
&\quad + R_{3222}\rho_{22} + R_{3233}\rho_{33}.
\end{aligned} \tag{16}$$

The rotating-wave approximation, which in our case implies

$$\Omega \ll |\omega_{31}|, |\omega_{32}|, \tag{17}$$

allows one to simplify the Eqs. (16):

$$\begin{aligned}
\dot{\rho}_{11} &= -(a_{12} + w_{13})\rho_{11} + a_{21}\rho_{22} + w_{31}\rho_{33} \\
&\quad - (a_{31} + a_1)\operatorname{Re}(\sigma_{32}e^{i\varphi}),
\end{aligned}$$

$$\begin{aligned}
\dot{\rho}_{22} + 2 \operatorname{Im}(\beta^* \sigma_{32}) &= a_{12}\rho_{11} - (a_{21} + w_{23})\rho_{22} + w_{32}\rho_{33} \\
&\quad + (a_1 - \tilde{a}_{32})\operatorname{Re}(\sigma_{32}e^{i\varphi}), \\
\dot{\rho}_{33} - 2 \operatorname{Im}(\beta^* \sigma_{32}) &= w_{13}\rho_{11} + w_{23}\rho_{22} - (w_{31} + w_{32})\rho_{33} \\
&\quad + (a_{31} + \tilde{a}_{32})\operatorname{Re}(\sigma_{32}e^{i\varphi}), \\
\dot{\sigma}_{32} + i\delta\sigma_{32} - i\beta n_{23} &= -\Gamma_{32}\sigma_{32} + e^{-i\varphi}R/2,
\end{aligned} \tag{18}$$

where

$$\begin{aligned}
R &= (a_2 - a_{13})\rho_{11} + (a_{31} + a_{32}^0)\rho_{22} + (a_1 + a_{32}^0)\rho_{33}, \\
\Gamma_{32} &= (a_{21} + w_{31} + w_{32} + w_{23})/2, \\
a_{31} &= 3w_{31}|\beta|/\omega_{31}, \\
a_{13} &= 3w_{13}|\beta|/\omega_{31}, \quad \tilde{a}_{31} = a_{31} + a_{13}, \\
a_{31}^0 &= a_{31} - a_{13}, \\
a_{32} &= 3w_{32}|\beta|/\omega_{32}, \\
a_{23} &= 3w_{23}|\beta|/\omega_{32}, \\
\tilde{a}_{32} &= a_{32} + a_{23}, \quad a_{32}^0 = a_{32} - a_{23}, \\
a_{21} &= s^2 w_{21}(\tilde{\omega}_{13}) + c^2 w_{21}(\tilde{\omega}_{12}), \\
a_{12} &= s^2 w_{12}(\tilde{\omega}_{31}) + c^2 w_{12}(\tilde{\omega}_{21}), \\
a_1 &= s c (w_{21}(\tilde{\omega}_{13}) - w_{21}(\tilde{\omega}_{12})), \\
a_2 &= s c (w_{12}(\tilde{\omega}_{21}) - w_{12}(\tilde{\omega}_{31})).
\end{aligned} \tag{19}$$

Here w_{mk} is the usual relaxation rate at the transition $|m\rangle \rightarrow |k\rangle$:

$$w_{mk} = \begin{cases} A_{mk}(N(\omega_{mk}) + 1), & \omega_{mk} > 0, \\ A_{km}N(\omega_{km}), & \omega_{mk} < 0, \end{cases} \tag{20}$$

where $A_{mk} = (4/3\hbar c_0^3)|\mu_{mk}|^2 w_{mk}^3 \eta(\omega_{mk})$ is the Einstein coefficient of the spontaneous relaxation at the corresponding atomic transition $|m\rangle \rightarrow |k\rangle$.

We will show below that, contrary to the traditional phenomenological master equations, the structure of the relaxation coefficients in Eqs. (18) reflects the fact that the relaxation of strongly driven atoms occurs between dynamic Stark (dressed) levels which result from the splitting of the atomic energy states. In the limit of a weak field, where this splitting is negligible, the generalized equations are reduced to traditional equations.

The set of equations (18) is valid for all four configurations displayed in Figs. 1(a)–1(d). However, the explicit form of the relaxation coefficients depends essentially on the sign of the transition frequencies. We have kept only the

terms of the first order with respect to the small parameter $\Omega/|\omega_{32}|$. However, we did not impose any constraint on the ratio $\Omega/|\omega_{21}|$. This allows for a change of sign of either $\tilde{\omega}_{31}$ or $\tilde{\omega}_{21}$ depending on the level configuration. This change of sign of the dressed frequency corresponds to the crossing between one dynamic Stark level and an unperturbed atomic state which we always label $|1\rangle$ in Figs. 1. In the bare-state basis this looks like a change of the direction of the spontaneous emission between levels $|1\rangle$ and $|2\rangle$.

Let us illustrate this property for the scheme of Fig. 1(a). The relaxation coefficients are simplified in case of pure spontaneous relaxation since then $N(\omega)=0$. According to Eqs. (19) and (20), we obtain

$$\begin{aligned} w_{31} &= A_{31}, \\ w_{32} &= A_{32}, \\ a_{31} &= 3A_{31}|\beta|/\omega_{31}, \quad a_{32} = 3A_{32}|\beta|/\omega_{32}, \quad (21) \\ \tilde{a}_{31} &= a_{31}^0 = a_{31}, \quad \tilde{a}_{32} = a_{32}^0 = a_{32}, \\ a_{13} &= a_{23} = w_{13} = w_{23} = 0. \end{aligned}$$

The relaxation rates at the low-frequency transition $|1\rangle$ - $|2\rangle$ are defined by the relative position of the low Stark level and an uncoupled state $|1\rangle$. When level $|\tilde{2}\rangle$ is situated above state $|1\rangle$, according to Eqs. (19) we have

$$\begin{aligned} a_{21} &= g_1 A_{21}, \\ a_1 &= g_2 A_{21}|\beta|/\omega_{21}, \\ a_{12} &= a_2 = 0, \quad (22) \\ g_1 &= 1 + (3 + \delta/\omega_{21})|\beta|^2/\omega_{21}^2, \\ g_2 &= 3(1 + \delta/\omega_{21}) + (\delta^2 + |\beta|^2)/\omega_{21}^2. \end{aligned}$$

But as soon as level $|\tilde{2}\rangle$ follows below level $|1\rangle$, we obtain

$$\begin{aligned} a_{21} &= s^2 A_{21}(\tilde{\omega}_{31}/\omega_{21})^3, \\ a_{12} &= c^2 A_{21}(\tilde{\omega}_{12}/\omega_{21})^3, \\ a_1 &= sc A_{21}(\tilde{\omega}_{31}/\omega_{21})^3, \\ a_2 &= sc A_{21}(\tilde{\omega}_{12}/\omega_{21})^3. \end{aligned} \quad (23)$$

The term a_{12} appearing when level $|\tilde{2}\rangle$ crosses level $|1\rangle$ corresponds formally to the spontaneous decay from state $|1\rangle$ to state $|2\rangle$. The appearance of this decay looks rather artificial because a bare-state basis is not appropriate in the case of strong driving. The structure of the master equations in the bare-state basis is quite complicated due to the presence of cross-relaxation terms, i.e., off-diagonal elements of the density matrix in the equations for diagonal elements, and vice versa.

B. Dressed-state description

The dressed-state picture is especially fruitful when the secular approximation with respect to the dressed frequencies $\tilde{\omega}_{m'm}$ can be used. This implies in Eq. (5) that

$$|\tilde{R}_{m'mn'n}| \ll |\tilde{\omega}_{m'm}|. \quad (24)$$

We use the secular approximation (24) in the dressed-state basis, neglecting terms which are smaller than the first order of $|\tilde{R}_{m'mn'n}|/|\tilde{\omega}_{m'm}|$. In this case the equations for the dressed populations do not depend on the equation for the dressed coherence $\tilde{\rho}_{32}$:

$$\begin{aligned} \frac{d}{dt}\tilde{\rho}_{11} &= \tilde{R}_{1111}\tilde{\rho}_{11} + \tilde{R}_{1122}\tilde{\rho}_{22} + \tilde{R}_{1133}\tilde{\rho}_{33}, \\ \frac{d}{dt}\tilde{\rho}_{22} &= \tilde{R}_{2211}\tilde{\rho}_{11} + \tilde{R}_{2222}\tilde{\rho}_{22} + \tilde{R}_{2233}\tilde{\rho}_{33}, \\ \frac{d}{dt}\tilde{\rho}_{33} &= \tilde{R}_{3311}\tilde{\rho}_{11} + \tilde{R}_{3322}\tilde{\rho}_{22} + \tilde{R}_{3333}\tilde{\rho}_{33}, \end{aligned} \quad (25)$$

$$\frac{d}{dt}\tilde{\rho}_{32} + i\tilde{\omega}_{32}\tilde{\rho}_{32} = \tilde{R}_{3232}\tilde{\rho}_{32} + \tilde{R}_{3211}\tilde{\rho}_{11} + \tilde{R}_{3222}\tilde{\rho}_{22} + \tilde{R}_{3233}\tilde{\rho}_{33},$$

where

$$\begin{aligned} \tilde{R}_{1122} &= c^2 w_{21}(\tilde{\omega}_{12}) + s^2 w_{31}(\tilde{\omega}_{12} - \omega), \\ \tilde{R}_{1133} &= s^2 w_{21}(\tilde{\omega}_{13}) + c^2 w_{31}(\tilde{\omega}_{13} - \omega), \\ \tilde{R}_{2211} &= c^2 w_{12}(\tilde{\omega}_{21}) + s^2 w_{13}(\tilde{\omega}_{21} + \omega), \\ \tilde{R}_{2233} &= s^4 w_{23}(\tilde{\omega}_{23} + \omega) + c^4 w_{32}(\tilde{\omega}_{23} - \omega), \\ \tilde{R}_{3311} &= s^2 w_{12}(\tilde{\omega}_{31}) + c^2 w_{13}(\tilde{\omega}_{31} + \omega), \\ \tilde{R}_{3322} &= s^4 w_{32}(\tilde{\omega}_{32} - \omega) + c^4 w_{23}(\tilde{\omega}_{32} + \omega), \\ \tilde{R}_{iiii} &= - \sum_{j \neq i} \tilde{R}_{jjii}, \quad (26) \\ \tilde{R}_{3232} &= -\frac{1}{2}(\tilde{R}_{1122} + \tilde{R}_{1133} + \tilde{R}_{2233} + \tilde{R}_{3322}) \\ &\quad - 2s^2 c^2 (w_{32}(-\omega) + w_{23}(\omega)), \\ \tilde{R}_{3211} &= \frac{1}{2} s c e^{-i\varphi} (w_{13}(\tilde{\omega}_{31} + \omega) + w_{13}(\tilde{\omega}_{21} + \omega) \\ &\quad - w_{12}(\tilde{\omega}_{31}) - w_{12}(\tilde{\omega}_{21})), \\ \tilde{R}_{3222} &= \frac{1}{2} s c e^{-i\varphi} (w_{21}(\tilde{\omega}_{12}) - w_{31}(\tilde{\omega}_{12} - \omega) + w_{23}(\omega) \\ &\quad - w_{32}(-\omega) + 2c^2 w_{23}(\tilde{\omega}_{32} + \omega) - 2s^2 w_{32}(\tilde{\omega}_{32} - \omega)), \\ \tilde{R}_{3233} &= \frac{1}{2} s c e^{-i\varphi} (w_{21}(\tilde{\omega}_{13}) - w_{31}(\tilde{\omega}_{13} - \omega) + w_{23}(\omega) \\ &\quad - w_{32}(-\omega) - 2c^2 w_{32}(\tilde{\omega}_{23} - \omega) + 2s^2 w_{23}(\tilde{\omega}_{23} + \omega)). \end{aligned}$$

