# Generalization of the Maxwell-Bloch equations to the case of strong atom-field coupling

Olga Kocharovskaya,[1,2,3] Shi-Yao Zhu,[1,4,5] Marlan O. Scully,[1,4] Paul Mandel,[2] and Y. V. Radeonychev[3]
[1]*Department of Physics, Texas A&M University, College Station, Texas 77843*
[2]*Université Libre de Bruxelles, Optique Nonliniaire Théorique, Campus Plaine CP 231, 1050 Bruxelles, Belgium*
[3]*Institute of Applied Physics, Russian Academy of Science, 603600 Nizhny Novgorod, Russia*
[4]*Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany*
[5]*Department of Physics, Hong Kong Baptist College, Hong Kong*
(Received 23 August 1993)

We derive the generalized set of Maxwell-Bloch equations which takes into account the dependence of relaxation coefficients on the amplitude and frequency of the coherent field. This has interesting implications in many problems in quantum optics and laser physics, e.g., the problem of lasing without inversion due to a strong coherent generating field.

PACS number(s): 42.50.−p

## I. INTRODUCTION

The Maxwell-Bloch equations are the basis for the analysis of many processes and problems in quantum optics and laser dynamics. As is well known, the interaction of atoms with a reservoir is introduced into this set of equations [1] of the density matrix

$$\frac{d}{dt}\rho^{(0)}_{m'm}+i\omega_{m'm}\rho^{(0)}_{m'm}+\frac{i}{\hbar}[V(t),\rho^{(0)}]_{m'm}=\sum_{n',n}R^{(0)}_{m'mn'n}\rho^{(0)}_{n'n} \quad (1)$$

phenomenologically via the supermatrix of relaxation constants $R^{(0)}$, which has the simplest form in the secular approximation

$$\sum_{n',n}R^{(0)}_{m'mn'n}\rho^{(0)}_{n'n}=\begin{cases}\sum_{k\neq m}(w_{km}\rho^{(0)}_{kk}-w_{mk}\rho^{(0)}_{mm})\ , & m=m'\\ -\gamma_{m'm}\rho_{m'm}\ , & m\neq m'\ .\end{cases} \quad (2)$$

Here $\omega_{m'm}$ is the transition frequency between atomic level $m'$ and $m$, $V(t)$ is the interaction Hamiltonian between atoms and field, $w_{mk}$ is a relaxation rate from level $m$ to level $k$, and $\gamma_{m'm}$ is a linewidth. Conversely, if we take into account that the master equation for an atomic system in the Born and Markovian approximations has indeed the form (1), in the absence of any coherent field we can say with the same success that it is the coupling with the coherent field which was introduced into the master equation for atomic system as a dynamical term. In other words, the influence of the coherent field on the interaction of the atoms with the reservoir is completely ignored by Eq. (1). (We use the index zero in these equations to note that they are only the zeroth-order approximation with respect to the dependence of the relaxation coefficients on field amplitude.) As a matter of fact, such an influence is there and its study has a long history rising from the earlier works of Redfield [2] and Argyres and Kelly [3] on the relaxation of spin systems in nuclear magnetic resonance. Many later papers have also been devoted to the influence of the strong field on the relaxation process in optics [4–13].

However, the appearance of the relaxation coefficients dependence on the amplitude and frequency of the field has been connected mainly with the violation of the Born and Markovian approximations. In this sense, Eq. (1) seems to be intrinsically consistent.

Here we show that even in the Born and Markovian approximation the coherent field influences the relaxation and incoherent pumping processes in atomic systems. As a result, the structure of the relaxation supermatrix under the action of the coherent field is essentially changed as compared to Eqs. (1) and (2). Even in the *secular approximation* there appear cross-relaxation terms (diagonal elements of a density matrix in the right-hand side of the equations for off-diagonal elements and vice versa).

In Sec. II we establish the general connection between the relaxation supermatrices of the multilevel atomic system in the absence of any field and under the action of the multifrequency resonant coherent field; this allows for the possibility of obtaining an exact generalized master equation in the presence of the field. As an illustration of the general approach we consider in Secs. III and IV the particular cases of the two-level and three-level systems driven by the resonant coherent field and interacting with the noncoherent field reservoir.

## II. GENERAL APPROACH

Let us consider $M$-level atoms which are described by the Hamiltonian $H_a$ and interact with the reservoir $H_r$ via $H_{\text{int}}$. As is well known, in the Born and Markovian approximations, and in the absence of any coherent field, the behavior of these atoms is described by the master equation (1) with

 

$$R^{(0)}_{m'mn'n}=\Gamma^{(0)}_{nmm'n'}+\Gamma^{(0)*}_{n'm'mn}-\sum_k(\delta_{mn}\Gamma^{(0)}_{m'kkn'}+\delta_{m'n'}\Gamma^{(0)*}_{mkkn}) , \quad (3)$$

$$\Gamma^{(0)}_{nmm'n'}(\omega_{m'n'})=\frac{1}{\hbar^2}\int_0^\infty dt' e^{-i\omega_{m'n'}t'}\mathrm{Tr}_r\left\{\langle\psi_n|H_{\rm int}|\psi_m\rangle\exp\left[\frac{-iH_r t'}{\hbar}\right]\langle\psi_{m'}|H_{\rm int}|\psi_{n'}\rangle\exp\left[\frac{iH_r t'}{\hbar}\right]\rho_r(0)\right\} , \quad (4)$$

