Two-photon atomic relaxation in the field of a monochromatic nonresonant electromagnetic wave

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A mechanism is suggested for two-photon spontaneous atomic relaxation in an external monochromatic field. A unitary transformation of the initial Hamiltonian and a technique involving Ito's quantum stochastic differential equation are employed to obtain an equation for the atomic density matrix and the relaxation constants that describe two-photon atomic relaxation. The equation for the density matrix is generalized to the case of relaxation involving one photon of the monochromatic field and one photon of the squeezed vacuum. The possibility of experimentally studying two-photon relaxation is also demonstrated.

INTRODUCTION

Analyzing spontaneous atomic relaxation in an external coherent field usually involves solving an equation for the denisty matrix with given spontaneous relaxation constants and then determining the effective relaxation times.[1] It is commonly assumed that spontaneous relaxation is due to one-photon transitions in the absorption or emission of resonant vacuum photons. But in addition to one-photon processes, there can be a complex hierarchy of multiphoton spontaneous processes in the external field, when the transitions in an atom that lead to additional relaxation occur with the absorption of one or several photons from the coherent field simultaneously with the absorption or emission of a vacuum photon. Owing to the intensity of the external field, such processes may be pronounced and can dominate ordinary one-photon spontaneous relaxation. Obviously, when the intensity of the coherent field is high and the role of these processes is important, one must re-examine even the ordinary two-level model of an atom,[2] since for any such field, resonant or nonresonant to two selected energy levels, other energy levels and an appropriate frequency of the vacuum photon can always be found, with the result that one is forced to allow for relaxation transitions from the two selected levels to other levels with photons of both coherent and vacuum fields being involved. The respective relaxation constant will depend here on the intensity of the coherent field in the starting equations for the density matrix of the atom and must be taken into account in the N-level approximation. Needless to say, this constant will affect many optical phenomena.

In this paper[1)] the above mechanisms of spontaneous relaxation in an external coherent field described in classical terms are taken into account systematically. For the simplest case of one- and two-photon relaxations an equation is derived for the atomic density matrix containing, in addition to the usual Einstein relaxation constant, relaxation parameters that depend on the intensity of the external coherent electromagnetic field. The equation is generalized to the case of a squeezed vacuum, whose photons participate only in two-photon relaxation transitions. Relaxation constants that allow for these processes are obtained for the three-level model of an atom. The possibility of experimentally studying two-photon spontaneous relaxation is illustrated by the example of Raman scattering of light in the coherent excitation

of a three-level system by a pair of ultrashort pulses in resonance with adjacent optically allowed transitions.[3]

The theoretical approach expounded here is based on the unitary transformation method[4,5] and the technique of Ito's quantum stochastic differential equation.[6] In Sec. 1 the unitary transformation method is used to derive an effective Hamiltonian that in addition to one-photon relaxation transitions allows for two-photon transitions involving a vacuum photon and a coherent photon. The quantum stochastic equations of Langevin and Ito are derived in Sec. 2. Section 3 is devoted to the equation for the density matrix and its generalization to the case of a squeezed vacuum. Section 4 gives the relaxation constants for various three-level models of an atom. Finally, Sec. 5 discusses the possibility of experimentally studying two-photon relaxation.

1. THE EFFECTIVE HAMILTONIAN OF SPONTANEOUS PROCESSES IN AN EXTERNAL COHERENT FIELD

We will describe the system consisting of an atom, the field of a classical electromagnetic wave of frequency v and electric field strength

$$E = \mathscr{E} e^{-ivt} + \text{c.c.} \tag{1}$$

and the vacuum electromagnetic field by the Hamiltonian

$$H = H_a + H_b + V_{coh.int} + V_b, \quad H_a = \sum_\alpha E_\alpha a_\alpha^+ a_\alpha,$$

$$H_b = \int d\omega \hbar \omega b_\omega^+ b_\omega,$$

$$\tag{2}$$

$$V_{coh.int} = -\mathscr{E} e^{-ivt} \sum d_{\alpha\alpha'} a_\alpha^+ a_{\alpha'} + \text{H.c.},$$