It can be shown that dressed coherences $\tilde{\rho}_{31}$ and $\tilde{\rho}_{21}$ vanish in the case [Eqs. (15)] under consideration. In particular this is easy to see from the transformation formulas

$$\begin{aligned}\tilde{\rho}_{31}e^{i\varphi} &= c\sigma_{31}e^{i\varphi} - s\rho_{21}, \\ \tilde{\rho}_{21} &= c\rho_{21} + s\sigma_{31}e^{i\varphi},\end{aligned}\quad (27)$$

taking into account that $\sigma_{31}=0$ and $\rho_{21}=0$. The set of equations (25), like Eqs. (18), is valid for any level configuration [Figs. 1(a)–1(d)]. Taking as an example the scheme of Fig. 1(a) let us show that the relaxation terms (26) can be viewed as the relaxation rates between quantum dressed states $|\overline{i,n}\rangle$ of the total system involving atom and the quantized coherent field [12]. These dressed states can be expressed via the states $|i,n\rangle=|i\rangle|n\rangle$ of the uncoupled atoms and field as

$$\begin{aligned}|\overline{1,n}\rangle &= |1,n\rangle, \\ |\overline{2,n}\rangle &= c|2,n\rangle + se^{-i\varphi}|3,n-1\rangle, \\ |\overline{3,n}\rangle &= c|3,n-1\rangle - se^{i\varphi}|2,n\rangle,\end{aligned}\quad (28)$$

where n is the number of photons of the quantized coherent field. The relations between nonzero dipole moments $\vec{\mu}_{i,n;j,m}=\langle\overline{i,n}|\vec{\mu}|\overline{j,m}\rangle$ and $\vec{\mu}_{m',m}=\langle m'|\vec{\mu}|m\rangle$ of the allowed one-photon transitions in the quantum dressed-state and bare-state bases, respectively, have the forms

$$\begin{aligned}\vec{\mu}_{3,n-1;3,n} &= -sce^{-i\varphi}\vec{\mu}_{23}, \\ \vec{\mu}_{2,n-1;3,n} &= c^2\vec{\mu}_{23}, \\ \vec{\mu}_{1,n-1;3,n} &= c\vec{\mu}_{13}, \\ \vec{\mu}_{3,n-1;2,n} &= -s^2e^{-i2\varphi}\vec{\mu}_{23}, \\ \vec{\mu}_{2,n-1;2,n} &= sce^{-i\varphi}\vec{\mu}_{23}, \\ \vec{\mu}_{1,n-1;2,n} &= se^{-i\varphi}\vec{\mu}_{13}, \\ \vec{\mu}_{3,n;1,n} &= -se^{-i\varphi}\vec{\mu}_{21}, \\ \vec{\mu}_{2,n;1,n} &= c\vec{\mu}_{21}, \\ \vec{\mu}_{i,n;j,m} &\equiv \vec{\mu}_{i,n-1;j,m-1}, \\ \vec{\mu}_{j,m;i,n} &= \vec{\mu}_{i,n;j,m}^*, \\ \langle i,n|\vec{\mu}|j,m\rangle &= \vec{\mu}_{ij}\delta_{mn}.\end{aligned}\quad (29)$$

The dressed frequencies corresponding to these quantum transitions are defined as

$$\begin{aligned}\omega_{3,n;3,n-1} &= \omega_{2,n;2,n-1} = \omega_{1,n;1,n-1} = \omega, \\ \omega_{3,n;2,n-1} &= \tilde{\omega}_{32} + \omega, \\ \omega_{3,n;1,n-1} &= \tilde{\omega}_{31} + \omega,\end{aligned}$$

$$\omega_{2,n;3,n-1} = \tilde{\omega}_{23} + \omega, \quad (30)$$

$$\omega_{2,n;1,n-1} = \tilde{\omega}_{21} + \omega,$$

$$\omega_{3,n;1,n} = \tilde{\omega}_{31},$$

$$\omega_{1,n;2,n} = \tilde{\omega}_{12},$$

where it was implied that

$$\begin{aligned}\omega_{j,n;i,m} &\equiv \omega_{j,n-1;i,m-1}, \\ \omega_{i,n;j,m} &= (E_{i,n} - E_{j,m})/\hbar, \\ H|\overline{i,n}\rangle &= E_{i,n}|\overline{i,n}\rangle.\end{aligned}\quad (31)$$

Thus, using Eqs. (29) and (30), the relaxation rates (26) can be rewritten in the forms

$$\begin{aligned}\tilde{R}_{1122} &= \bar{w}_{21} + \bar{W}_{21}, \\ \tilde{R}_{2211} &= \bar{w}_{12} + \bar{Q}_{12}, \\ \tilde{R}_{1133} &= \bar{w}_{31} + \bar{W}_{31}, \\ \tilde{R}_{3311} &= \bar{w}_{13} + \bar{Q}_{13}, \\ \tilde{R}_{2233} &= \bar{W}_{32} + \bar{Q}_{32}, \\ \tilde{R}_{3322} &= \bar{W}_{23} + \bar{Q}_{23},\end{aligned}\quad (32)$$

$$\begin{aligned}\tilde{R}_{3232} &= -\frac{1}{2}(\tilde{R}_{1122} + \tilde{R}_{1133} + \tilde{R}_{2233} + \tilde{R}_{3322}) + 2(\bar{W}_{2233} + \bar{Q}_{2233}), \\ \tilde{R}_{3211} &= \bar{Q}_{1231} + \bar{Q}_{1321}^* + \bar{w}_{1231} + \bar{w}_{1321}^*, \\ \tilde{R}_{3222} &= \bar{Q}_{2232} + \bar{W}_{2232} + \bar{Q}_{2322}^* + \bar{W}_{2322}^* - \bar{w}_{3112} \\ &\quad - \bar{W}_{3112} - \bar{Q}_{3222} - \bar{W}_{3222} - \bar{Q}_{3332} - \bar{W}_{3332}, \\ \tilde{R}_{3233} &= \bar{Q}_{3233} + \bar{W}_{3233} + \bar{W}_{3323}^* + \bar{Q}_{3323}^* - \bar{w}_{2113} \\ &\quad - \bar{W}_{2113}^* - \bar{W}_{2223}^* - \bar{Q}_{2223}^* - \bar{W}_{2333}^* - \bar{Q}_{2333}^*.\end{aligned}$$

For convenience, we distinguish generalized rates of the one-photon transitions at the optical dressed frequencies, from the upper dressed level to the lower dressed level:

$$\begin{aligned}\bar{W}_{geji} &= \frac{2}{3\hbar c_0^3} \vec{\mu}_{g,n;e,n-1} \vec{\mu}_{j,n-1;i,n} \omega_{i,n;j,n-1}^3 \\ &\quad \times \eta(\omega_{i,n;j,n-1}) [N(\omega_{i,n;j,n-1}) + 1],\end{aligned}\quad (33)$$

$$\bar{W}_{mn} = 2\bar{W}_{mnmn}, \quad (34)$$

and from the lower dressed level to the upper dressed level (incoherent pumping):

$$\bar{Q}_{geji} = \frac{2}{3\hbar c_0^3} \vec{\mu}_{g,n-1;e,n} \vec{\mu}_{j,n;i,n-1} \omega_{j,n;i,n-1}^3 \times \eta(\omega_{j,n;i,n-1}) N(\omega_{j,n;i,n-1}), \quad (35)$$

$$\bar{Q}_{mn} = 2\bar{Q}_{mnmn}. \quad (36)$$

Generalized relaxation rates at the low-frequency dressed transitions are

$$\bar{w}_{geji} = \frac{2}{3\hbar c_0^3} \vec{\mu}_{g,n;e,n} \vec{\mu}_{j,n;i,n} \times \begin{cases} \omega_{i,n;j,n}^3 \eta(\omega_{i,n;j,n}) [N(\omega_{i,n;j,n}) + 1], & \omega_{i,n;j,n} > 0, \\ \omega_{j,n;i,n}^3 \eta(\omega_{j,n;i,n}) N(\omega_{j,n;i,n}), & \omega_{i,n;j,n} < 0, \end{cases} \quad (37)$$

$$\bar{w}_{mn} = 2\bar{w}_{mnmn}. \quad (38)$$

All the transitions are depicted in Fig. 2.

In the particular case $N(\omega) = 0$, i.e., if there is only spontaneous decay, the set of equation (25) for the case of level crossing [Fig. 1(a)], according to Eqs. (32)–(38), takes the most transparent forms

$$\begin{aligned} \frac{d}{dt} \tilde{\rho}_{11} &= -\bar{a}_{12} \tilde{\rho}_{11} + \bar{A}_{21} \tilde{\rho}_{22} + (\bar{a}_{31} + \bar{A}_{31}) \tilde{\rho}_{33}, \\ \frac{d}{dt} \tilde{\rho}_{22} &= \bar{a}_{12} \tilde{\rho}_{11} - (\bar{A}_{21} + \bar{A}_{23}) \tilde{\rho}_{22} + \bar{A}_{32} \tilde{\rho}_{33}, \\ \frac{d}{dt} \tilde{\rho}_{33} &= \bar{A}_{23} \tilde{\rho}_{22} - (\bar{a}_{31} + \bar{A}_{31} + \bar{A}_{32}) \tilde{\rho}_{33}, \end{aligned} \quad (39)$$

$$\begin{aligned} \frac{d}{dt} \tilde{\rho}_{32} + i\tilde{\omega}_{32} \tilde{\rho}_{32} &= -\bar{\Gamma}_{3232} \tilde{\rho}_{32} + \bar{\Gamma}_{3211} \tilde{\rho}_{11} \\ &+ \bar{\Gamma}_{3222} \tilde{\rho}_{22} + \bar{\Gamma}_{3233} \tilde{\rho}_{33}, \end{aligned}$$

where

$$\bar{\Gamma}_{3232} = \bar{A}_{21} + \bar{A}_{31} + \bar{A}_{32} + \bar{A}_{23} - 2\bar{A}_{3322},$$

$$\bar{\Gamma}_{3211} = (\bar{a}_{1321})^*, \quad (40)$$

$$\bar{\Gamma}_{3222} = -(\bar{A}_{3112} + \bar{A}_{3222} + \bar{A}_{3332}) + \bar{A}_{2232} + (\bar{A}_{2322})^*,$$

$$\bar{\Gamma}_{3233} = \bar{A}_{3233} + (\bar{A}_{3323} - \bar{A}_{2113} - \bar{a}_{2113} - \bar{A}_{2223} - \bar{A}_{2333})^*$$

and

$$\bar{A}_{geji} = \frac{2}{3\hbar c_0^3} \mu_{g,n;e,n-1} \mu_{j,n-1;i,n} \omega_{i,n;j,n-1}^3 \eta(\omega_{i,n;j,n-1}), \quad (41)$$

$$\bar{a}_{geji} = \frac{2}{3\hbar c_0^3} \mu_{g,n;e,n} \mu_{j,n;i,n} \omega_{i,n;j,n}^3 \eta(\omega_{i,n;j,n}) \quad (42)$$

are generalized spontaneous relaxation rates at the optical and low-frequency dressed transitions, respectively, $\bar{A}_{ij} = 2\bar{A}_{ijji}$ and $\bar{a}_{ij} = 2\bar{a}_{ijji}$ are the Einstein coefficients of the spontaneous relaxation at the corresponding optical dressed transition $|i,n\rangle \rightarrow |j,n-1\rangle$ and the low-frequency dressed transition $|i,n\rangle \rightarrow |j,n\rangle$:

$$\bar{A}_{ij} = \frac{4}{3\hbar c_0^3} |\mu_{j,n-1;i,n}|^2 \omega_{i,n;j,n-1}^3 \eta(\omega_{i,n;j,n-1}), \quad (43)$$

$$\bar{a}_{ij} = \frac{4}{3\hbar c_0^3} |\mu_{j,n;i,n}|^2 \omega_{i,n;j,n}^3 \eta(\omega_{i,n;j,n}). \quad (44)$$

IV. STRUCTURE OF THE FIELD-DEPENDENT RELAXATION

It follows from Sec. III that the relaxation rates of the dressed atoms are defined by the traditional Wigner-Weisskopf formulas where both bare dipole moments and bare frequencies are replaced by the dipole moments and frequencies of the dressed quantum transitions. This is especially transparent for coefficients (43) and (44) in the case $N(\omega) = 0$. In other words, we find that the relaxation terms $\bar{R}'_{m'n'n}$ depend on the dressed dipole moments and frequen-

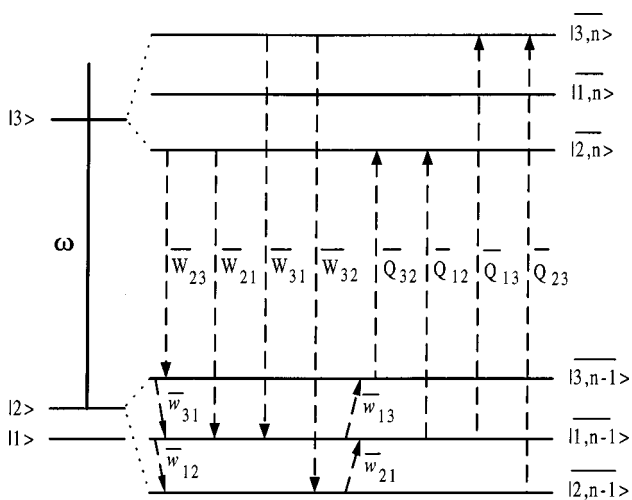


FIG. 2. Quantum dressed-state representation of a strongly driven three-level atomic system with a lower-level splitting when the Rabi splitting provides a crossing of one ac Stark level and a nearby unperturbed atomic state. Index n means the photon number of the driving field. Only one link with $n \gg 1$ of the infinite staircase, corresponding to different values of n , is plotted here.

cies in the same way as the bare relaxation terms $R_{m'mn'n}^{(0)}$ depend on the bare dipole moments and frequencies. With this recipe, it is quite simple to obtain the evolution equations of the driven atoms as the rate equations in the dressed-state basis by simply looking at Fig. 2.

Using Eqs. (25) with $\tilde{R}_{m'mn'n}$, and going back to the bare-state basis, we find evolution equations (18) with field-dependent relaxation rates. Conversely, starting from the usual evolution equations in the Wigner-Weisskopf approximation, and making the transformation to the dressed-state basis leads to equations similar to Eqs. (25) but where the relaxation terms depend on the bare frequencies instead of the dressed frequencies.