where the basis of bare atomic states $|\psi_n\rangle$ has been used:

$$H_a\psi_k=E_k\psi_k , \quad \omega_{ln}=(E_l-E_n)/\hbar$$

and the trace has been taken over the reservoir variables [$\rho_r(0)$ is the density matrix of the reservoir in the absence of interaction with the atoms]. Now, let us suppose that apart from the interaction with the reservoir the atoms are driven by the resonant $M-1$ frequency field. Then the Hamiltonian of our dynamical system (atomic system driven by the field) depends on time via $H_d(t)=H_a+V(t)$. The straightforward generalization of the traditional method of derivation of the master equation in the Born and Markovian approximations for this case [1] leads to the same form of the master equation

$$\frac{d\rho_{m'm}}{dt}+i\omega_{m'm}\rho_{m'm}+\frac{i}{\hbar}[V(t),\rho]_{m'm}=\sum_{n',n}R_{m'mn'n}(t)\rho_{n'n} , \quad (5)$$

where the supermatrix $R$ is expressed via $\Gamma_{nmn'm'}$ by the same way as $R^{(0)}$ is expressed via $\Gamma^{(0)}_{nmn'm'}$ in Eq. (3) and

$$\Gamma_{nmm'n'}(t)=\frac{1}{\hbar^2}\int_0^\infty dt'\mathrm{Tr}_r\{\langle\psi_n|H_{\rm int}|\psi_m\rangle\times\langle\psi_{m'}|H^I_{\rm int}(t-t',t)|\psi_{n'}\rangle\times\rho_r(0)\} , \quad (6)$$

$$H^I_{\rm int}(t-t',t)=U_d(t,0)U_d^{-1}(t-t',0)\exp\left[\frac{-iH_r t'}{\hbar}\right]\times H_{\rm int}\exp\left[\frac{+iH_r t'}{\hbar}\right]\times U_d(t-t',0)U_d^{-1}(t,0) . \quad (7)$$

Thus the only difference, as compared with the case $V=0$, is the coupling Hamiltonian with the reservoir in the interaction picture $H^I_{\rm int}(t-t',t)$, which is defined in general by the evolution operator of the dynamical system $U_d(t,0)=T\exp\{[-i\int_0^t H_d(t')dt'/\hbar]\}$, where $T$ is a chronological operator, ordering the products of successive operators in accordance with the decreasing of the time from the left to the right. In the case $V=0$ the evolution operator takes a simple form $U_d(t,0)=\exp[(-iH_a t/\hbar)]$ and, as a result, Eq. (6) becomes Eq. (4). In order to express $R$ via $\Gamma$ we use the lemma of "effective evolution" which allows us to reduce the evolution operator $U_d(t,0)$ acting on any hermitian operator $B$ to the successive operation of $\exp[(i\tilde{H}_d t/\hbar)]$ and $U(t)$:

$$U_d(t,0)BU_d^{-1}(t,0)=U(t)\exp\left[-\frac{i}{\hbar}\tilde{H}_d(t-t_0)\right]\times B\exp\left[\frac{i}{\hbar}\tilde{H}_d(t-t_0)\right]U^{-1}(t) . \quad (8)$$

Here $U(t)$ is a unitary operator of transformation to the basis of rotating states which transforms the Hamiltonian of the dynamical system $H_d(kt)$ to the time independent one: $\tilde{H}_d=U^{-1}(t)[H_d(t)-i\hbar(dU/dt)U^{-1}]U(t)$ with $U(t=0)=1$. Applying twice the lemma (8) to (7) we have

$$H^I_{\rm int}-U(t)e^{-(i/\hbar)(\tilde{H}_d+H_r)t'}U^{-1}(t-t')\times H_{\rm int}U(t-t')e^{(i/\hbar)(\tilde{H}_d+H_r)t'}U^{-1}(t) . \quad (9)$$

For simplicity we restrict ourselves to the rotating-wave approximation (RWA), i.e., use an effective interaction Hamiltonian

$$V(t)=-\tfrac{1}{2}\sum_n[\mathcal{E}_n\exp(-i\Omega_n t)\mu_{g(n),f(n)}|\psi_{g(n)}\rangle\times\langle\psi_{f(n)}|+\mathrm{H.c.}] ,$$

where $\mathcal{E}_n$ is a complex amplitude of the $n$th component of the field and $\mu_{g(n),f(n)}$ is a dipole moment at the transition $g(n)$ to $f(n)$. This approximation, as is well known [1], implies small values of the Rabi frequency compared with the corresponding atomic frequency. We consider the situation when at least one in each couple of levels either does not interact with the field at all or interacts only with one component of the field. [The integer functions $f(n)$ and $g(n)$ enumerate the lower and upper levels of the transition coupled to the $n$the component of the field correspondingly.] In this case the unitary transformation matrix $U(t)$ is a diagonal one:

$$U_j^m(t)=\delta_{mj}\exp(iS_j\Omega_j t/\hbar) , \quad (10)$$

where $S_j=1$ if level $j$ interacts only with the $n$th monochromatic component of the field and $j=f(n)$; $S_j=-1$ if level $j$ interacts only with the $n$th monochromatic component of the field and $j=g(n)$ and $S_{f(n)}=0$; it vanishes in all other cases. Substitution of (10) into (9) and in turn (9) into (6) leads to our main formula:

$$\Gamma_{nmm'n'}=\sum_{k,k',l,l'}\zeta^{k*}_{m'}\zeta^l_{n'}\zeta^k_{k'}\zeta^{l*}_{l'}\Gamma^{(0)}_{nmk'l'}(\tilde{\omega}_{kl}-S_{k'}\Omega_{k'}+S_{l'}\Omega_{l'})\times\exp[it(S_{m'}\Omega_{m'}-S_{n'}\Omega_{n'}-S_{k'}\Omega_{k'}+S_{l'}\Omega_{l'})] , \quad (11)$$

which expresses the new relaxation coefficients (in the presence of the coherent field) via the old ones (in the absence of coherent field) and via the matrix of transformation from the basis of depressed states to the basis of bare states $\zeta_i^j$:

$$\psi_i = \sum_j \zeta_i^j \tilde{\psi}_j \ , \quad \tilde{H}_a \tilde{\psi}_j = \tilde{E}_j \tilde{\psi}_j \ , \quad \tilde{\omega}_{ij} = (\tilde{E}_i - \tilde{E}_j)/\hbar \ . \tag{12}$$

Obviously, in the absence of the field we get the usual master equation since in the limit $\mathcal{E}_n \to 0$ the transformation matrix becomes the unit matrix.

The coherent field action not only incorporates the dynamical term $i[V(t),\rho]/\hbar$ to the Bloch equation as it was done usually, but also includes the action of the field on the structure of the relaxation supermatrix, i.e., introduces new nonzero elements. Moreover, all supermatrix elements become the functions of the amplitudes and frequencies of the field via the dressing frequency shift mentioned above transformation matrix $\zeta_i^j$. This dependence vanishes only in a very particular case when the relaxation rates at the all transitions do not depend on the frequencies of these transitions. In general this dependence appears via (a) the shift of the energy levels; (b) the mixing of the relaxation coefficients of different energy levels coupled by the external fields; and (c) the dependence of the efficiency of incoherent pumping process on the position of the quasienergy levels relative to the distribution of the energy levels of the reservoir (in particular, the density modes and the number of photons in these modes). This dependence can be especially important when we are interested in self-consistent lasing problems and hence have to add the equations for the field amplitudes

$$\dot{\mathcal{E}}_n + i(\Omega_n - \omega_n)\mathcal{E}_n + \frac{\mathcal{E}_n}{2T_c} = \frac{2\pi\omega_n \mu_{g(n),f(n)} N \sigma_n}{c_0} \tag{13}$$

to the set of equations (5). Here $\Omega_n$ and $T_c$ are the cavity resonant frequencies and relaxation time, $N$ is an atomic density, and $c_0$ is the light velocity. The changes in the master equation structures as well as the nonlinear dependence of the relaxation supermatrix $R$, even when weak (for example, if it is of the order of the ratio of the Rabi frequencies to the transition frequencies), introduces a new nonlinearity into the set of generalized Maxwell-Bloch equations (5) and (13), and therefore can substantially influence the nonlinear dynamics of systems under consideration.

## III. TWO-LEVEL SYSTEM

In order to illustrate the general results let us consider the case of a two-level system driven by a strong field with a Rabi frequency $\alpha$ and interacting with the field reservoir.

In the representation of the second quantization in the dipole and RWA approximation the interaction Hamiltonian is

$$H_{\text{int}} = -\sum_{\mathbf{k}} \sqrt{2\pi\hbar\omega_{\mathbf{k}}} \sum_{j,j'} [(\boldsymbol{\mu}_{jj'} \cdot \mathbf{e}_{\mathbf{k}}) | \psi_j \rangle \times \langle \psi_{j'} | A_{\mathbf{k}} b_{\mathbf{k}} + \text{H.c.}] \ , \tag{14}$$

where $\mu_{jj'}$ is a dipole momentum at the transition $j \to j'$; $j,j'=1,2$; $b_{\mathbf{k}}$ is an annihilation operator in the mode $\mathbf{k}$; $\mathbf{e}_{\mathbf{k}}$ is a unit polarization vector; and $A_{\mathbf{k}}$ is an amplitude of the $\omega_{\mathbf{k}}$ mode at the atom's position. Then in the Born and Markovian approximation, and in accordance with (4), we have, as is well known,

$$\Gamma^{(0)}_{mpln}(\omega_{ln}) = \frac{A_{mpln}}{2} \int_0^\infty \frac{\omega_k^3}{|\omega_{ln}|^3} |A_{\mathbf{k}}|^2 \eta(\omega_{\mathbf{k}}) \times \{ n(\omega_{\mathbf{k}}) \delta(\omega_{ln} - \omega_k) + [n(\omega_{\mathbf{k}}) + 1] \delta(\omega_{ln} + \omega_{\mathbf{k}}) \} \times d\omega_{\mathbf{k}} \ , \tag{15}$$

where $A_{mpln} = 4\pi\mu_{mp}\mu_{ln}|\omega_{ln}|^3/3\hbar c_0^3$, $A_{ln} = A_{nlln}$ is the Einstein coefficient of the spontaneous emission in the absence of the coherent field, $\eta(\omega_{ln}) = c_0^3 k^2(\omega_{ln})/\omega_{ln}^2(d\omega_k/dk)$ is a dimensionless parameter characterizing the distinction of the mode density of the reservoir from the vacuum (in vacuum $\eta(\omega_{ln})=1$), $n(\omega_k)$ is the average number of photons in the $\mathbf{k}$ mode of the reservoir, and $|A_{\mathbf{k}}|^2 = 1$ for running plane waves. In Eq. (15) the Lamb shift has been neglected. The transformation matrix to the basis of dressed states is