$$V_b = -i \sum \int d\omega K(\omega) d_{\alpha\alpha'} a_\alpha^+ a_{\alpha'} b_\omega + \text{H.c.},$$

where H_a is the Hamiltonian of the isolated atom, H_b the Hamiltonian of the photon thermostate, $V_{coh.int}$ the operator of the interaction of the atom with field (1), and V_b the operator of the interaction of the atom with the thermostat photons. We have introduced the following notation: a_α^+ and a_α are the creation and annihilation operators of an atom in a state with energy E_α, satisfying the commutation relations $[a_\alpha, a_\alpha^+] = \delta_{\alpha\alpha'}$; b_ω^+ and b_ω are the creation and annihilation operators of photons of frequency ω, with

$[b_\omega, b_{\omega'}^+] = \delta(\omega - \omega')$; $d_{\alpha\alpha'}$ is the matrix element of the operator of the dipole moment between states with energies E_α and $E_{\alpha'}$; and $K(\omega)$ is the coupling constant.[2)] We have ignored polarization effects and recoil and employed the one-dimensional approximation, while the interaction with fields is considered in the electric dipole approximation.

The initial Hamiltonian H describes the evolution of the density matrix ρ of the entire system:

$$i\hbar \partial\rho/\partial t = [H, \rho].\tag{3}$$

To identify in this Hamiltonian the effective terms responsible for the hierarchy of the spontaneous processes under discussion, we perform the following unitary transformation:[5]

$$\bar\rho = e^{-iS}\rho e^{iS}, \quad i\hbar \partial\bar\rho/\partial t = [\tilde H, \bar\rho], \quad \tilde H = e^{-iS}He^{iS} - i\hbar e^{-iS}\partial e^{iS}/\partial t.\tag{4}$$

We write S and $\tilde H$ in the form of series in powers of the strengths of the coherent and vacuum fields:

$$S = S^{(10)} + S^{(01)} + S^{(11)} + \ldots,$$

$$\tilde H = \tilde H^{(00)} + \tilde H^{(10)} + \tilde H^{(01)} + \tilde H^{(11)} + \ldots,$$

where the left index in each pair of superscripts refers to the external coherent field and the right index to the vacuum field. Obviously,

$$\tilde H^{(00)} = H_a + H_b,$$
$$\tilde H^{(10)} = V_{coh.int} - i[S^{(10)}, \tilde H^{(00)}] + \hbar\partial S^{(10)}/\partial t,$$
$$\tilde H^{(01)} = V_b - i[S^{(01)}, \tilde H^{(00)}] + \hbar\partial S^{(01)}/\partial t,$$
$$\tilde H^{(11)} = -(i/2)[S^{(01)}, V_{coh.int}] - (i/2)[S^{(10)}, V_b]$$
$$-(i/2)[S^{(01)}, \tilde H^{(10)}]$$
$$-(i/2)[S^{(10)}, \tilde H^{(01)}] - i[S^{(11)}, \tilde H^{(00)}] + \hbar\partial S^{(11)}/\partial t, \ldots\tag{5}$$

Now we must require that $\tilde H^{(10)}$ vanish since the external electromagnetic field (1) is assumed to be out of resonance with the atom. The quantity $\tilde H^{(01)}$ must contain only terms with the appropriate frequency dependence, corresponding to absorption and emission of resonant vacuum photons:

$$\tilde H^{(01)} = -i\sum\int d\omega_l(\alpha\alpha')K[\omega_l(\alpha\alpha')]d_{\alpha\alpha'}a_\alpha^+ a_{\alpha'} b_{\omega_l(\alpha\alpha')}$$

$$\times\theta(E_\alpha - E_{\alpha'}) + \text{H.c.}\tag{6}$$

According to Ref. 7, the frequency spectrum of the vacuum photons is divided so that to each atomic transition there corresponds an individual source of noise related to the resonant interaction with thermostat photons, whose frequencies are denoted by $\omega_l(\alpha\alpha')$. Here $\omega_l(\alpha\alpha') = \omega_l(\alpha'\alpha)$, and the central frequency of this noise source, $\bar\omega_l(\alpha\alpha')$, is equal to $|\omega_{\alpha\alpha'}|$, where $\omega_{\alpha\alpha'} = (E_\alpha - E_{\alpha'})/\hbar$. We must also bear in mind that all the sources of noise just mentioned act as a single source in relation to an atomic transition out of resonance with these sources. We have also used the standard notation for the unit step function, that is, $\theta(x) = 1$ for $x > 0$ and $\theta(x) = 0$ for $x < 0$. Then, assuming that the coherent field is switched on adiabatically, we get