Another way to obtain Eqs. (25) is to use a full quantum description calculating the relaxation rates between quantum states according to the traditional Wigner-Weisskopf formula with dipole moments and frequencies of the dressed transitions instead of the bare ones, and afterward applying the quasiclassical approximation

$$\langle i, n | \rho | i, n \rangle \cong \langle i, n-1 | \rho | i, n-1 \rangle \cong p_0(n) \langle \tilde{i} | \rho | \tilde{i} \rangle = p_0(n) \tilde{\rho}_{ii}. \quad (45)$$

This implies that the driving field is in a coherent state with a Poisson distribution $p_0(n)$ for the photon number. This approach was suggested and developed earlier in a number of works [12]. It proved to be very efficient and transparent for analyzing the fluorescence and the probe field absorption spectra in atomic systems driven by a strong coherent field. Indeed, dressed frequencies define the position of the extrema in the spectra. However, the dependence of the relaxation rates on the dressed frequencies in this approach was usually ignored. Only the dependence of these constants on the dressed dipole moments was taken into account. In this case the evolution equations derived in that approach in the quasiclassical approximation coincide with those obtained from the traditional phenomenological equations (with field-independent relaxation), resulting from the basis transformation from bare to dressed states. Neglecting the dependence of the relaxation rates $R_{m'mn'n}$ on the driving field in the bare-state basis is correct only when the generalized Rabi frequency is the smallest parameter in the system. Recently it was shown that in a two-level system driven by a monochromatic field, such a dependence leads to a symmetry breaking of the atomic response with respect to the resonance [9]. Since the difference between dressed and bare frequencies in this case is always small (of the order of Ω/ω_{21} , where ω_{21} is the atomic transition frequency), the modification of the relaxation rates and hence atomic response is typically also weak. However, in principle, this can be observed even in free space [9]. Moreover, it can be strongly enhanced by placing atoms into the cavity or photonic band-gap material, exploiting the frequency dependence of the mode density $\eta(\omega)$ when it sharply varies on the scale of the Rabi frequency [2–5,8,9].

In a three-level system the relaxation rates are drastically modified even in free space when the Rabi splitting provides a crossing of the dynamic Stark sublevel and the nearby unperturbed atomic level. This leads to a change of sign of one of the dressed frequencies, and hence to a change of direction of the spontaneous emission between these two levels

due to the change of their relative position. For the scheme plotted in Fig. 1(a), this means the appearance of spontaneous emission from the former ground state to the lower Stark level which falls below the ground state [10].

It is worth emphasizing that this result implies the semiclassical approximation, when the driving field is characterized by a large number of photons. The crossing of a former ground state and a dynamic Stark level means a crossing of two quantum dressed states $|\bar{1}, n\rangle$ and $|\bar{2}, n\rangle$ of the total dynamical system with $n \gg 1$. There are of course no energy levels in the full dynamic system, which would fall below the ground state $|1, 0\rangle$ (see Ref. [13] and [10]).

It follows from Eq. (3) that if the relaxation rates of the bare atomic system are frequency independent, then dressing of atoms would not lead to field-dependent relaxation effects. This does not mean, however, that these effects would be eliminated in the case of a flat density of states of the field reservoir. Indeed, in this case the atomic relaxation rates would be proportional to the frequency, due to the fact that the transition probability between the two states of the system dipole coupled with the electromagnetic field is proportional to the frequency.

V. GENERAL SOLUTION OF THE MASTER EQUATIONS

The general solutions of the master equations (18) for arbitrary intensity and detuning of the driving field have the forms:

$$\begin{aligned} \sigma_{32} e^{i\varphi} &= \sigma'_{32} + i\sigma''_{32}, \\ \sigma'_{32} &= (R + 4\delta|\beta|n_{23}/W)/\tilde{W}, \\ \sigma''_{32} &= 2(|\beta|n_{23} - \delta R/W)/\tilde{W}, \\ \rho_{22} &= (C_2 A_3 - C_1 A_4)/D, \\ \rho_{33} &= (C_1 A_2 - C_2 A_1)/D, \\ D &= A_1 A_4 - A_2 A_3, \end{aligned} \quad (46)$$

where the coefficients A_j and C_j are given explicitly in Appendix B. Using transformation (13), one can also obtain the steady-state solution in the basis of the dressed states

$$\begin{aligned} \tilde{\rho}_{33} &= s^2 \rho_{22} + c^2 \rho_{33} - 2sc \operatorname{Re}(\sigma_{32} e^{i\varphi}), \\ \tilde{\rho}_{22} &= c^2 \rho_{22} + s^2 \rho_{33} + 2sc \operatorname{Re}(\sigma_{32} e^{i\varphi}), \\ \tilde{\rho}_{11} &= \rho_{11}, \\ \tilde{\rho}_{32} e^{i\varphi} &= sc(\rho_{33} - \rho_{22}) + c^2 \sigma_{32} e^{i\varphi} - s^2 \sigma_{23} e^{-i\varphi}. \end{aligned} \quad (47)$$

In the limit of high intensity, when the secular approximation and hence the set of equations (25) are valid, the physical picture in the dressed-state basis is especially transparent. According to Eqs. (25), off-diagonal elements do not influence the evolution of dressed populations, and one can easily obtain steady-state solutions for both populations of the

dressed states and dressed coherence $\tilde{\rho}_{32}$:

$$\begin{aligned}\tilde{\rho}_{22} &= r_2/D, \\ \tilde{\rho}_{33} &= r_3/D, \\ \tilde{\rho}_{32} &= -i(\tilde{R}_{3211}\tilde{\rho}_{11} + \tilde{R}_{3222}\tilde{\rho}_{22} + \tilde{R}_{3233}\tilde{\rho}_{33})/\tilde{\omega}_{32}, \\ r_2 &= \tilde{R}_{3311}\tilde{R}_{2233} - \tilde{R}_{2211}\tilde{R}_{3333}, \\ r_3 &= \tilde{R}_{2211}\tilde{R}_{3322} - \tilde{R}_{3311}\tilde{R}_{2222}, \\ D &= r_2 + r_3 + \tilde{R}_{1133}(\tilde{R}_{1122} + \tilde{R}_{3322}) + \tilde{R}_{1122}\tilde{R}_{2233}.\end{aligned}\quad (48)$$

In particular, at $N(\omega)=0$, when Eqs. (25) reduce to Eqs. (39), we have

$$\begin{aligned}r_2 &= \bar{a}_{12}(\bar{a}_{31} + \bar{A}_{31} + \bar{A}_{32}), \\ r_3 &= \bar{a}_{12}\bar{A}_{23}, \\ D &= r_2 + r_3 + (\bar{a}_{31} + \bar{A}_{31})(\bar{A}_{21} + \bar{A}_{23}) + \bar{A}_{21}\bar{A}_{32}, \\ \tilde{\rho}_{32} &= -i(\tilde{\Gamma}_{3211}\tilde{\rho}_{11} + \tilde{\Gamma}_{3222}\tilde{\rho}_{22} + \tilde{\Gamma}_{3233}\tilde{\rho}_{33})/\tilde{\omega}_{32}.\end{aligned}\quad (49)$$

In analogy with Eqs. (47), we can also find steady-state solutions for all the elements of the density matrix in the bare atomic basis:

$$\begin{aligned}\rho_{11} &= \tilde{\rho}_{11}, \\ \rho_{22} &= c^2\tilde{\rho}_{22} + s^2\tilde{\rho}_{33}, \\ \rho_{33} &= s^2\tilde{\rho}_{22} + c^2\tilde{\rho}_{33}, \\ \sigma_{32}e^{i\varphi} &= sc(\tilde{\rho}_{22} - \tilde{\rho}_{33}) + i\text{Im}(\tilde{\rho}_{32}e^{i\varphi}).\end{aligned}\quad (50)$$

To derive this result, we took into account that in the secular approximation [Eq. (24), $\text{Re}(\tilde{\rho}_{ij})=0$ if $i \neq j$. In the domain of the parameters Eq. (24)] where the secular approximation is valid, solutions (46) and (50) coincide. As noticed above, both sets of equations (18) and (25) are valid for any level configuration [Figs.1(a)–1(c)]. Only the relaxation rates are modified according to definitions (19) and (26) due to the change of the relative position of levels.

The effects caused by field-dependent relaxation are the most vivid when the spontaneous decay between levels $|2\rangle$ and $|1\rangle$ is the dominant relaxation process. This corresponds to a relatively large frequency interval between these two levels, $\hbar\omega_{21} \gg kT$, or to a ‘‘cold’’ reservoir, $N(\omega) \cong 0$ for $\omega \gg \omega_{21}$ (where k is the Boltzmann constant and T is the reservoir temperature).

VI. SCHEMES WITH A LOWER-LEVEL SPLITTING

The difference between the steady-state solutions of the traditional and generalized master equations is most drastic for the scheme plotted in Fig. 1(a). Indeed, according to the traditional equations, $N(\omega) \cong 0$ implies that all the atoms are in a ground state which is not coupled to the field. Since both

levels $|2\rangle$ and $|3\rangle$ are empty, there is no interaction at all at any intensity of the field. However, the steady-state solution (49) gives an essentially different picture.

A. Resonant driving

Let us first consider the case of resonant driving: $\delta=0$. Solutions (49) in this case and in the limit $|\beta| \gg \omega_{21}$ take the particularly simple forms [7]

$$\begin{aligned}\tilde{\rho}_{33} &= [1 + 2(a_0 + A_{31})(a_0 + A_{31} + A_{32})/a_0A_{32}]^{-1}, \\ \tilde{\rho}_{22} &= \tilde{\rho}_{33}[1 + 2(a_0 + A_{31})/A_{32}], \quad \tilde{\rho}_{11} = 1 - \tilde{\rho}_{22} - \tilde{\rho}_{33}, \\ \tilde{\rho}_{32}e^{i\varphi} &= \frac{i}{8|\beta|}(a_0 + \tilde{\rho}_{33}[a_0 + 2(3A_{31} + 2A_{32}) \\ &\quad + 2(A_{31}^2 - a_0^2)/A_{32}]), \\ a_0 &= A_{21}(|\beta|/\omega_{21})^3.\end{aligned}\quad (51)$$

In the bare atomic basis, according to Eqs. (50), we have

$$\begin{aligned}\rho_{22} &= \rho_{33} = \tilde{\rho}_{33}[1 + (a_0 + A_{31})/A_{32}], \\ \sigma_{32}e^{i\varphi} &= \tilde{\rho}_{33}(a_0 + A_{31})/A_{32} + \tilde{\rho}_{32}e^{i\varphi}.\end{aligned}\quad (52)$$

Solutions (52) correspond to the case when the low dynamic Stark level $|\tilde{2}\rangle$ is much lower than the ground state $|1\rangle$ (since $|\beta| \gg \omega_{21}$). In this case, according to Eq. (38), the spontaneous relaxation rate between these two levels $\bar{a}_{12}=a_0/2$ is proportional to the cube of the Rabi frequency: $a_0 \sim |\beta|^3$. This relaxation leads to a population in level $|\tilde{2}\rangle$ (and hence levels $|2\rangle$ and $|3\rangle$) which provides the interaction of atoms with the field. If this relaxation is weak, most of the atoms still remain in the ground state due to the fast spontaneous decay at the optical transition A_{31} . However, if $a_0 \gg A_{31}$ the situation is changed dramatically. In this case, according to Eqs. (52),

$$\tilde{\rho}_{33} \cong [1 + 2(a_0 + A_{32})/A_{32}]^{-1}.\quad (53)$$

If $A_{31} \ll a_0 \ll A_{32}$, all levels are equally populated, both in the bare and dressed bases. If $a_0 \gg \{A_{31}, A_{32}\}$, most of atoms are trapped at the lower Stark level: $\tilde{\rho}_{22} \cong 1$. This condition implies that the following inequalities are satisfied:

$$\begin{aligned}\mu_{21}^2 \eta(|\beta|)(|\beta|/\omega_{31})^3/\mu_{31}^2 \eta(\omega_{31}) &\gg 1, \\ \mu_{21}^2 \eta(|\beta|)(|\beta|/\omega_{32})^3/\mu_{32}^2 \eta(\omega_{32}) &\gg 1.\end{aligned}\quad (54)$$

These results are quite different from those which would be obtained in the case of a driven two-level system where populations of the dressed states at resonance are equal. In the bare basis, trapping to the lower Stark level corresponds to a saturation of the $|2\rangle$ - $|3\rangle$ transition, $\rho_{22} \cong \rho_{33} \cong 1/2$, while the ground state becomes almost fully depleted [14]. Hence in the bare atomic basis, the population distribution is similar to that which would be obtained in a closed two-level system $|2\rangle$ - $|3\rangle$ driven by a coherent field. However since the Stark

level $|\bar{2}\rangle$ is a coherent superposition of the two atomic levels $|2\rangle$ and $|3\rangle$, the atomic coherence reaches its maximum value

$$\sigma_{32}e^{i\varphi} \cong 1/2.$$

This is a rather rare example wherein spontaneous decay helps to prepare atoms in an almost pure quantum state with the maximal possible coherence: $|\sigma_{32}| \cong \sqrt{\rho_{22}\rho_{33}}$. This result is in opposition to what would take place in a closed two-level system driven by a coherent field, where the coherence tends to zero in the limit of high intensity [15], i.e., saturation is not accompanied by high coherence.