$$(\zeta_j^i) = \begin{bmatrix} c & se^{-i\phi} \\ -se^{i\phi} & c \end{bmatrix} , \quad c = \frac{\theta}{\sqrt{\theta^2 + |\alpha|^2}} \ ,$$
$$s = \frac{|\alpha| S}{\sqrt{\theta^2 + |\alpha|^2}} \ , \quad S = \text{sgn}\delta \ , \quad \alpha = |\alpha| e^{-i\phi} \ , \tag{16}$$
$$\theta = \tfrac{1}{2}(|\delta| + \sqrt{\delta^2 + 4|\alpha|^2}) \ , \quad \delta = \omega_{21} - \Omega \ .$$

If we substitute (15) and (16) into (11) and then (11) into (5) we immediately obtain the generalized set of the Bloch equations. For simplicity we write these equations in the secular approximation. This means that we keep on the right-hand side only the terms which give the same time dependence as we have on the left-hand side supposing that $\rho_{ln} = \sigma_{ln} e^{-i\omega_{ln} t}$, where $\sigma_{ln}$ are the slow varying functions of time in the scale $\omega_{ln}^{-1}$. In the absence of the coherent field, this approximation implies well-separated levels ($\omega_{m'm} \gg \gamma_{m'm}$) and leads to the simplest form (1) to (2) of the master equation because the relaxation supermatrix (3) or (4) does not depend on time. without this approximation, the cross-relaxation terms (i.e., off-diagonal density matrix elements $\rho_{ij}$ in the equations for populations $\rho_{ii}$ and vice versa) could appear in the master equation (1) and (5) even under the RWA. In the presence of the field, $R$ depends on time and as a result the structure of the Bloch equations is essentially changed. They are

$$
\begin{aligned}
&\dot{\sigma}_{21}+i\delta\sigma_{21}=-i\alpha(\rho_{22}-\rho_{11})+R_{2121}\sigma_{21}\\
&\qquad +R_{2112}\sigma_{12}+R_{2111}\rho_{11}+R_{2122}\rho_{22}\ ,\\
&\dot{\rho}_{11}=-2\,\mathrm{Im}(\sigma_{21}\alpha^*)+R_{1111}\rho_{11}\\
&\qquad +R_{1122}\rho_{22}+R_{1112}\sigma_{12}+R_{1121}\sigma_{21}\ ,\\
&\dot{\rho}_{22}=2\,\mathrm{Im}(\sigma_{21}\alpha^*)+R_{2222}\rho_{22}\\
&\qquad +R_{2211}\rho_{11}+R_{2221}\sigma_{21}+R_{2212}\sigma_{12}\ ,\\
&R_{1111}=-[c^4(a_1-a_1^{(0)})+2c^2s^2(a_0-a_0^{(0)})\\
&\qquad +s^4(a_{-1}-a_{-1}^{(0)})]\ ,\\
&R_{2222}=-[c^4(a_1+a_1^{(0)})+2c^2s^2(a_0+a_0^{(0)})\\
&\qquad +s^4(a_{-1}+a_{-1}^{(0)})]\ ,\\
&R_{2111}=[c^2a_1^{(0)}-s^2a_{-1}^{(0)}+(s^2-c^2)a_0^{(0)}]cse^{-i\phi}\ ,\\
&R_{2112}=[2a_0-a_{-1}-a_1]c^2s^2e^{2i\phi}\ ,\\
&R_{1112}=[(c^2-s^2)a_0+s^2a_{-1}-c^2a_1]cse^{i\phi}\ ,\\
&R_{2121}=(R_{1111}+R_{2222})/2\ ,\\
&R_{1122}=-R_{2222}\ ,\\
&R_{2211}=-R_{1111}\ ,\\
&R_{2221}=-R^*_{1112}\ ,\\
&R_{1121}=R^*_{1112}\ ,\\
&R_{2112}=-R_{1112}\ ,\\
&R_{2122}=R_{2111}\ ,
\end{aligned}
\tag{17}
$$

where

$$
\begin{aligned}
&a_j=\frac{A_{21}}{\omega_{21}^3}(\Omega+j\tilde{\omega}_{21})^3\eta(\Omega+j\tilde{\omega}_{21})[n(\Omega+j\tilde{\omega}_{21})+\tfrac{1}{2}]\ ,\\
&a_j^{(0)}=a_j(n\equiv 0)\ ,\\
&\tilde{\omega}_{21}=S\sqrt{\delta^2+4|\alpha|^2}\ .
\end{aligned}
\tag{18}
$$