$$S^{(10)} = -i\sum\frac{d_{\alpha\alpha'}\mathscr{E}e^{-i\nu t}a_\alpha^+ a_{\alpha'}}{\hbar(\omega_{\alpha\alpha'} - \nu)} + \text{H.c.},$$

$$S^{(01)} = \sum\int' d\omega\,\frac{K(\omega)d_{\alpha\alpha'}a_\alpha^+ a_{\alpha'}b_\omega}{\hbar(\omega_{\alpha\alpha'} - \omega)}$$

$$+\sum\int d\omega_l(\alpha\alpha')$$

$$\times\frac{K(\omega_l(\alpha\alpha'))d_{\alpha\alpha'}a_\alpha^+ a_{\alpha'}b_{\omega_l(\alpha\alpha')}\theta(E_{\alpha'} - E_\alpha)}{\hbar[\omega_{\alpha\alpha'} - \omega_l(\alpha\alpha')]} + \text{H.c.},$$

where the prime on the integral sign indicates the absence of terms with resonant denominators.

We now substitute these expressions into the formula for $\tilde H^{(11)}$ and retain only the terms with the correct frequency dependence (by defining $S^{(11)}$ appropriately), which in the final analysis corresponds to the approximation of slowly varying amplitudes:

$$\tilde H^{(11)} = -\frac{i}{2}\sum\int d\omega_\theta(\alpha\gamma)K((\omega_\theta(\alpha\gamma))\mathscr{E}e^{-i\nu t}a_\alpha^+ a_\gamma b_{\omega_\theta(\alpha\gamma)}$$

$$\times[\Pi_{\alpha\gamma}(\nu) + \Pi_{\alpha\gamma}(\omega_\theta(\alpha\gamma))]\theta(E_\alpha - E_\gamma)$$

$$+\frac{i}{2}\sum\int d\omega_q(\alpha\gamma)K(\omega_q(\alpha\gamma))$$

$$\times\mathscr{E}e^{-i\nu t}a_\alpha^+ a_\gamma b_{\omega_q(\alpha\gamma)}^+[\Pi_{\alpha\gamma}(\nu) + \Pi_{\alpha\gamma}(-\omega_q(\alpha\gamma))]\theta(E_\gamma - E_\alpha)$$

$$+\frac{i}{2}\sum\int d\omega_p(\alpha\gamma)K(\omega_p(\alpha\gamma))\mathscr{E}e^{-i\nu t}a_\alpha^+ a_\gamma b_{\omega_p(\alpha\gamma)}^+$$

$$\times[\Pi_{\alpha\gamma}(\nu) + \Pi_{\alpha\gamma}(-\omega_p(\alpha\gamma))]\theta(E_\alpha - E_\gamma)$$

$$+\frac{i}{2}\sum\int d\omega_\nu K(\omega_\nu)\mathscr{E}^* e^{i\nu t}a_\alpha^+ a_\alpha b_{\omega_\nu}$$

$$\times[\Pi_\alpha(\nu) + \Pi_\alpha(\omega_\nu)] + \text{H.c.}\tag{7}$$

The noise source $\omega_\theta(\alpha\gamma)$ with the central frequency $\bar\omega_\theta(\alpha\gamma)$ is in two-photon resonance with the two-photon (optically forbidden) transition $E_\alpha \to E_\gamma$, that is, $\bar\omega_\theta(\alpha\gamma) + \nu = (E_\alpha - E_\gamma)/\hbar(E_\alpha > E_\gamma)$; the noise source $\omega_q(\alpha\gamma)$ with central frequency $\bar\omega_q(\alpha\gamma)$ and the noise source $\omega_p(\alpha\gamma)$ with central frequency $\bar\omega_p(\alpha\gamma)$ are in combination resonance with the two-photon transitions $E_\alpha \to E_\gamma$, that is, $\bar\omega_q(\alpha\gamma) - \nu = (E_\alpha - E_\gamma)/\hbar(E_\alpha > E_\gamma)$ and $\nu - \bar\omega_p(\alpha\gamma) = (E_\alpha - E_\gamma)/\hbar,(E_\alpha > E_\gamma)$; and the central frequency $\bar\omega_\nu$ of the noise source ω_ν coincides with the frequency ν of the nonresonant wave (1). Here we have introduced the following notation:[5]