Resonant driving of the $|3\rangle$ - $|2\rangle$ transition provides a method for depleting the ground state. It is different from optical pumping and saturation. This depletion leads to a population inversion both at the $|2\rangle$ - $|1\rangle$ and $|3\rangle$ - $|1\rangle$ transitions. The last result is unexpected since, according to the traditional master equations, coherent driving in a three-level system cannot produce population inversion at a transition whose frequency is higher than the frequency of the driven transition. Apparently the population inversion at the $|3\rangle$ - $|1\rangle$ transition is possible due to multiphoton absorption of the driving field. The magnitude of $\text{Re}(\sigma_{32}e^{i\varphi})$, characterizing the refractive index, tends to 1/2 in the limit of high intensity, while in a closed two-level driven system at resonance it vanishes. It follows from this that the ratio of the real and imaginary parts of the susceptibility is, $\text{Re}(\sigma_{32}e^{i\varphi})/\text{Im}(\sigma_{32}e^{i\varphi}) \gg 1$, when $|\beta| \gg \omega_{21}$, while in a closed two-level system it is zero.

The absorption power remains at zero until the level crossing occurs at $|\beta| = \omega_{21}$. Then it increases with the driving intensity. In the limit of high intensity ($a_0 \gg \{A_{31}, A_{32}\}$), according to Eqs. (46), we obtain

$$\begin{aligned} P &= 2N\hbar\omega \text{Im}(\sigma_{32}\beta^*)|_{\delta=0} \\ &\cong 2N\hbar\omega \frac{2|\beta|^2(A_{31}+A_{32})}{8|\beta|^2+3a_0(A_{31}+A_{32})/4}. \end{aligned} \quad (55)$$

Comparing this result with the absorption power in an equivalent two-level system,

$$P_2 = 2N\hbar\omega \text{Im}(\sigma_{32}\beta^*)|_{\delta=0} \cong 2N\hbar\omega \frac{2|\beta|^2A}{8|\beta|^2+A^2}$$

(where the spontaneous decay rate is taken to be equal to the sum of the two optical decay rates in a three-level system: $A = A_{31} + A_{32}$), we conclude that P is smaller than P_2 . The two absorption powers tend toward the same value for $3A^2 \ll 3a_0A \ll 32|\beta|^2$, i.e., at $(A/A_{21})^{1/3}\omega_{21} \ll |\beta| \ll 32\omega_{21}^3/(3AA_{21})$. Hence trapping of atoms in a dressed state does not lead to a reduction of the absorption power. This result is due to the fact that the absorption power is proportional to the product of the number of absorbing (untrapped) atoms times their decay rate. Although the number of absorbing atoms NA/a_0 decreases, their decay rate a_0 increases with an increase of the intensity. As a result, the absorption power in this limit is the same as in a two-level system ($P = N\hbar\omega A/2$). This result is also quite obvious from the fact

that the population distribution in a three-level system in this case is the same ($\rho_{22} \cong \rho_{33} \cong 1/2, \rho_{11} \cong 0$) as in a closed two-level system $|2\rangle$ - $|3\rangle$.

Since $a_0 \sim |\beta|^3$, it seems at first glance that in the limit of very high intensity, when $32|\beta|^2 \ll 3a_0A \ll 3a_0^2$, the absorption power in a three-level system turns to zero. However, this limit implies that the decay rates of the dressed states exceed the frequency spacing between them ($\bar{a}_{31} \gg \omega_{3,n;1,n}$, $\bar{a}_{12} \gg \omega_{1,n;2,n}$, see Fig. 2), and hence violate the Markov approximation. Apparently, in the limit $|\beta| \gg \omega_{21}$ the relative position of levels $|2\rangle$ and $|1\rangle$ is not important, and hence solutions (51) and (52) also remain true for the scheme plotted in Fig. 1(b).

B. Arbitrary detuning

In Fig. 3 we plot $\text{Re}(\sigma_{32}e^{i\varphi})$, the absorption power $P = 2N\hbar\omega \text{Im}(\sigma_{32}\beta^*)$, as well as the populations of both bare and dressed levels as the functions of the normalized detuning $\delta/|\beta|$ for different values of the field intensity, according to solutions (46), for the first scheme [Fig. 1(a)]. Contrary to the case of a two-level driven system, all these curves are asymmetric functions of the detuning. This is due to the fact that negative detuning is favorable for the level crossing while positive detuning prevents it. For each value of the Rabi frequency $|\beta|$ there exists a critical magnitude of the detuning corresponding to a crossing between the lower Stark level and the ground state:

$$\delta^* = -\omega_{21}(1 - |\beta|^2/\omega_{21}^2). \quad (56)$$

Depending on the ratio $|\beta|/\omega_{21}$, the value of δ^* may be positive or negative. For $\delta \gg \delta^*$, the field does not interact with atoms. This would also be the case with traditional master equations with field-independent relaxation. But for $\delta < \delta^*$ (i.e., as soon as the lower Stark level falls below the ground state) spontaneous relaxation via the channel $|1\rangle$ - $|\bar{2}\rangle$ provides some population to level $|\bar{2}\rangle$ [see Fig. 3(d)]. This leads to the population of the atomic levels $|2\rangle$ and $|3\rangle$ [Fig. 3(c)] and therefore to the appearance of a nonzero atomic response [Figs. 3(a) and 3(b)]: $\text{Re}(\sigma_{32}e^{i\varphi}) \neq 0$ and $\text{Im}(\sigma_{32}e^{i\varphi}) \neq 0$. According to the traditional master equations, interaction would still be impossible. It is worth emphasizing that even a very weak coherent field ($|\beta| \rightarrow 0$) leads to a nontrivial atomic response if the magnitude of the negative detuning is large enough to provide a crossing of the lower Stark level with the ground state ($\Omega \cong |\delta| > \omega_{21}$), and hence spontaneous decay from the ground state to this Stark level. Increasing the intensity, a nonzero atomic response is produced in a larger and larger domain of positive detuning [Figs. 3(a) and 3(b)]. For each value of the detuning, as in the case of exact resonance, a distribution of dressed populations is defined by the ratio of the spontaneous decay rates \bar{a}_{ij} [Eq. (44)] at the low-frequency transitions and the spontaneous decay rates at the dressed optical transitions \bar{A}_{mn} [Eq. (43)] [Fig. 3(d)].

In the limit of large negative detuning ($\delta < 0$, $|\delta|/|\beta| \gg 1$), according to Eqs. (43) and (44) we have

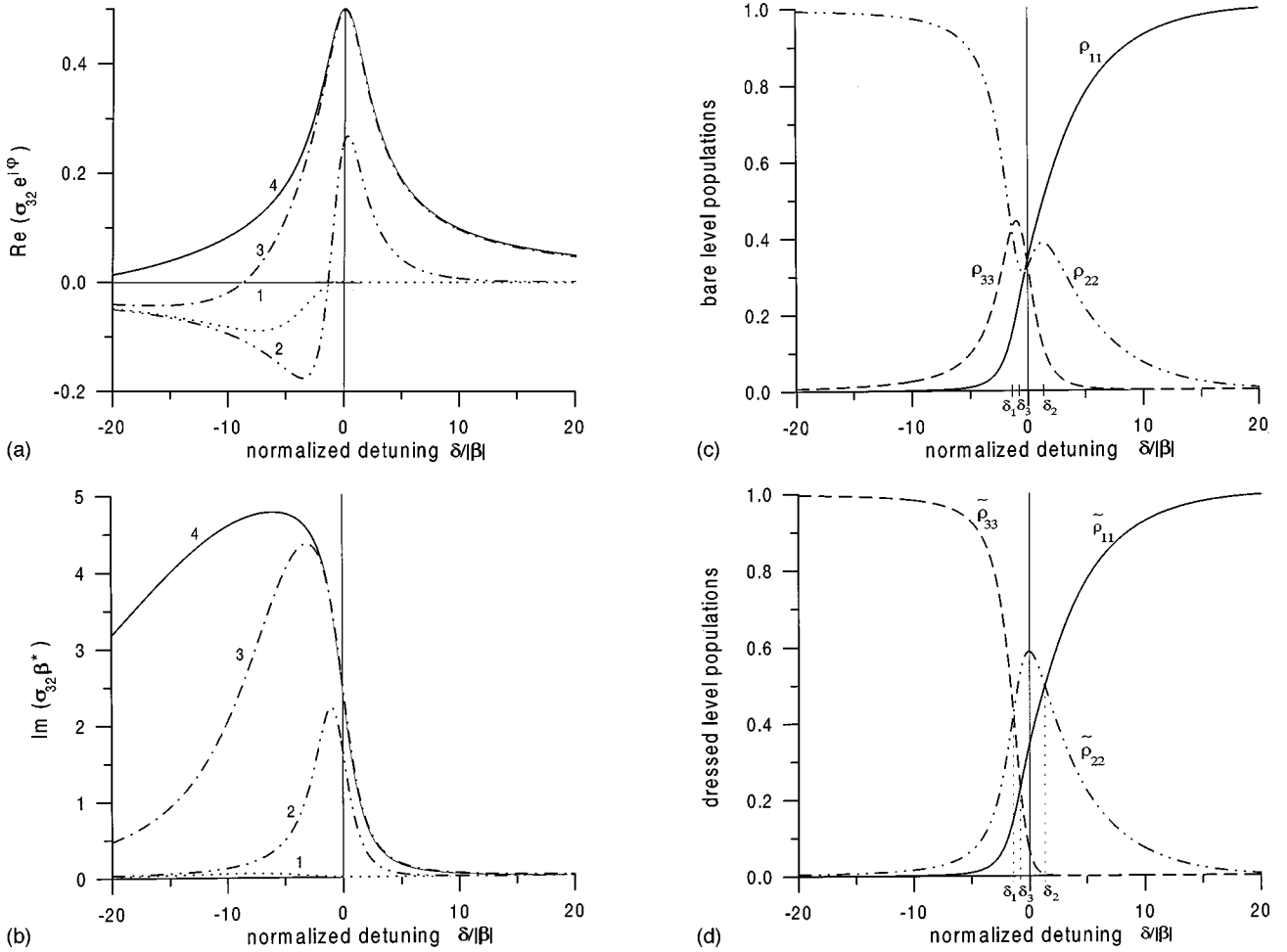


FIG. 3. (a) $\text{Re}(\sigma_{32}e^{i\varphi})$, which is proportional to the real part of susceptibility; (b) $\text{Im}(\sigma_{32}\beta^*)$, which is proportional to the absorption power; (c) populations ρ_{ii} of the bare atomic levels; and (d) populations $\tilde{\rho}_{ii}$ of the dressed levels for the scheme of lower level splitting [Fig. 1(a)] vs the driving field detuning δ in units of resonant Rabi frequency $|\beta|$ for $A_{21}/\omega_{21}=10^{-6}$, $A_{31}/\omega_{21}=A_{32}/\omega_{21}=10^{-2}$, $\omega_{31}/\omega_{21}=\omega_{32}/\omega_{21}=10^5$, $\eta(\omega)=1$, and $N(\omega)=0$. (1) $|\beta|/\omega_{21}=5$. (2) $|\beta|/\omega_{21}=30$. (3) $|\beta|/\omega_{21}=180$. (4) $|\beta|/\omega_{21}=500$ for (a) and (b) and $|\beta|/\omega_{21}=30$ for (c) and (d).

$$\bar{a}_{12} \cong |\delta|A_{21}|\beta|^2/\omega_{21}^3,$$

$$\bar{a}_{31} \cong A_{21}, \quad \bar{A}_{21} \cong A_{31},$$

$$\bar{A}_{23} \cong A_{32}, \quad \bar{A}_{31} \rightarrow 0,$$

$$\bar{A}_{32} \rightarrow 0.$$

After substitution of these expressions into Eqs. (49), we find

$$\begin{aligned} \tilde{\rho}_{22} &\cong \frac{A_{21}|\beta|^2|\delta|/\omega_{21}^3}{A_{31}+A_{32}+(A_{21}+A_{32})|\beta|^2|\delta|/\omega_{21}^3}, \\ \tilde{\rho}_{33} &\cong \frac{A_{32}|\beta|^2|\delta|/\omega_{21}^3}{A_{31}+A_{32}+(A_{21}+A_{32})|\beta|^2|\delta|/\omega_{21}^3}. \end{aligned} \quad (57)$$

Below we consider the particular case $A_{31} \leq A_{32}$ since, according to the selection rules, if transitions $|1\rangle-|2\rangle$ and $|3\rangle-|2\rangle$ are allowed, transition $|3\rangle-|1\rangle$ typically should be forbidden ($A_{31}=0$). Then for a weak coherent driving, $|\beta|^2|\delta|/\omega_{21}^3 \ll 1$, according to Eqs. (57) $\tilde{\rho}_{22} \cong \tilde{\rho}_{33} \cong 0$ and

hence, according to Eqs. (50), for $|\delta| \gg |\beta|$, we find that $\rho_{22} \cong \tilde{\rho}_{33} \cong 0$ and $\rho_{33} \cong \tilde{\rho}_{22} \cong 0$, i.e., atoms remain in the ground state. Nevertheless spontaneous emission from the ground state still may be observed in this weak-field limit for $|\delta| > \omega_{21}$, since level crossing takes place due to the large detuning. As noticed recently [15,10] this weak-field limit also allows for a transparent interpretation of this process in the bare-state picture via Raman scattering, involving spontaneous emission of a photon with the frequency $\omega_1 \cong |\delta| - \omega_{21}$ and absorption of a photon of the driving field [Fig. 4(a)].