There appears the cross-relaxation terms, i.e., terms which are proportional to $\sigma_{21}$ and $\sigma_{12}$ in the equations for $\rho_{11}$ and $\rho_{22}$ and vice versa. The elements of the relaxation supermatrix depend on the amplitude and frequency of the field via the dependence of the mode density $\eta$, number of photons in the modes, transformation coefficients $c$ and $s$, as well as via the coefficient $(\Omega+j\tilde{\omega}_{21})^3$. In the absence of the field ($\alpha=0$) we obtain from (17) the usual Bloch equations in the form

$$
\begin{aligned}
&\dot{\sigma}_{21}=-\sigma_{21}(w_{21}+w_{12})/2\ ,\\
&\dot{\rho}_{11}=-w_{12}\rho_{11}+w_{21}\rho_{22}\ ,\\
&\dot{\rho}_{22}=-w_{21}\rho_{22}+w_{12}\rho_{11}\ ,
\end{aligned}
\tag{19}
$$

where $w_{21}=A_{21}[n(\omega_{21})+1]$, $w_{12}=A_{21}n(\omega_{21})$, or taking into account $\rho_{11}+\rho_{22}=1$, $w_{21}=\rho_{11}^{(0)}/T_1$, $w_{12}=\rho_{22}^{(0)}/T_1$, $(w_{21}+w_{12})/2=1/2T_1=1/T_2$, $d=\rho_{22}-\rho_{11}$, and $d_0=A_{21}T_1$ in the form

$$
\dot{\sigma}_{21}=-\frac{\sigma_{21}}{T_2}\ ,\quad \dot{d}=-\frac{d-d_0}{T_1}\ . \tag{20}
$$

If we neglect everywhere the frequency shift, Eq. (17) returns to the traditional Eq. (1). Strictly speaking it is cumbersome to define the order of corrections in Eq. (17) as compared to (1). Taking into account that $|s|,|c|,|\rho_{nm}|<1$ we can estimate it very roughly as $\tilde{\omega}_{21}/\omega_{21}$. However, taking into account that the coefficients $c$ and $s$ vary from 0 to $\frac{1}{2}$ and from 1 to $\frac{1}{2}$ with the increasing of the field amplitude, and that in the absence of the field cross-relaxation terms were absent, and that apart from comparison with the old relaxation terms we have to compare the additional terms also with the dynamical terms $\sigma_{21}\alpha^*$, we can conclude that our estimation certainly is not correct in general. The only correct way to estimate the order of corrections would be to solve the Eq. (17). The influence of the additional terms can be quite different depending on the problem under consideration: the steady-state regime, its stability analysis, transient effects, the behavior of atoms driven by the field or self-consistent laser problem, etc.

However, to demonstrate the possibility of new effects, we consider the atoms behavior in the simplest resonant case when $\eta=1$ and $n(\Omega+j\tilde{\omega}_{21})=\text{const}$. It is worth noting that even under these conditions, moreover even at $n(\Omega+j\tilde{\omega}_{21})=0$ for large $\Omega+j\tilde{\omega}_{21}$, field-dependent relaxation rate depends on the frequency [see Eq. (18)] and hence on the amplitude of the coherent field $\alpha$. Keeping the linear and second-order terms with respect to $x=2|\alpha|/\omega_{21}$ we obtain from (17)

$$
\begin{aligned}
&\dot{\sigma}_{21}=-\frac{\sigma_{21}}{T_2}-i\alpha d-x(3+x^2)e^{i\phi}\frac{d_0}{2T_2}\\
&\qquad -3x^2e^{i\phi}\frac{\mathrm{Re}(e^{i\phi}\sigma_{21})}{T_2}\ ,\\
&\dot{d}=-\frac{(d-d_0)}{T_1}(1+\tfrac{3}{2}x^2)\\
&\qquad +4\,\mathrm{Im}(\alpha^*\sigma_{21})-x(3+x^2)\mathrm{Re}(e^{i\phi}\sigma_{21})/T_1\ ,
\end{aligned}
\tag{21}
$$

and the steady-state solution of (21) is

$$
\begin{aligned}
&\frac{\sigma_{21}}{\alpha}=-iT_2d-\frac{d_0}{\omega_{21}}(3-8x^2)\ ,\\
&d=\frac{d_0}{1+4|\alpha|^2T_1T_2+12x^2(|\alpha|^2T_1T_2+\frac{3}{8})}\ .
\end{aligned}
\tag{22}
$$

Thus in a steady state there is a saturation of population difference but with a slightly reduced saturation intensity. But it is important that the magnitude $\mathrm{Re}\sigma_{21}/\alpha$, which characterizes the dispersion, is not zero as it was in Eq. (1). It was noticed also in Refs. [4,5] on the basis of a different approach. If we look to the origin of this term we can see that it appeared because we took first the resonant case $\delta=0$ and only then the limit $\alpha\to 0$. If we use the opposite sequence (first take $\alpha\to 0$ and then the limit $\delta\to 0$, supposing $\alpha\ll\delta$) we would get the usual result. This means that under the action of even a weak field there appears a dispersion (frequency shift) in the case of very small detunings ($\delta \ll \alpha$). This effect has been completely ignored in the usual Eq. (1).