$$\Pi_{\alpha\gamma}(\omega) = \sum\frac{d_{\alpha\beta}d_{\beta\gamma}}{\hbar}\left(\frac{1}{\omega_{\beta\alpha} + \omega} + \frac{1}{\omega_{\beta\gamma} - \omega}\right),$$

determines the effective dipole moment of the two-photon transition, and

$$\Pi_\alpha(\omega) = \sum\frac{|d_{\alpha\alpha'}|^2}{\hbar}\left(\frac{1}{\omega_{\alpha\alpha'} + \omega} + \frac{1}{\omega_{\alpha\alpha'} - \omega}\right),$$

determines the Stark shift of the level E_α. Since

$$\Pi_{\alpha\gamma}(\nu) = \Pi_{\alpha\gamma}(\bar\omega_\theta(\alpha\gamma)), \quad \Pi_{\alpha\gamma}(\nu) = \Pi_{\alpha\gamma}(-\bar\omega_q(\alpha\gamma))$$

and $\Pi_{\alpha\gamma}(\nu) = \Pi_{\alpha\gamma}(-\bar\omega_p(\alpha\gamma))$,

ignoring the frequency dependence of the Π parameters, we can write

$$\tilde{H}^{(11)} = -i \sum \int d\omega_\theta (\alpha\gamma) K((\omega_\theta(\alpha\gamma)) \mathscr{E} e^{-i\nu t} a_\alpha^+ a_\gamma b_{\omega_\theta(\alpha\gamma)}$$

$$\times \Pi_{\alpha\gamma}(\nu)\theta(E_\alpha - E_\gamma)$$

$$+i\sum \int d\omega_q(\alpha\gamma) K(\omega_q(\alpha\gamma)) \mathscr{E} e^{-i\nu t} a_\alpha^+ a_\gamma b_{\omega_q(\alpha\gamma)}^+$$

$$\times \Pi_{\alpha\gamma}(\nu)\theta(E_\gamma - E_\alpha)$$

$$+i\sum \int d\omega_p(\alpha\gamma) K(\omega_p(\alpha\gamma)) \mathscr{E} e^{-i\nu t} a_\alpha^+ a_\gamma b_{\omega_p(\alpha\gamma)}^+$$

$$\times \Pi_{\alpha\gamma}(\nu)\theta(E_\alpha - E_\gamma)$$

$$+i\sum \int d\omega_\nu K(\omega_\nu) \mathscr{E}^* e^{i\nu t} a_\alpha^+ a_\alpha b_{\omega_\nu} \Pi_\alpha(\nu) + \text{H.c.}$$

$$(7')$$

The term $\tilde{H}^{(20)}$ is obtained in the same way as in the classical case[5] and describes the Stark shift of atomic levels:

$$\tilde{H}^{(20)} = \sum |\mathscr{E}|^2 \Pi_\alpha(\omega) a_\alpha^+ a_\alpha. \qquad (8)$$

Thus, the effective Hamiltonian of the atomic system and vacuum photons in the field of a classical coherent electromagnetic wave can be written as the sum of four terms determined by Eqs. (5)–(8):

$$\tilde{H} = \tilde{H}^{(00)} + \tilde{H}^{(01)} + \tilde{H}^{(11)} + \tilde{H}^{(20)}. \qquad (9)$$