At the same time, at $|\beta|^2|\delta|/\omega_{21}^3 \gg 1$, we obtain, according to Eqs. (57),

$$\begin{aligned} \tilde{\rho}_{22} &\rightarrow \frac{A_{21}}{A_{21}+A_{32}}, \\ \tilde{\rho}_{33} &\rightarrow \frac{A_{32}}{A_{21}+A_{32}}. \end{aligned} \quad (58)$$

Note that the last inequality along with the rotating-wave approximation ($|\delta| \ll \omega_{32}$), implies a sufficiently large driving field intensity: $|\beta|^2 \gg \omega_{21}^3/\omega_{32}$. In the most interesting

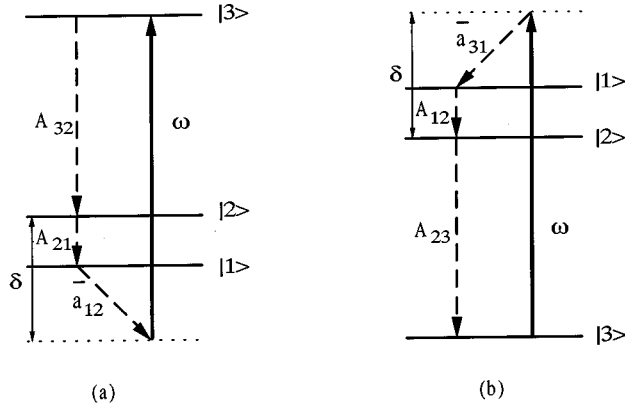


FIG. 4. Raman scattering of the strong driving field in the case of the lower-level splitting (a), and the upper-level splitting (b).

and typical situation, when spontaneous decay at the driven optical transition is much faster than at the unperturbed low-frequency transition ($A_{32}/A_{21} \gg 1$), we have

$$\begin{aligned} \tilde{\rho}_{22} &\cong \frac{A_{21}}{A_{32}} \ll 1, \\ \tilde{\rho}_{33} &\cong \frac{A_{32}}{A_{31}} = 1. \end{aligned} \quad (59)$$

Hence atoms are trapped in the upper ac Stark level, as would be the case for the closed two-level system $|2\rangle$ - $|3\rangle$ at a large negative detuning. Accordingly, the atomic response in this domain of δ (Fig. 3) looks similar to the atomic response of the two-level closed system. In the limit under consideration, $|\delta|/|\beta| \gg 1$, $\delta < 0$ (which implies $c \rightarrow 0$ and $s \rightarrow 1$), trapping in the dressed state $|\bar{3}\rangle$ is equivalent to trapping in the bare state $|2\rangle$ [see Eqs. (50)]. At first sight, this result seems amazing. Indeed, it seems that the interaction with the field should become weaker with an increase of the detuning, and hence atoms should rather remain in the ground state. However, the distribution of populations among levels $|1\rangle$ and $|2\rangle$ is defined by the ratio of the transition rate of the two-photon Raman process [Fig. 4(a)] to the spontaneous decay rate at the $|2\rangle$ - $|1\rangle$ transition. The first rate increases with a detuning due to the fact that spontaneous photons with higher frequencies participate in the process. It is easy to verify that condition $|\beta|^2 |\delta|/\omega_{21}^3 \gg 1$ corresponds precisely to prevailing of Raman scattering over the spontaneous emission at the $|2\rangle$ - $|1\rangle$ transition [10].

Close to resonance, spontaneous decay (modified by the weak driving field) at the low-frequency transition remains sufficiently small compared to both optical transitions, so that most of the atoms remain almost unaffected by the field i.e., they remain in the ground state: $\tilde{\rho}_{11} = \rho_{11} \cong 1$. With a further increase of the driving intensity and hence of the spontaneous decay rate at the low-frequency transition ($\bar{a}_{12} > A_{31}, A_{32}$) around resonance there appears a domain of detuning:

$$\delta_1 < \delta < \delta_2,$$

$$\delta_1 \cong \frac{|\beta|}{1 - (A_{32}/A_{21})^{1/3}},$$

$$\delta_2 \cong \frac{p^3 |\beta|}{2(3p^2 + A_{31}/A_{21})},$$

$$p = |\beta|/\omega_{21},$$

where $\tilde{\rho}_{22} > \{\tilde{\rho}_{11}, \tilde{\rho}_{33}\}$. Hence atoms can be trapped in the lower Stark level [Fig. 3(d)], contrary to the case of a closed two-level system where populations of both dressed states would be equal. In the bare atomic basis for the range of negative detuning,

$$\delta_1 < \delta < 0,$$

population inversion becomes possible both at the driven $|3\rangle$ - $|2\rangle$ transition and at the adjacent $|3\rangle$ - $|1\rangle$ transition: $\rho_{33} > \{\rho_{11}, \rho_{22}\}$. This is an interesting effect which is completely due to field-dependent relaxation.

Figures 3(a) and 3(b) illustrate the striking difference between the atomic response of the closed two-level system $|2\rangle$ - $|3\rangle$ and the atomic response of our three-level system in the case $\bar{a}_{12} > A_{31}, A_{32}$. For increasing intensity, the dispersion curve $\text{Re}(\sigma_{32} e^{i\varphi})$, as a function of $\delta/|\beta|$, becomes more symmetric with respect to the resonant driving, and looks rather similar to the absorption profile of a two-level system; meanwhile the absorption curve $\text{Im}(\sigma_{32} \beta^*)$, as a function of $\delta/|\beta|$, acquires some dispersive features. In particular, its maximum is shifted from resonance to the domain of negative detuning [Fig. 3(b)], and it may exceed the maximum value occurring in a two-level case. Let us note that the nonmonotonic behavior of the maxima of these curves with an increase of intensity is due to our normalization of δ to the Rabi frequency $|\beta|$.

VII. MODIFICATION OF THE PROBE FIELD ABSORPTION SPECTRA

The distribution of dressed populations allows one to define the major characteristics of both fluorescence and probe field absorption spectra for a probe field coupled with any of three atomic transitions. Drastic modifications of this distribution due to field-dependent relaxation provide the appearance of qualitatively interesting features in such spectra. Let us consider these features for the scheme plotted in Fig. 1(a).

According to traditional master equations in the case under consideration ($\hbar\omega_{21} \gg kT$), there should be no fluorescence at all since the field does not interact with the atoms. However, according to generalized equations, if the driving of the $|3\rangle$ - $|2\rangle$ transition provides a crossing of the lower Stark level with the ground atomic state, an interaction with the coherent field is switched on and the fluorescence appears, in general, at all the allowed dressed transitions (see Fig. 2), including the $|1, n-1\rangle \rightarrow |2, n-1\rangle$ transition. Trapping atoms into one of two ac Stark levels leads to an increase of the fluorescence from this trapped state and a decrease of the fluorescence from another state.

According to traditional master equations, if the probe field is coupled to the $|2\rangle$ - $|3\rangle$ transition, it does not interact with the atoms for any intensity of the driving field. However, according to our results, interaction with both driving

and probe fields is switched on as soon as one ac Stark level crosses a nearby atomic state. Qualitatively the absorption spectrum looks similar to the well-known Mollow spectrum for a two-level driven system. That is, there are two extrema at the dressed frequencies $\omega_{2,n;3,n-1}$ and $\omega_{3,n;2,n-1}$. For $\delta < \delta_1 < 0$ [Fig. 3(d)], there is an absorption peak at $\omega_{2,n;3,n-1}$ and an amplification peak at $\omega_{3,n;2,n-1}$ since $\tilde{\rho}_{33} > \tilde{\rho}_{22}$. Conversely, for $\delta > \delta_1$ there is amplification at $\tilde{\omega}_{23}$ and absorption at $\tilde{\omega}_{32}$. Let us note that amplification at one of the dressed transitions and absorption at another one can occur either with population inversion at the bare $|3\rangle$ - $|2\rangle$ transition (at $\delta_1 < \delta < 0$) or without population inversion at the bare $|3\rangle$ - $|2\rangle$ transition (at $\delta > 0$ or at $\delta < \delta_1$), as shown in Figs. 3(c) and 3(d).

If the probe field is coupled to the $|1\rangle$ - $|2\rangle$ transition, two extremes at the frequencies $\omega_{3,n;1,n}$ and $\omega_{1,n;2,n}$ should be observed. When $\delta < \delta_3$ [Fig. 3(d)], the probe field is amplified at the frequency $\omega_{3,n;1,n}$ (since $\tilde{\rho}_{33} > \tilde{\rho}_{11}$) and absorbed at the frequency $\omega_{1,n;2,n}$ (since $\tilde{\rho}_{22} > \tilde{\rho}_{11}$). When $\delta_3 < \delta < \delta_2$, the probe field is absorbed at both dressed transitions. When $\delta > \delta_2$, the probe field is amplified at the frequency $\omega_{1,n;2,n}$ and absorbed at the frequency $\omega_{3,n;1,n}$.

Finally, when the probe field is coupled with the $|1\rangle$ - $|3\rangle$ transition, according to traditional master equations one would obtain the well-known absorption profile described by the Autler-Townes doublet with two maxima in the absorption spectrum corresponding to a tuning of the probe field to the dressed transitions $|1,n-1\rangle$ - $|3,n\rangle$ or $|1,n-1\rangle$ - $|2,n\rangle$ (Fig. 2). According to our analysis, in the domain of driving field detuning $\delta < \delta_2$ [Fig. 3(d)] we have $\tilde{\rho}_{22} > \tilde{\rho}_{11}$, and hence amplification of the probe field tuned to the frequency $\omega_{2,n;1,n-1}$ of the dressed transition $|2,n\rangle \rightarrow |1,n-1\rangle$ should appear. Note that this amplification may occur without population inversion at the bare $|1\rangle$ - $|3\rangle$ transition ($\rho_{11} > \rho_{33}$) at $\delta^* < \delta < \delta_2$ [Fig. 3(c)], where δ^* corresponds to a crossing of the ρ_{11} line with the ρ_{33} line. At $\delta < \delta^*$, amplification of the probe field is accompanied by population inversion at the bare $|1\rangle$ - $|3\rangle$ transition (which is created by the monochromatic field). Moreover, for $\delta_3 < \delta < \delta^*$ [Fig. 3(c)], absorption at the $\omega_{3,n;1,n-1}$ frequency should occur in spite of a steady-state population inversion at the bare transition $|1\rangle$ - $|3\rangle$.