## IV. THREE-LEVEL SYSTEM

Using the expression for unperturbed coefficients (15) as well as the connection between perturbed and unperturbed relaxation supermatrices (11) and following the same procedure as for a two-level case, we obtain a master equation in the secular approximation for three-level atoms driven by the monochromatic field resonant to the transition between upper levels 2 and 3:

$$\begin{aligned}
&\dot\rho_{22}+2\,\mathrm{Im}(\sigma_{32}\alpha^*)=R_{2211}\rho_{11}+R_{2222}\rho_{22}+R_{2233}\rho_{33}+2\,\mathrm{Re}(R_{2223}\rho_{23})\ ,\\
&\dot\rho_{33}-2\,\mathrm{Im}(\sigma_{32}\alpha^*)=R_{3311}\rho_{11}+R_{3322}\rho_{22}+R_{3333}\rho_{33}+2\,\mathrm{Re}(R_{3323}\rho_{23})\ ,\\
&\dot\sigma_{32}+i\delta\sigma_{32}=i\alpha n_{23}+R_{3232}\sigma_{32}+R_{3211}\rho_{11}+R_{3222}\rho_{22}+R_{3233}\rho_{33}+R_{3223}\sigma_{23}\ ,\\
&\sigma_{31}=i\alpha\sigma_{21}+R_{3131}\sigma_{31}+R_{3121}\sigma_{21}\ ,\\
&\dot\sigma_{21}-i\delta\sigma_{21}=i\alpha^*\sigma_{31}+R_{2121}\sigma_{21}+R_{2131}\sigma_{31}\ ;
\end{aligned} \tag{23}$$

$$\rho_{32}=\sigma_{32}e^{-i\Omega t}\ ,\quad \rho_{31}=\sigma_{31}e^{-i\omega_{31}t}\ ,$$

$$\rho_{21}=\sigma_{21}e^{-i(\omega_{31}-\Omega)t}\ ,\quad \delta=\omega_{32}-\Omega\ ,$$

$$R_{2211}=c^2w_{12}(\tilde\omega_{21}-\Omega)+s^2w_{12}(\tilde\omega_{31}-\Omega)\ ,$$

$$R_{3311}=s^2w_{13}(\tilde\omega_{21})+c^2w_{13}(\tilde\omega_{31})\ ,$$

$$R_{2222}=-(R_{3322}+R_{1122})\ ,$$

$$R_{3333}=-(R_{1133}+R_{2233})\ ,$$

$$R_{1122}=c^2w_{21}(\tilde\omega_{12}+\Omega)+s^2w_{21}(\tilde\omega_{13}+\Omega)\ ,$$

$$R_{1133}=c^2w_{31}(\tilde\omega_{13})+s^2w_{31}(\tilde\omega_{12})\ ,$$

$$R_{3322}=c^4w_{23}(\tilde\omega_{32}+\Omega)+s^4w_{23}(\tilde\omega_{23}+\Omega)+2s^2c^2w_{23}(\Omega)\ ,$$

$$R_{2233}=2s^2c^2(w_{32}-\Omega)+s^4w_{32}(\tilde\omega_{32}-\Omega)+c^4w_{32}(w_{23}-\Omega)\ ,$$

$$\begin{aligned}R_{2223}=e^{-i\phi}\frac{sc}{2}\{&(c^2-s^2)[w_{32}(-\Omega)+w_{23}(\Omega)]\\&+s^2[w_{32}(\tilde\omega_{32}-\Omega)+w_{23}(\tilde\omega_{23}+\Omega)]\\&-c^2[w_{32}(\tilde\omega_{23}-\Omega)+w_{23}(\tilde\omega_{32}+\Omega)]\\&+w_{21}(\tilde\omega_{13}+\Omega)-w_{21}(\tilde\omega_{12}+\Omega)\}\ ,\end{aligned}$$

$$\begin{aligned}R_{3323}=e^{-i\phi}\frac{sc}{2}\{&(s^2-c^2)[w_{23}(\Omega)+w_{32}(-\Omega)]\\&+c^2[w_{32}(\tilde\omega_{23}-\Omega)+w_{23}(\tilde\omega_{32}+\Omega)]\\&-s^2[w_{32}(\tilde\omega_{32}-\Omega)+w_{23}(\tilde\omega_{23}+\Omega)\\&-w_{31}(\tilde\omega_{12})+w_{31}(\tilde\omega_{13})]\}\ ,\end{aligned}$$

$$R_{3232}=\frac{R_{2222}+R_{3333}}{2}\ ,\quad R_{2121}=\frac{R_{1111}+R_{2222}}{2}\ ,\quad R_{3131}=\frac{R_{1111}+R_{3333}}{2}\ , \tag{24}$$

$$\begin{aligned}R_{3211}=e^{-i\phi}\frac{sc}{2}[&w_{12}(\tilde\omega_{21}-\Omega)-w_{12}(\tilde\omega_{31}-\Omega)\\&+w_{13}(\tilde\omega_{21})-w_{13}(\tilde\omega_{31})]\ ,\end{aligned}$$

$$\begin{aligned}R_{3222}=e^{-i\phi}\frac{sc}{2}\{&(c^2-s^2)[w_{23}(\Omega)-w_{32}(-\Omega)]\\&+s^2[w_{23}(\tilde\omega_{23}+\Omega)-w_{32}(\tilde\omega_{32}-\Omega)]\\&-c^2[w_{23}(\tilde\omega_{32}+\Omega)-w_{32}(\tilde\omega_{23}-\Omega)]\\&+w_{31}(\tilde\omega_{13})-w_{31}(\tilde\omega_{12})\}\ ,\end{aligned}$$

$$\begin{aligned}R_{3233}=e^{-i\phi}\frac{sc}{2}\{&(s^2-c^2)[w_{32}(-\Omega)-w_{23}(\Omega)]\\&+s^2[w_{23}(\tilde\omega_{23}+\Omega)-w_{32}(\tilde\omega_{32}-\Omega)]\\&+c^2[w_{32}(\tilde\omega_{23}-\Omega)-w_{23}(\tilde\omega_{32}+\Omega)]\\&+w_{21}(\tilde\omega_{12}+\Omega)-w_{21}(\tilde\omega_{12}+\Omega)\}\ ,\end{aligned}$$