The first term $\tilde{H}^{(00)}$ is the sum of the Hamiltonians of the atomic and photon subsystems isolated from each other and from the other fields. The term $\tilde{H}^{(20)}$ characterizes the Stark shifts of the levels in the field of the nonresonant wave (1). Both $\tilde{H}^{(01)}$ and $\tilde{H}^{(11)}$ correspond to spontaneous relaxation processes in the slowly-varying-amplitude approximation, with $\tilde{H}^{(01)}$ corresponding to one-photon relaxation and $\tilde{H}^{(11)}$ to two-photon relaxation involving thermostat photon and the coherent wave. In what follows it is convenient to think of the effective Hamiltonian (9) as the sum of the Hamiltonian of the atomic system in the external coherent nonresonant field H_{sys}, the vacuum-photon Hamiltonian H_b, and the operator of one- and two-photon interaction of the atom with the vacuum photons:

$$\tilde{H} = H_{\text{sys}} + H_b + H_{\text{int}}, \qquad (9')$$

where $H_{\text{sys}} = H_a + H^{(20)}$ and $H_{\text{int}} = \tilde{H}^{(01)} + \tilde{H}^{(11)}$, and for the sake of brevity we write

$$H_{\text{int}} = -i\hbar\Sigma \int d\omega_j K(\omega_j) [f_j(t) R_j^+ b_{\omega_j} - f_j^*(t) R_j b_{\omega_j}^+]. \qquad (10)$$

In such expressions the sum index j numbers the atomic transitions and all the above-mentioned noise sources (Fig. 1) that are in resonance both with optically allowed transitions ($j = l$ and $\omega_l \approx \omega_{\alpha'\alpha} > 0$) and with two-photon transitions ($j = \theta$ for the double resonance $\omega_\theta + \nu \approx \omega_{\gamma\alpha} > 0$, and $j = q$ and $j = p$ for the combination resonances $\omega_q - \nu \approx \omega_{\gamma\alpha} > 0$ and $\nu - \omega_p \approx \omega_{\gamma\alpha} > 0$). The operators R_j and the parameters $f_j(t)$ for the respective transitions are

$$R_l = a_\alpha^+ a_{\alpha'} \cdot 0(E_{\alpha'} - E_\alpha), \qquad f_l(t) = d_{\alpha'\alpha}/\hbar,$$
$$R_\theta = a_\alpha^+ a_\gamma \theta(E_\gamma - E_\alpha), \qquad f_\theta(t) = \mathscr{E} e^{-i\nu t} \Pi_{\gamma\alpha}(\nu)/\hbar,$$
$$R_q = a_\alpha^+ a_\gamma \theta(E_\gamma - E_\alpha), \qquad f_q(t) = \mathscr{E}^* e^{i\nu t} \Pi_{\gamma\alpha}(-\nu)/\hbar,$$
$$R_p = a_\gamma^+ a_\alpha \theta(E_\gamma - E_\alpha), \qquad f_p(t) = \mathscr{E}^* e^{i\nu t} \Pi_{\alpha\gamma}(-\nu)/\hbar.$$

In addition, j "incorporates" the noise source ω_ν at the frequency of the nonresonance wave (1) ($j = \nu$ at $\omega_\nu \approx \nu$). In contrast to the noise sources ω_l, ω_θ, ω_q, and ω_p, the ω_ν photons cause random variations to occur in the Stark frequency shifts rather than participate in atomic transition. For this source,

$$R_\nu = \sum a_\alpha^+ a_\alpha \Pi_\alpha(\nu), \qquad f_\nu(t) = -\mathscr{E}^* e^{i\nu t}/\hbar.$$

If in addition to the nonresonant field (1) there is a resonant classical electromagnetic field acting on the atomic system, the atomic Hamiltonian H_{sys} incorporates the term[5] that allows for this resonant interaction in the slowly varying amplitude approximation. This introduces additional terms into $S^{(10)}$ that contribute to (10) and to the Stark shifts of the resonant levels. In relation to nonresonant transitions some of these terms are the same as those considered above but are determined by this resonance frequency. The other fraction differs for resonant transitions and, among other things, is the cause of the Bloch–Siegert shift.[5] Often, however, these terms can be ignored, which is done in the actual calculation in Sec. 5.