In order to analyze the absorption spectra in detail on the basis of the generalized master equations, we need to add to this set of equations dynamical terms describing the interaction with the probe field. It is supposed that this field is too weak to alter the relaxation processes in the system. Hence the set of master equations has forms:

$$\begin{aligned} \dot{\rho}_{11} + 2 \operatorname{Im}(\alpha^* \sigma_{31}) &= -(a_{12} + w_{13})\rho_{11} + a_{21}\rho_{22} + w_{31}\rho_{33} \\ &\quad - (a_{31} + a_1)\operatorname{Re}(\sigma_{32}e^{i\varphi}), \\ \dot{\rho}_{22} + 2 \operatorname{Im}(\beta^* \sigma_{32}) &= a_{12}\rho_{11} - (a_{21} + w_{23})\rho_{22} + w_{32}\rho_{33} \\ &\quad + (a_1 - \tilde{a}_{32})\operatorname{Re}(\sigma_{32}e^{i\varphi}), \\ \dot{\rho}_{33} - 2 \operatorname{Im}(\beta^* \sigma_{32} + \alpha^* \sigma_{31}) \\ &= w_{13}\rho_{11} + w_{23}\rho_{22} - (w_{31} + w_{32})\rho_{33} \\ &\quad + (a_{31} + \tilde{a}_{32})\operatorname{Re}(\sigma_{32}e^{i\varphi}), \end{aligned} \quad (60)$$

$$\dot{\sigma}_{32} + i\delta\sigma_{32} - i\beta n_{23} - i\alpha\sigma_{21}^* = -\Gamma_{32}\sigma_{32} + e^{-i\varphi}R/2,$$

$$\begin{aligned} \dot{\sigma}_{31} + i\delta_a\sigma_{31} - i\beta\sigma_{21} - i\alpha n_{13} &= -\Gamma_{31}\sigma_{31} \\ &\quad + e^{-i\varphi}(a_{31} + a_{32})\sigma_{21}/2, \end{aligned}$$

$$\begin{aligned} \dot{\sigma}_{21} + i\delta_c\sigma_{21} - i\beta^*\sigma_{31} + i\alpha\sigma_{32}^* \\ = -\Gamma_{21}\sigma_{21} + e^{-i\varphi}(a_1 - a_{23})\sigma_{31}/2, \end{aligned}$$

where $\alpha = (\tilde{\mu}_{31}\tilde{E}_a)/(2\hbar) = |\alpha|e^{-i\varphi_a}$ is the complex amplitude of the probe weak field, and

$$\Gamma_{31} = (a_{21} + w_{31} + w_{13} + w_{32})/2,$$

$$\Gamma_{21} = (a_{21} + a_{12} + w_{13} + w_{23})/2,$$

$$\delta_a = \omega_{31} - \omega_a,$$

$$\delta_c = \omega_{21} - (\omega_a - \omega) = \delta_a - \delta.$$

The absorption coefficient is defined by

$$\begin{aligned} \operatorname{Im}(\sigma_{31}/\alpha) &= \frac{1}{|D_a|^2} \{ [\Gamma_{21}n_{13} - |\beta|\sigma_{32}'' - (a_{31} + a_{32})\sigma_{32}'/2]D_a' \\ &\quad + [\delta_c n_{13} - |\beta|\sigma_{32}' + (a_{31} + a_{32})\sigma_{32}''/2]D_a'' \}, \end{aligned} \quad (61)$$

where

$$D_a = D_a' + iD_a'',$$

$$D_a' = \Gamma_{31}\Gamma_{21} - (a_{31} + a_{32})(a_1 - a_{23})/4 + |\beta|^2 - \delta_a\delta_c,$$

$$D_a'' = \Gamma_{31}\delta_c + \Gamma_{21}\delta_a - (a_{31} + a_{32} + a_{21} - a_{23})|\beta|/2.$$

This is plotted in Fig. 5 as a function of the probe field detuning for two different values of driving field frequency. In the case of resonant driving [Fig. 5(a)], the probe field is absorbed when tuned to the $|1,n-1\rangle$ - $|3,n\rangle$ dressed transition, and it is amplified when it is tuned to $|1,n-1\rangle$ - $|2,n\rangle$ dressed transition in the absence of population inversion between bare states. In the case of detuned driving [$\delta \cong \delta_1$ in Fig. 3(d)], amplification of the probe field occurs at both the $|1,n-1\rangle$ - $|3,n\rangle$ and $|1,n-1\rangle$ - $|2,n\rangle$ dressed transitions. This is in a full agreement with the signs of dressed-state population differences, as was explained above.

VIII. SCHEMES WITH AN UPPER-LEVEL SPLITTING

For these schemes, the lower operating level is the ground state. Hence at any intensity of the field there is a finite atomic response according to both the traditional and generalized master equations. In this sense, the modification of the atomic response due to field-dependent relaxation is not as drastic as in the first scheme of Fig. 1, where the atomic response (if there exists only spontaneous relaxation) is simply zero at any intensity of the field in the frame of the traditional master equations neglecting field-dependent relaxation. Nevertheless there is still a striking difference between the solutions of the traditional and generalized master equa-

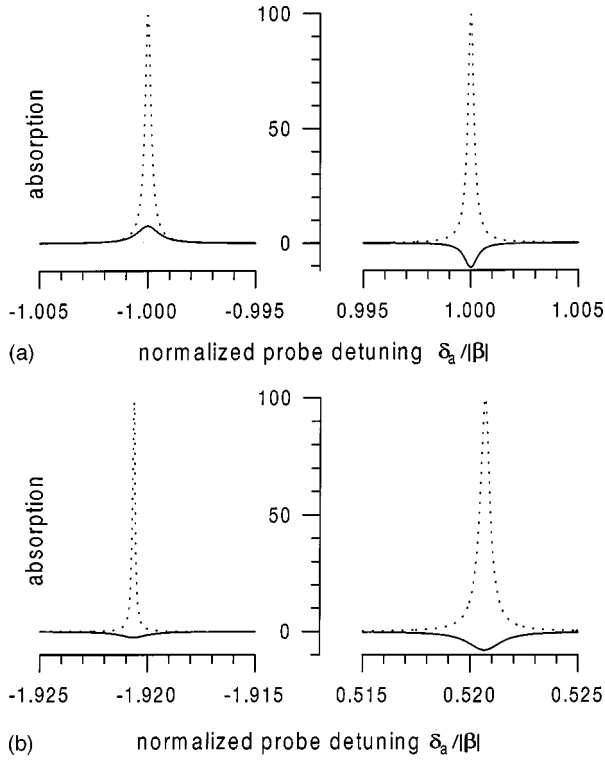


FIG. 5. Amplification profile of the probe field at the $|3\rangle\text{-}|1\rangle$ transition (bold line) in comparison with the solution of the traditional master equations (dashed line) when the driving field detuning from the $|3\rangle\text{-}|2\rangle$ transition is fixed at (a) $\delta=0$ [in this case, $\tilde{\rho}_{22} > \tilde{\rho}_{11}$ and $\tilde{\rho}_{33} < \tilde{\rho}_{11}$ in Fig. 3(d)]. (b) $\delta/|\beta| \approx \delta_1 = -1.4$ [in this case, $\tilde{\rho}_{33} \approx \tilde{\rho}_{22} > \tilde{\rho}_{11}$ in Fig. 3(d)].

tions in the case of sufficiently high intensity. In fact, the third scheme of Fig. 1 is the most convenient for a comparison of these solutions. Indeed, in the case of spontaneous decay according to the traditional master equations, the upper level in this scheme is not involved at all in the interaction with the field, and hence the atomic response is the same as in the two-level system $|2\rangle\text{-}|3\rangle$, whose properties are well known [15]. In particular, at resonance $\delta=0$ and in the limit of high intensity $|\beta| \gg A_{23}$, one obtains

$$\begin{aligned}
 \tilde{\rho}_{22} &\approx \tilde{\rho}_{33} \approx 1/2, \\
 \tilde{\rho}_{11} &\approx 0, \\
 \tilde{\sigma}_{32} &\approx 0, \quad \rho_{22} \approx \rho_{33} \approx 1/2, \\
 \text{Re}(\sigma_{32}e^{-i\varphi}) &= 0, \\
 \text{Im}(\sigma_{32}e^{-i\varphi}) &\approx 0, \\
 P &\approx N\hbar\omega A_{23}/2.
 \end{aligned} \tag{62}$$

According to the generalized equations, as long as the upper Stark level does not cross level $|3\rangle$ (which for $\delta=0$ means $|\beta| \leq \omega_{12}$) the atomic response is the same as in a two-level system. However, as soon as such a crossing occurs, spontaneous decay at the transition $|\tilde{3}\rangle \rightarrow |1\rangle$ [Fig. 1(c)] involves

the upper atomic state into the interaction process. On resonance ($\delta=0$) and in the limit $|\beta| \gg \omega_{12}$ from Eq. (48) we obtain

$$\begin{aligned}
 \tilde{\rho}_{33} &= A_{23}(a_0 + 2A_{13})/D, \\
 \tilde{\rho}_{22} &= [A_{13}A_{23} + (a_0 + A_{13})(2a_0 + A_{23})]/D, \\
 \rho_{22} = \rho_{33} &= [A_{13}A_{23} + (a_0 + A_{13})(a_0 + A_{23})]/D, \\
 \sigma_{32}e^{i\varphi} &= a_0(a_0 + A_{13})/D,
 \end{aligned} \tag{63}$$

$$D = A_{13}A_{23} + (a_0 + A_{13})(2a_0 + 3A_{23}).$$

In the limit $|\beta| \gg \omega_{12}$, the relative position of levels $|1\rangle$ and $|2\rangle$ is indifferent, and the same solution holds for the scheme of Fig. 1(d). According to Eqs. (63), for $a_0 \gg A_{23}$ but independently of the ratio a_0/A_{13} , all atoms are trapped to the lower Stark level $|\tilde{2}\rangle$:

$$\begin{aligned}
 \tilde{\rho}_{22} &\approx 1, \\
 \tilde{\rho}_{11} &\approx \tilde{\rho}_{33} \approx 0,
 \end{aligned}$$

in analogy with the schemes with lower level splitting. In the bare atomic states, trapping to a Stark level, as already discussed above, corresponds to an equalization of populations $\rho_{22} \approx \rho_{33} \approx 1/2$ as in a two-level system. However, opposite to the case of a two-level system where $\sigma_{32} \approx 0$, the excitation of the coherence is maximal ($\sigma_{32}e^{i\varphi} \approx 1/2$). Accordingly $\text{Re}(\sigma_{32}e^{i\varphi})/\text{Im}(\sigma_{32}e^{i\varphi}) \gg 1$ (instead of zero) in the limit of high intensity, while the absorption power at $a_0 \gg A_{23}$ saturates to its maximal value $P = N\hbar\omega A_{23}/2$.

As in schemes with lower-level splitting, both the dispersion profile, given by $\text{Re}(\sigma_{32}e^{i\varphi})$, and the absorption profile, given by $\text{Im}(\sigma_{32}e^{i\varphi})$, as well as the populations of both dressed and bare states are asymmetric functions of the detuning (Fig. 6). But contrary to the case of lower-level splitting, where spontaneous relaxation of the rate \bar{a}_{12} at the dressed transition $|1\rangle \rightarrow |\tilde{2}\rangle$ [Fig. 1(a)] leads to a depletion of state $|1\rangle$ and a population of state $|2\rangle$, for the scheme with upper level splitting spontaneous relaxation of the rate \bar{a}_{31} at the dressed transition $|\tilde{3}\rangle\text{-}|1\rangle$ [Fig. 1(c)] depletes level $|\tilde{3}\rangle$ and populates level $|1\rangle$. In the limit of sufficiently large negative detuning, $|\delta'| \gg |\beta| \gg \sqrt{\omega_{21}^3/|\delta'|}$, ($\delta' = -\delta$), this means that most of the atoms can be trapped in the upper atomic state: $\rho_{11} \approx 1$ [Figs. 6(c) and 6(d)]. This provides a quite unexpected method for producing fully inverted three-level atoms at the $|3\rangle\text{-}|1\rangle$ transition by driving them at the lower frequency transition $|3\rangle\text{-}|2\rangle$. This result is unexpected from the traditional point of view. Indeed, according to the traditional master equations, level $|1\rangle$ is not coupled to the field, independently of its intensity and detuning, and hence level $|1\rangle$ should remain empty. The process which leads to pumping of the upper state is a two-photon Stokes scattering of the driving field [Fig. 4(b)]. Under the condition $|\beta|^2|\delta'| \gg \omega_{12}^3$ this process prevails over spontaneous decay from the upper state (since the rate \bar{a}_{31} exceeds the rates of all other transitions), and hence leads to a trapping of atoms