$$\begin{aligned}R_{3223}=e^{-2i\phi}\frac{s^2c^2}{2}[&2w_{32}(-\Omega)+2w_{23}(\Omega)\\&-w_{32}(\tilde\omega_{32}-\Omega)-w_{23}(\tilde\omega_{23}+\Omega)\\&-w_{32}(\tilde\omega_{23}-\Omega)-w_{23}(\tilde\omega_{32}+\Omega)]\ ,\end{aligned}$$

$$\begin{aligned}R_{2131}=e^{-i\phi}\frac{sc}{2}\{&(c^2-s^2)w_{23}(\Omega)-c^2w_{23}(\tilde\omega_{32}+\Omega)\\&+s^2w_{23}(\tilde\omega_{23}+\Omega)+w_{21}(\tilde\omega_{13}+\Omega)\\&-w_{21}(\tilde\omega_{12}+\Omega)\}\ ,\end{aligned}$$

$$\begin{aligned}R_{3121}=e^{-i\phi}\frac{sc}{2}[&w_{31}(\tilde\omega_{12})-w_{31}(\tilde\omega_{13})\\&+(c^2-s^2)w_{32}(-\Omega)-c^2w_{32}(\tilde\omega_{23}-\Omega)\\&+s^2w_{32}(\tilde\omega_{32}-\Omega)]\ ,\end{aligned}$$

$$w_{mk}(\omega_{ij})=\begin{cases}A_{mk}\left[\dfrac{\omega_{ij}}{\omega_{mk}}\right]^3\eta(\omega_{ij})n(\omega_{ij}) & \text{if } \omega_{ij}>0\ ,\\[2ex] A_{mk}\left[\dfrac{\omega_{ji}}{\omega_{mk}}\right]^3\eta(\omega_{ji})[n(\omega_{ji})+1] & \text{if } \omega_{ij}<0\ .\end{cases}$$

The first three equations of the set (23) describe a two-level system (levels 2 and 3) with different decays from the levels 2 and 3 if we substitute everywhere $\rho_{11}=1-\rho_{22}-\rho_{33}$. In our case the dressed frequencies are $\tilde\omega_{32}=s\sqrt{\delta^2+4|\alpha|^2}$, $\tilde\omega_{21}=\omega_{31}-(\delta+\tilde\omega_{32})/2$, and $\tilde\omega_{31}=\omega_{31}-(\delta-\tilde\omega_{32})/2$.

One can see that relaxation rates depend not only on the ratio of the Rabi frequency to the frequency of the resonant transition (as was the case in a two-level problem) but also on the ratio of the Rabi frequency to the frequency of the adjacent transition. Even in the Markovian approximation and the RWA the large values of this parameter are allowed. Physically it corresponds to the situation when the splitted sublevels (as a result of an ac Stark effect) cross the unperturbed atomic levels adjacent to the resonant one. Consequently, the scheme of the relaxation processes and the structure of the master equation are essentially modified. It leads to new field-dependent relaxation effects in a three-level systems (in particular, under definite conditions to the large value $\mathrm{Re}[\sigma_{21}\exp(-i\phi)]\sim\frac{1}{2}$, characterizing resonant dispersion). These effects will be considered in detail elsewhere.

## V. DISCUSSION

We demonstrated that even in the simplest Born, Markovian, RWA, and secular approximations there is an influence of the coherent field on the relaxation processed in the atomic system. The important parameter which often (but by no means always) defines the order of the additional terms in the master equation, as compared to the traditional relaxation terms, is the ratio of the Rabi frequency to the frequency of the resonant transition ($|\alpha|/\omega_{21}$). Since we used the RWA, secular, and Markov approximations we have to consider this parameter as a small one. However, we should note that (a) for a circular polarized transitions we do not need the RWA if the magnetic sublevels are well resolved, (b) the major formula (11) does not imply a secular approximation and hence we can easily avoid it also in concrete applications, (c) non-Markovian terms do not influence some steady state processes [14], and (d) in a multilevel system (beginning with the three-level one) there is a new important parameter which is a ratio of the Rabi frequency to the frequency of the adjacent transition. This last parameter also defines the order of the corrections and in comparison with $|\alpha|/\omega_{21}$, it can be quite large. Apart from that, the influence of the coherent fields on the relaxation processes is important because (a) it introduces a new nonlinearity into the Maxwell-Bloch equations and (b) it essentially changes the structure of the master equation. As a result it is difficult to define the order of the corrections in advance (i.e., before the solution of the master equation). Essential corrections are possible even when $|\alpha|/\omega_{21}\ll 1$.