2. THE QUANTUM EQUATIONS OF LANGEVIN AND ITO

We go over to the Heisenberg representation and write the equation of motion for an operator of the atomic system, say A:

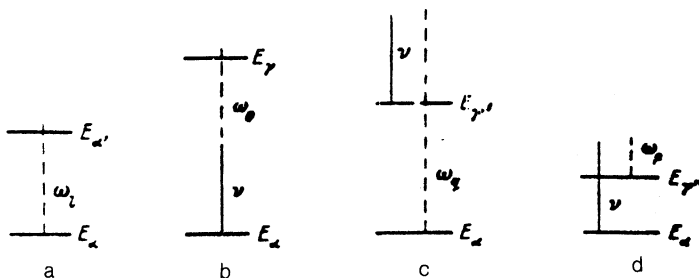FIG. 1. The energy levels and noise sources participating in (a) one-photon and (b)–(d) two-photon relaxation processes.

$$A = -\frac{i}{\hbar}[A, H_{sys}] - \sum_j \int d\omega_j K(\omega_j)\{f_j(t)[A, R_j^+]b_{\omega_j}$$

$$-f_j^*(t)[A, R_j]b_{\omega_j}^+\}.$$

In this equation we replace b_{ω_j} with the integral representation

$$b_{\omega_j} = \exp[-i\omega_j(t-t_0)]b_{0\omega_j} + K(\omega_j)\int_{t_0}^{t} f_j^*(t')R_j(t')$$

$$\times \exp[-i\omega_j(t-t')]dt',$$

where $b_{0\omega_j}$ is the value of b_{ω_j} at the initial time t_0, and apply the Markov approximation[6]

$$K(\omega_j) = (\varkappa_j/2\pi)^{1/2}. \tag{11}$$

Assuming that

$$\int_{t_0}^{t} R_j(t')\delta(t-t')dt' = \frac{1}{2}R_j(t),$$

we arrive at the quantum Langevin equation

$$A = -\frac{i}{\hbar}[A, H_{sys}] - \sum_j \left\{ [A, R_j^+] \right.$$

$$\times \left(\varkappa_j^{1/2} b_{(in)j}(t)f_j(t) + \frac{\varkappa_j}{2}|f_j(t)|^2 R_j \right)$$

$$\left. - \left(\varkappa_j^{1/2} b_{(in)j}^+(t)f_j(t) + \frac{\varkappa_j}{2}|f_j(t)|^2 R_j^+ \right)[A, R_j] \right\}. \tag{12}$$

Here for each noise source we have introduced the in-fields

$$b_{(in)j}(t) = \frac{1}{(2\pi)^{1/2}}\int d\omega_j \exp[-i\omega_j(t-t_0)]b_{0\omega_j}, \tag{13}$$

which satisfy the commutation relations

$$[b_{(in)j}(t), b_{(in)j'}^+(t')] = \delta_{jj'}\delta(t-t'), \quad [b_{(in)j}(t), b_{(in)j'}(t')] = 0,$$

Note that $\sqrt{\varkappa_j}\, b_{(in)j}(t)f_j(t) + \frac{1}{2}\varkappa_j|f_j(t)|^2 R_j$ commutes with any atomic operator.

We assume that the in-fields are sources of white noise:

$$\text{Tr}\{\rho_{(in)}b_{(in)j}^+(t)b_{(in)j'}(t')\} \equiv \langle b_{(in)j}^+(t)b_{(in)j'}(t')\rangle = N_j\delta_{jj'}(t-t'),$$

$$\text{Tr}\{\rho_{(in)}b_{(in)j}(t)b_{(in)j'}^+(t')\} \equiv \langle b_{(in)j}(t)b_{(in)j'}^+(t')\rangle$$

$$= (N_j+1)\delta_{jj'}\delta(t-t'), \tag{14}$$

$$\langle b_{(in)j}(t)b_{(in)j'}(t')\rangle = \langle b_{(in)j}^+(t)b_{(in)j'}^+(t')\rangle = 0, \tag{15}$$

which corresponds to

$$\langle b_{0\omega_j}^+ b_{0\omega_j'}\rangle = N_j\delta(\omega_j-\omega_j'), \quad \langle b_{0\omega_j}b_{0\omega_j'}^+\rangle = (N_j+1)\delta(\omega_j-\omega_j'),$$

$$\langle b_{0\omega_j}b_{0\omega_j'}\rangle = \langle b_{0\omega_j}^+ b_{0\omega_j'}^+\rangle = 0,$$

with $\rho_{(in)}$ the density matrix of the in-fields. If we also assume that $N_j = \bar{N}(\omega_j)$, where $\bar{N}(\omega) = 1/[\exp(\hbar\omega/$

$kT) - 1]$, Eqs. (14) will correspond to the case of a photon thermostat of a temperature T.