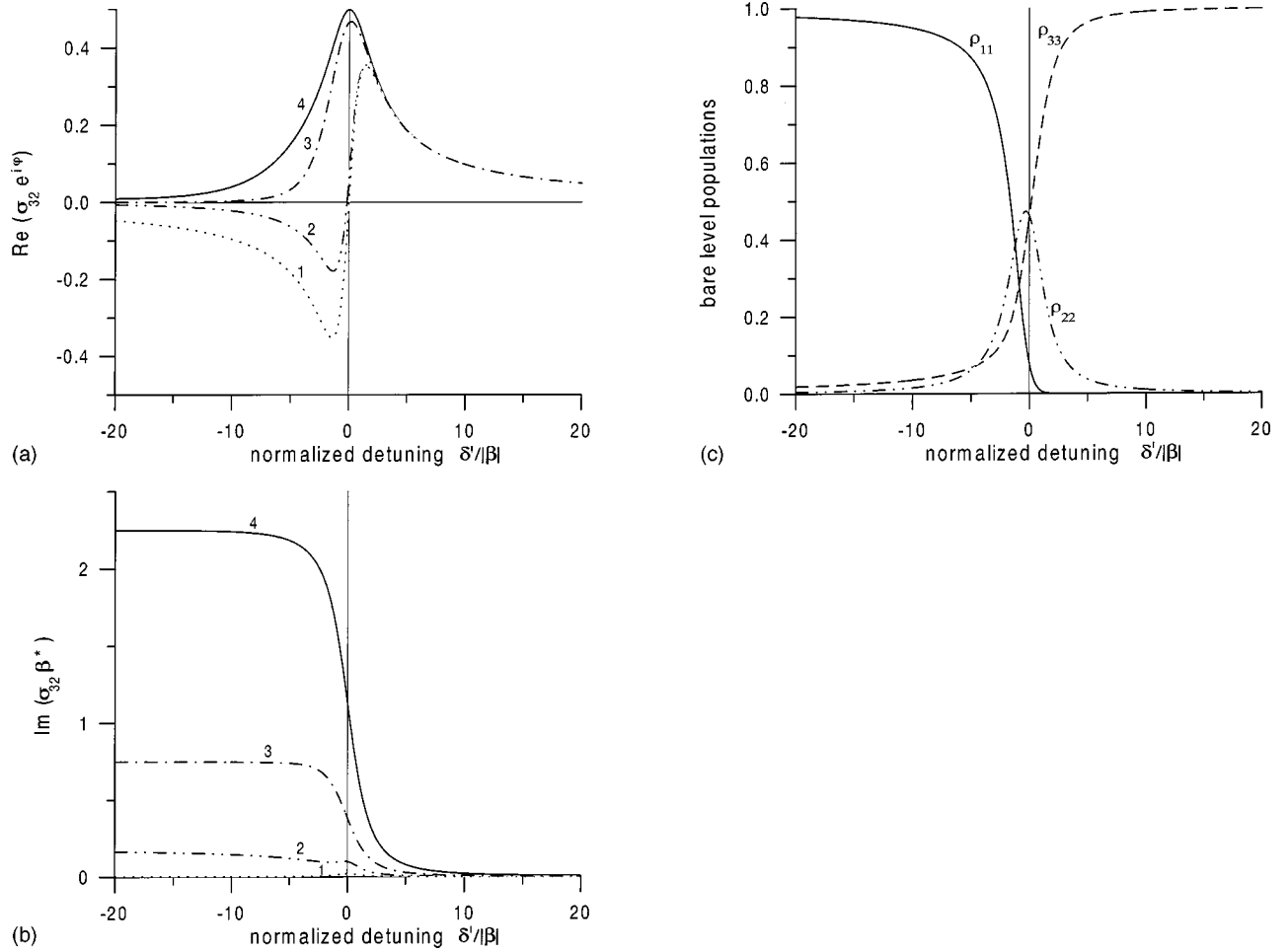


FIG. 6. (a) $\text{Re}(\sigma_{32}e^{-i\varphi})$, which is proportional to the real part of the susceptibility; (b) $\text{Im}(\sigma_{32}\beta^*)$, which is proportional to the absorption power; (c) populations ρ_{ii} of the bare levels; (d) populations $\tilde{\rho}_{ii}$ of the dressed levels for the scheme of the upper-level splitting [Fig. 1(c)] vs the driving field detuning $\delta' = -\delta$ in units of the resonant Rabi frequency $|\beta|$ for $A_{12}/\omega_{12} = 10^{-6}$, $A_{13}/\omega_{12} = A_{23}/\omega_{12} = 10^{-2}$, $\omega_{13}/\omega_{12} = \omega_{23}/\omega_{12} = 10^5$, $\eta(\omega) = 1$, and $N(\omega) = 0$. (1) $|\beta|/\omega_{12} = 3$. (2) $|\beta|/\omega_{12} = 15$. (3) $|\beta|/\omega_{12} = 60$. (4) $|\beta|/\omega_{12} = 180$ for (a) and (b) and $|\beta|/\omega_{12} = 30$ for (c) and (d).

into the state $|1\rangle$. The traditional result follows from our treatment only if $|\beta|^2|\delta'| \ll \omega_{12}^3$.

IX. CONCLUSIONS

We have shown that coherent driving at one of three atomic transitions in a three-level system can strongly influence relaxation processes. Drastic modifications appear in the case of crossing between one of the ac Stark levels and a nearby unperturbed atomic level. Such crossing results in a reversal of the direction of spontaneous emission between these levels. Modification of the relaxation scheme induces a very unusual atomic response compared to that which would be obtained on the basis of the traditional master equations (where the decay rates are assumed to be constants independent of driving field).

Level crossing occurs when the generalized Rabi frequency of the driving field exceeds the frequency of the adjacent transition. Negative detuning favors such crossing, and allows for the observation of some interesting effects even at a low intensity of the driving field. For each value of the driving intensity there is an optimal value of the detuning which provides the most effective absorption of energy from

the field, and allows one to achieve a large population inversion at the driven atomic transition. For a scheme with lower-level splitting [Fig. 1(a)], in the case of a large negative detuning, all atoms can be trapped into the upper ac Stark level due to two-photon Raman process. For a scheme with upper-level splitting [Fig. 1(c)], the two-photon Raman scattering can provide a full population inversion at the highest frequency transition.

In the vicinity of the resonance ($\delta \cong 0$) atoms can be trapped into the lower ac Stark level $\tilde{\rho}_{22} \cong 1$. Such a trapping results in the excitation of the maximal atomic coherence $\sigma_{32} \cong \sqrt{\rho_{33}\rho_{22}}$, the maximal value of $\text{Re}(\sigma_{32}e^{i\varphi})$, and a maximal ratio $\text{Re}(\sigma_{32}e^{i\varphi})/\text{Im}(\sigma_{32}e^{i\varphi})$. For the lower-level splitting schemes this also provides full depletion of the ground state and population inversion simultaneously at the $|2\rangle$ - $|1\rangle$ and $|3\rangle$ - $|1\rangle$ bare-state transitions.

The redistribution of the dressed populations leads, in turn, to drastic modifications of both the fluorescence and probe field absorption spectra. In particular, at a certain domain of detuning of the driving field, a weak-field probing $|1\rangle$ - $|3\rangle$ transition can be amplified in the absence of population inversion at this transition in the vicinity of either one or

both of the dressed transitions. Hence field-dependent relaxation provides an interesting mechanism for amplification without inversion.

All the field-dependent relaxation effects discussed above were considered within the simplest model of a three-level atomic system with nondegenerate levels coupled to the field reservoir when a strong monochromatic field drives only one atomic transition. For an experimental realization of this simplest model in the case of Figs. 1(a) and 1(b), it would be appropriate to use the vapors of alkali metals driven on a $D1$ transition, and placed in a constant magnetic field.

For example, for potassium (^{39}K) vapor in a magnetic field, sublevels with $M_J = \pm 1/2$ of its ground state $4s\ ^2S_{1/2}$ and one of the levels with $M_J = -1/2$ or $M_J = +1/2$ of the first excited state $4p\ ^2P_{1/2}^0$ form a three-level configuration corresponding to Figs. 1(a) and 1(b) with optical frequencies $\omega_{31} \approx \omega_{32} \sim 2.45 \times 10^{15}\ \text{s}^{-1}$. When the magnetic-field strength essentially exceeds some critical value $H \sim 170\ \text{G}$, the frequency ω_{21} formed by this magnetic field sufficiently exceeds hyperfine splitting $\omega_h \sim 2.9 \times 10^9\ \text{s}^{-1}$ of the ground state $4s\ ^2S_{1/2}$. Hence one can neglect the hyperfine structure of atomic levels. Radiation of a Ti:sapphire or dye laser which has an appropriate frequency and a sufficiently high power can be used as the driving field. Due to resonant tuning of the driving field to the $D1$ transition, it is possible to neglect the interaction of the field with the $4p\ ^2P_{3/2}^0$ atomic state nearest to level $|3\rangle$. Left circular polarization of the driving field provides its interaction only with the $|3\rangle$ - $|2\rangle$ transition corresponding to Fig. 1(a) (where level $|3\rangle$ has $M_J = -1/2$), whereas a right circularly polarized field interacts only with the $|3\rangle$ - $|1\rangle$ transition [Fig. 1(b)] (where the level $|3\rangle$ has $M_J = +1/2$).

Because of the small value of the magnetic dipole moment of the $|1\rangle$ - $|2\rangle$ transition with respect to values of electric dipole moments of optical transitions ($|\mu_{21}| \sim 1.3 \times 10^{-20}\ \text{cgs}$, $|\mu_{31}| \sim |\mu_{32}| \sim 1.6 \times 10^{-17}\ \text{cgs}$) it is meaningful to look for modifications of the fluorescence spectrum or probe field absorption which have different polarizations at different transitions. For instance, for the scheme of Fig. 1(a), the fluorescence at rf transition $|1\rangle$ - $|2\rangle$ of weakly driven atoms (the lower dynamic Stark level is above the ground state) is right circularly polarized along the magnetic field. However, as soon as the lower dynamic Stark level crosses the ground state (for the field resonant to the $|3\rangle$ - $|2\rangle$ transition, it corresponds to intensity $I_\beta \sim 400\ \text{W}/\text{sm}^2$), rf radiation at the $|\tilde{1}\rangle$ - $|\tilde{2}\rangle$ transition with left circular polarization should appear as a direct experimental evidence of field-induced level crossing and spontaneous relaxation from the ground atomic state. For example, for potassium vapor of volume $V \sim 1\ \text{cm}^3$ under a temperature $T \sim 10^3\ \text{K}$ and pressure $p \sim 1\ \text{torr}$ (concentration $n \sim 10^{16}\ \text{sm}^{-3}$) placed in magnetic field $H \sim 500\ \text{G}$ and driven by a field of intensity $I_\beta \sim 160\ \text{kW}/\text{sm}^2$ resonant to the $D1$ line, one can expect fluorescence with a power $P_f \sim 4 \times 10^{-15}\ \text{W}$ at a wavelength $\lambda \sim 1\ \text{sm}$.

A probe field with an appropriate polarization can test either optical transitions or the rf transition $|1\rangle$ - $|2\rangle$. When a rf probe field is left circularly polarized and is directed along the magnetic field, no interaction with atoms in the scheme of Fig. 1(a) occurs until the driving field provides a level

crossing. As soon as such a crossing takes place, the rf field is amplified (without population inversion in a bare atomic system) due to population inversion at the dressed transition $|\tilde{1}\rangle$ - $|\tilde{2}\rangle$. Under the conditions described above, one can expect a probe field gain $G_\alpha \sim 10^{-3}\ \text{sm}^{-1}$. Placing the system into a resonator with $Q \sim 10^3$ and length $L \sim 1.5\ \text{cm}$ provides a predominance of amplification over losses, and a realization of stimulated emission at the rf transition.

Changing the intensity or frequency of the driving field as well as the magnetic-field strength leads to a change of the intensity and frequency of both the fluorescence emission and generated rf field. For example, an increase of the driving field frequency corresponding to detuning from the $D1$ line on the value $\delta \sim 160\ \text{GHz}$ allows one to obtain fluorescence with a power $P_f \sim 4 \times 10^{-15}\ \text{W}$ at a wavelength $\lambda \sim 2\ \text{mm}$ with a driving field intensity $I_\beta \sim 15\ \text{kW}/\text{sm}^2$. We would like to stress that observation of the described rf effects does not require a low temperature of the reservoir.

Atoms with a ground state 1S_0 and nuclear spin $I=0$ could be chosen for an experimental realization of the schemes of Figs. 1(c) and 1(d). For example, for the case of barium (^{138}Ba), the ground state $6s\ ^1S_0$ and magnetic sublevels with $M_F = -1$ and $M_F = 0$ of the excited state $6p\ ^1P_1^0$ form a three-level configuration corresponding to Fig. 1(c) if the driving field has a left circular polarization. The ground state and magnetic sublevels with $M_F = 0$ and $M_F = +1$ of the excited state with a right circularly polarized field form a three-level configuration corresponding to Fig. 1(d). Radiation of the dye laser can be used as a driving field resonant to the $6s\ ^1S_0$ - $6p\ ^1P_1^0$ transition with a wavelength $\lambda_0 \approx 553.6\ \text{nm}$ and a dipole moment $\mu_0 \sim 1.4 \times 10^{-17}\ \text{cgs}$. Experimental conditions similar to conditions for potassium lead to rf fluorescence and amplification of the rf probe field, which are of the same order as described above.

Since barium atoms have no levels close to the $6p\ ^1P_1^0$ state, one can also drive them by radiation slightly detuned from optical resonance, for instance, by radiation of a Nd:YAG (yttrium aluminum garnet) laser at wavelength $\lambda \approx 540\ \text{nm}$. In this case barium vapor of volume $V \sim 1\ \text{cm}^3$ under a temperature $T \sim 10^3\ \text{K}$ and pressure $p \sim 1\ \text{torr}$ driven by a left circularly polarized field of intensity $I_\beta \sim 50\ \text{kW}/\text{sm}^2$ can produce a left circularly polarized rf emission corresponding to relaxation at the dressed transition $|\tilde{3}\rangle$ - $|\tilde{1}\rangle$ [Fig. 1(c)], with power $P_f \sim 3 \times 10^{-15}\ \text{W}$ at a wavelength $\lambda \sim 20\ \mu\text{m}$.

Finally, let us emphasize that we studied field-dependent relaxation effects in the simplest possible model allowing for both an analytical solution and a clear physical interpretation. At the same time, a crossing of the dynamic Stark levels with some neighboring unperturbed energy states can occur under interaction of the driving field with different multilevel quantum systems coupled to different kinds of reservoirs. Interesting examples of this include interaction of a strong laser field with vibrational-rotational transitions of molecules and semiconductor structures having appropriate energy spectra. The quantitative analysis of these physical situations requires a generalization of the above model.