It is worth mentioning that all the corrections in the master equation which are due to the influence of the coherent field on the relaxation supermatrix depend on the form of the Hamiltonian which we have chosen for the interaction of atoms with the field reservoir. Everywhere above we used the following form of the Hamiltonian: $H_{\mathrm{int}}=-\mathbf{d}\cdot\mathcal{E}$. If we would use the form $H_{\mathrm{int}}=-e\mathbf{p}\cdot\mathbf{A}/mc$, then instead of the coefficient $\sqrt{2\pi\omega_k}$ in Eq. (14) we had the coefficient $\sqrt{2\pi/\omega_k\omega_{21}}$. It does not influence, in fact, the relaxation coefficients in the absence of the field $\Gamma^{(0)}_{mpln}$ since anyway we have $\delta(\omega_{21}\pm\omega_k)$ under the integral on the right-hand side of Eq. (15). But it does influence the relaxation coefficients under the action of the field since we have to change everywhere $\omega_k$ (but not $\omega_{21}$) by the dressed frequencies in accordance with Eq. (11). As a result, in a two-level system the expression for $a_j$ (18) will contain the coefficient $(\Omega+j\tilde{\omega}_{21})/\omega_{21}$ (for $H_{\mathrm{int}}=-e\mathbf{p}\cdot\mathbf{A}/mc$) instead of the coefficient $(\Omega+j\tilde{\omega}_{21})^3/\omega_{21}^3$, which we had for $H_{\mathrm{int}}=-\mathbf{d}\cdot\mathcal{E}$.

In particular, in the last case if we would keep just the linear terms with respect to the ratio $\tilde{\omega}_{21}/\Omega$ we would obtain three times smaller coefficients as compared to the case $H_{\mathrm{int}}=-e\mathbf{p}\cdot\mathbf{A}/mc$. [Let us also note that in this linear approximation at $n(\omega)=\mathrm{const}, \eta(\omega)=1$, the relaxation coefficients depend only on the amplitude but not on the frequency of the field.]

However, the above-mentioned difference becomes nonessential when the scale $\Delta\omega$ of the functions $n(\omega)$ and/or $\eta(\omega)$ is much smaller than $\omega_{21}$ and an increasing parameter $(\omega_{21}/\Delta\omega)\gg 1$ defines a more abrupt behavior of these functions as compared to the smooth behavior of the coupling constants ($\omega^2$ or $\omega$). As a result, strong effects which are due to the dependence of relaxation coefficients on the coherent field via $n(\Omega+j\tilde{\omega}_{21})$ and $\eta(\Omega+j\tilde{\omega}_{21})$ play the dominant role.

Finally we found a generalized set of Maxwell-Bloch equations which possesses a new nonlinearity. Many processes in quantum optics and laser dynamics will be influenced by these results. One important application refers to the problem of amplification without population inversion in schemes involving strong coherent pumping [15–17]. Until now the analysis of this problem has been based on the traditional set of Maxwell-Bloch equations. Since the action of the coherent pumping leads to the appearance of new cross-relaxation terms in the master equation, it can essentially modify the conditions of inversionless amplification.

It is worth mentioning once again that the connection between the relaxation supermatrix in the presence and in the absence of the field has been obtained in the Born-Markovian approximation. At the same time the theory of the interaction of the atomic system with the reservoir has been developed beyond this approximation. It would be of interest to generalize this situation especially as it concerns our main result Eq. (11).

## ACKNOWLEDGMENTS

M.O.S. wishes to thank the ONR and the Welsh foundation for their support. S.Y.Z. acknowledges the support of Hong Kong Baptist College.

[1] H. Haken, in *Laser Theory in Encyclopedia of Physics*, edited by L. Genzel (Springer-Heidelberg, 1970), Vol. XXV/2c; G. S. Agarwal, in *Quantum Statistics Theories of Spontaneous Emission and Their Relation to Other Approaches*, edited by G. Hohler, Springer Tracts in Modern Physics Vol. 70 (Springer-Verlag, Heidelberg, 1974); G. K. Blum, *Density Matrix Theory and Applications* (Plenum, New York, 1981).

[2] A. G. Redfield, Phys. Rev. **98**, 1787 (1955).
[3] P. N. Argyres and P. L. Kelly, Phys. Rev. **134**, 98 (1964).
[4] R. H. Lehmber, Phys. Lett. **33A**, 501 (1970).
[5] G. S. Agarwal, in *Progress in Optics,* edited by E. Wolf (North-Holland, Amsterdam, 1973), Vol. XI.
[6] H. J. Carmichael and D. F. Walls, Phys. Rev. A **9**, 2686 (1974).
[7] C. Cohen-Tannoudji and S. Reynaund, J. Phys. B **10**, 345 (1977).
[8] P. A. Apanasevich, S. Ya. Kilin, A. P. Nizovtsev, and N. S. Onishchenko, Opt. Commun. **52**, 279 (1984).
[9] K. Wódkiewicz and J. H. Eberly, Phys. Rev. A **32**, 992 (1985).
[10] P. Berman and R. G. Brewer, Phys. Rev. A **32**, 2784 (1985).
[11] An. V. Vinogradov, Kvant. Elektron. (Moscow) **13**, 293 (1986).
[12] E. G. Pestov, Tr. FIAN (in Russian) **187**, 60 (1988).
[13] G. Gangopadhyay and D. S. Ray, Phys. Rev. A **46**, 1507 (1992); J. D. Cresser, J. Mod. Opt. **39**, 2187 (1992).
[14] A. A. Villiae, J. C. Vallet, and S. H. Lin, Phys. Rev. A **43**, 5030 (1991).
[15] O. Kocharovskaya, Phys. Rep. **219**, 175 (1992).
[16] M. Scully, Phys. Rep. **219**, 191 (1992).
[17] J. Seke and W. Herfort, Physica A **178**, 561 (1991).