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APPENDIX A: RELAXATION RATES OF THE DRIVEN THREE-LEVEL ATOM

The elements of the relaxation supermatrix in the set of equations (8) are calculated according to Eqs. (2)–(4), (10), and (11). They are grouped into two sets. The first one is as follows:

$$\begin{aligned}
 R_{1111} &= -(R_{2211} + R_{3311}), \\
 R_{1122} &= c^2 w_{21}(\tilde{\omega}_{12}) + s^2 w_{21}(\tilde{\omega}_{13}), \\
 R_{1133} &= c^2 w_{31}(\tilde{\omega}_{13} - \omega) + s^2 w_{31}(\tilde{\omega}_{12} - \omega), \\
 R_{2211} &= c^2 w_{12}(\tilde{\omega}_{21}) + s^2 w_{12}(\tilde{\omega}_{31}), \\
 R_{2222} &= -(R_{3322} + R_{1122}), \\
 R_{2233} &= c^4 w_{32}(\tilde{\omega}_{23} - \omega) + s^4 w_{32}(\tilde{\omega}_{32} - \omega) + 2s^2 c^2 w_{32}(-\omega), \\
 R_{3311} &= c^2 w_{13}(\tilde{\omega}_{31} + \omega) + s^2 w_{13}(\tilde{\omega}_{21} + \omega), \\
 R_{3322} &= c^4 w_{23}(\tilde{\omega}_{32} + \omega) + s^4 w_{23}(\tilde{\omega}_{23} + \omega) + 2s^2 c^2 w_{23}(\omega), \\
 R_{3333} &= -(R_{1133} + R_{2233}), \\
 R_{1132} &= e^{i\varphi} \frac{SC}{2} [w_{31}(\tilde{\omega}_{12} - \omega) - w_{31}(\tilde{\omega}_{13} - \omega) + w_{21}(\tilde{\omega}_{12}) - w_{21}(\tilde{\omega}_{13})], \\
 R_{2232} &= e^{i\varphi} \frac{SC}{2} \{ (c^2 - s^2) [w_{32}(-\omega) + w_{23}(\omega)] + s^2 [w_{32}(\tilde{\omega}_{32} - \omega) + w_{23}(\tilde{\omega}_{23} + \omega)] \\
 &\quad - c^2 [w_{32}(\tilde{\omega}_{23} - \omega) + w_{23}(\tilde{\omega}_{32} + \omega)] + w_{21}(\tilde{\omega}_{13}) - w_{21}(\tilde{\omega}_{12}) \}, \\
 R_{3332} &= e^{i\varphi} \frac{SC}{2} \{ (s^2 - c^2) [w_{23}(\omega) + w_{32}(-\omega)] + c^2 [w_{32}(\tilde{\omega}_{23} - \omega) + w_{23}(\tilde{\omega}_{32} + \omega)] \\
 &\quad - s^2 [w_{32}(\tilde{\omega}_{32} - \omega) + w_{23}(\tilde{\omega}_{23} + \omega)] - w_{31}(\tilde{\omega}_{12} - \omega) + w_{31}(\tilde{\omega}_{13} - \omega) \}, \\
 R_{3211} &= e^{-i\varphi} \frac{SC}{2} [w_{13}(\tilde{\omega}_{21} + \omega) - w_{13}(\tilde{\omega}_{31} + \omega) + w_{12}(\tilde{\omega}_{21}) - w_{12}(\tilde{\omega}_{31})], \\
 R_{3222} &= e^{-i\varphi} \frac{SC}{2} \{ (c^2 - s^2) [w_{23}(\omega) - w_{32}(-\omega)] + s^2 [w_{23}(\tilde{\omega}_{23} + \omega) - w_{32}(\tilde{\omega}_{32} - \omega)] \\
 &\quad - c^2 [w_{23}(\tilde{\omega}_{32} + \omega) - w_{32}(\tilde{\omega}_{23} - \omega)] + w_{31}(\tilde{\omega}_{13} - \omega) - w_{31}(\tilde{\omega}_{12} - \omega) \}, \\
 R_{3233} &= e^{-i\varphi} \frac{SC}{2} \{ (s^2 - c^2) [w_{32}(-\omega) - w_{23}(\omega)] + s^2 [w_{23}(\tilde{\omega}_{23} + \omega) - w_{32}(\tilde{\omega}_{32} - \omega)] \\
 &\quad + c^2 [w_{32}(\tilde{\omega}_{23} - \omega) - w_{23}(\tilde{\omega}_{32} + \omega)] + w_{21}(\tilde{\omega}_{13}) - w_{21}(\tilde{\omega}_{12}) \},
 \end{aligned} \tag{A1}$$

$$\begin{aligned}
R_{3232} &= (R_{2222} + R_{3333})/2, \\
R_{3223} &= 0, \\
R_{3131} &= (R_{1111} + R_{3333})/2, \\
R_{3113} &= 0, \\
R_{3121} &= -e^{-i\varphi}[(c^3s - s^3c)w_{32}(-\omega) - c^3sw_{32}(\tilde{\omega}_{23} - \omega) + s^3cw_{32}(\tilde{\omega}_{32} - \omega)]/2, \\
R_{3112} &= sc e^{-i\varphi}[w_{2121}(\tilde{\omega}_{21}) - w_{2121}(\tilde{\omega}_{31})], \\
R_{2121} &= (R_{1111} + R_{2222})/2, \\
R_{2112} &= c^2w_{2121}(\tilde{\omega}_{21}) + s^2w_{2121}(\tilde{\omega}_{31}) + c^2w_{1212}^*(\tilde{\omega}_{12}) + s^2w_{1212}^*(\tilde{\omega}_{13}), \\
R_{2131} &= e^{i\varphi} \frac{sc}{2} [(c^2 - s^2)w_{23}(\omega) + s^2w_{23}(\tilde{\omega}_{23} + \omega) - c^2w_{23}(\tilde{\omega}_{32} + \omega) + w_{21}(\tilde{\omega}_{13}) - w_{21}(\tilde{\omega}_{12})], \\
R_{2113} &= e^{-i\varphi} sc [w_{1212}^*(\tilde{\omega}_{12}) - w_{1212}^*(\tilde{\omega}_{13})].
\end{aligned}$$

For the second set, one obtains

$$\begin{aligned}
R_{1121} &= -2s^2c^2w_{1332}(\omega) - s^4w_{1332}(\tilde{\omega}_{23} + \omega) - c^4w_{1332}(\tilde{\omega}_{32} + \omega), \\
R_{1131} &= e^{i\varphi}[(c^3s - s^3c)w_{1332}(\omega) - c^3sw_{1332}(\tilde{\omega}_{32} + \omega) + s^3cw_{1332}(\tilde{\omega}_{23} + \omega)], \\
R_{2221} &= -s^2w_{2331}^*(\tilde{\omega}_{21} + \omega) - c^2w_{2331}^*(\tilde{\omega}_{31} + \omega), \\
R_{2231} &= 0, \\
R_{3321} &= 2s^2c^2w_{1332}(\omega) + s^4w_{1332}(\tilde{\omega}_{23} + \omega) + c^4w_{1332}(\tilde{\omega}_{32} + \omega), \\
R_{3331} &= -e^{i\varphi}[(c^3s - s^3c)w_{1332}(\omega) - c^3sw_{1332}(\tilde{\omega}_{32} + \omega) + s^3cw_{1332}(\tilde{\omega}_{23} + \omega)], \\
R_{3221} &= sc e^{-i\varphi}[w_{2331}^*(\tilde{\omega}_{21} + \omega) - w_{2331}^*(\tilde{\omega}_{31} + \omega) + c^2w_{2321}^*(\tilde{\omega}_{21}) + s^2w_{2321}^*(\tilde{\omega}_{31})], \\
R_{3212} &= e^{-i\varphi}[(c^3s - s^3c)w_{1332}^*(\omega) - c^3sw_{1332}^*(\tilde{\omega}_{32} + \omega) + s^3cw_{1332}^*(\tilde{\omega}_{23} + \omega)], \\
R_{3231} &= -s^2w_{2331}^*(\tilde{\omega}_{21} + \omega) - c^2w_{2331}^*(\tilde{\omega}_{31} + \omega), \\
R_{3213} &= e^{-i2\varphi} s^2 c^2 [2w_{1332}^*(\omega) - w_{1332}^*(\tilde{\omega}_{23} + \omega) - w_{1332}^*(\tilde{\omega}_{32} + \omega)], \\
R_{3111} &= 0, \\
R_{3122} &= 0, \\
R_{3133} &= e^{-i\varphi} \{ (c^3s - s^3c)[w_{1332}^*(\omega) - w_{3123}(-\omega)] - c^3s[w_{1332}^*(\tilde{\omega}_{32} + \omega) - w_{3123}(\tilde{\omega}_{23} - \omega)] \\
&\quad + s^3c[w_{1332}^*(\tilde{\omega}_{23} + \omega) - w_{3123}(\tilde{\omega}_{32} - \omega)] \}, \\
R_{3132} &= -2s^2c^2w_{1332}^*(\omega) - s^4w_{1332}^*(\tilde{\omega}_{23} + \omega) - c^4w_{1332}^*(\tilde{\omega}_{32} + \omega), \\
R_{3123} &= e^{-i2\varphi} s^2 c^2 [2w_{3123}(-\omega) - w_{3123}(\tilde{\omega}_{23} - \omega) - w_{3123}(\tilde{\omega}_{32} - \omega)], \\
R_{2111} &= -s^2w_{2331}(\tilde{\omega}_{21} + \omega) - c^2w_{2331}(\tilde{\omega}_{31} + \omega), \\
R_{2122} &= -2s^2c^2w_{1332}^*(\omega) - s^4w_{1332}^*(\tilde{\omega}_{23} + \omega) - c^4w_{1332}^*(\tilde{\omega}_{32} + \omega),
\end{aligned} \tag{A2}$$

$$R_{2133} = 2s^2c^2w_{3123}(-\omega) + c^4w_{3123}(\tilde{\omega}_{23} - \omega) + s^4w_{3123}(\tilde{\omega}_{32} - \omega) + s^2w_{3213}^*(\tilde{\omega}_{12} - \omega) + c^2w_{3213}^*(\tilde{\omega}_{13} - \omega),$$

$$R_{2132} = sce^{i\varphi}[w_{3213}^*(\tilde{\omega}_{12} - \omega) - w_{3213}^*(\tilde{\omega}_{13} - \omega)],$$

$$R_{2123} = e^{-i\varphi}\{(c^3s - s^3c)[w_{3123}(-\omega) + w_{1332}^*(\omega)] - c^3s[w_{1332}^*(\tilde{\omega}_{32} + \omega) + w_{3123}(\tilde{\omega}_{23} - \omega)] + s^3c[w_{3123}(\tilde{\omega}_{32} - \omega) + w_{1332}^*(\tilde{\omega}_{23} + \omega)]\}.$$

APPENDIX B: COEFFICIENTS OF THE GENERAL SOLUTION (45)

$$A_1 = \alpha_1 + \beta_1 \delta |\beta| - 4|\beta|^2 / \tilde{W},$$

$$A_2 = \alpha_2 - \beta_2 \delta |\beta| + 4|\beta|^2 / \tilde{W},$$

$$A_3 = \alpha_3 + \beta_3 \delta |\beta| + 4|\beta|^2 / \tilde{W},$$

$$A_4 = \alpha_4 - \beta_4 \delta |\beta| - 4|\beta|^2 / \tilde{W},$$

$$C_1 = a_{12} + (a_2 - a_{13})(a_1 - \tilde{a}_{32}) / \tilde{W} + \eta \delta |\beta|,$$

$$C_2 = w_{13} + (a_2 - a_{13})(a_{31} + \tilde{a}_{32}) / \tilde{W} - \eta \delta |\beta|,$$

$$\eta = 4(a_2 - a_{13}) / (W\tilde{W}),$$

$$\alpha_1 = (a_1 - \tilde{a}_{32})(\tilde{a}_{31} + a_{32}^0 - a_2) / \tilde{W} - a_{21} - a_{12} - w_{23},$$

$$\beta_1 = 4(a_1 - a_2 + \tilde{a}_{31} - 2a_{23}) / (W\tilde{W}),$$

$$\alpha_2 = (a_{31} + \tilde{a}_{32})(\tilde{a}_{31} + a_{32}^0 - a_2) / \tilde{W} + w_{23} - w_{13},$$

$$\beta_2 = 4(a_{13} - 2a_{23} - a_2) / (W\tilde{W}),$$

$$\alpha_3 = (a_1 - \tilde{a}_{32})(a_1 - a_2 + a_{13} + a_{32}^0) / \tilde{W} + w_{32} - a_{12},$$

$$\beta_3 = 4(2a_{32} + a_{13} - a_2) / (W\tilde{W}),$$

$$\alpha_4 = (a_{31} + \tilde{a}_{32})(a_1 - a_2 + a_{13} + a_{32}^0) / \tilde{W} - w_{31} - w_{13} - w_{32},$$

$$\beta_4 = 4(\tilde{a}_{31} + 2a_{32} + a_1 - a_2) / (W\tilde{W}),$$

$$\tilde{W} = W(1 + 4\delta^2 / W^2),$$

$$W = 2\Gamma_{32}.$$

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