If we reason along lines similar to Ref. 6, we can also examine out-fields and the respective quantum Langevin equation reversed in time. But this will not be needed here.

We introduce the quantum Wiener processes $B_j(t,t_0)$ in the following manner:

$$B_j(t, t_0) = \int_{t_0}^{t} b_{(in)j}(t')dt', \tag{16}$$

with $[B_j(t,t_0), B_j^+(t,t_0)] = (t-t_0)\delta_{jj'}$. Defining the Ito integral and differential in the usual way,[6] we have the following basic formulas of stochastic analysis:

$$dB_j^+(t)dB_{j'}(t) = N_j\delta_{jj'}dt, \quad dB_j(t)dB_{j'}^+(t) = (N_j+1)\delta_{jj'}dt,$$

$$dB_j(t)dB_{j'}(t) = dB_j^+(t)dB_{j'}^+(t) = dtdt$$

$$= dtdB_j^+(t) = dtdB_j(t) = 0. \tag{17}$$

To obtain Ito's quantum stochastic differential equation from the quantum Langevin equation (12), we must replace $b_{(in)j}(t)dt$ with $dB_j(t)$ and then add terms that will guarantee the validity in the obtained equation of Ito's rule of differentiation; namely, that

$$d(A_1 A_2) = (dA_1)A_2 + A_1 dA_2 + (dA_1)(dA_2)$$

for each pair of atomic operators A_1 and A_2. This leads to the quantum Ito equation

$$dA = -\frac{i}{\hbar}[A, H_{sys}]dt - \sum_j \frac{\varkappa_j}{2}|f_j(t)|^2$$

$$\times ([A, B_j^+]R_j - R_j^+[R_j, A])dt$$

$$- \sum_j \frac{\varkappa_j}{2}|f_j(t)|^2 N_j([[A, R_j^+], R_j] + [[A, R_j], R_j^+])dt$$

$$+ \sum_j \varkappa_j^{1/2} f_j^*(t)[A, R_j]dB_j^+(t) - \sum_j \varkappa_j^{1/2} f_j(t)[A, R_j^+]dB_j(t), \tag{18}$$

which can be written as

$$dA = -\frac{i}{\hbar}[A, H_{sys}]dt + \sum_j \frac{\varkappa_j}{2}|f_j(t)|^2(N_j+1)(R_j^+[A, R_j]$$

$$+ [R_j^+, A]R_j)dt + \sum_j \frac{\varkappa_j}{2}|f_j(t)|^2 N_j(R_j[A, R_j^+] + [R_j, A]R_j^+)dt$$

$$+ \sum_j \varkappa_j^{1/2} f_j^*(t)[A, R_j]dB_j^+(t) - \sum_j \varkappa_j^{1/2} f_j(t)[A, R_j^+]dB_j(t). \tag{18'}$$

The structure of this equation coincides with that of the Ito equation of Ref. 6, but because of the factor $|f_j(t)|^2$ the terms with $j = \theta, j = q, j = p$, and $j = v$ are proportional to the intensity of the external field (1) and describe the spontaneous processes of two-photon relaxation with simultaneous participation of the coherent wave (1) and vacuum photons.

3. THE EQUATION FOR THE ATOMIC DENSITY MATRIX

In the Ito equation the coefficients of $dB_j(t)$ and $dB_j^+(t)$ are nonadvanced operator functions. Hence, averaging (18), we get

$$\frac{\langle dA(t)\rangle}{dt} = -\left\langle \frac{i}{\hbar}[A, H_{sys}]\right\rangle - \left\langle \sum \frac{\varkappa_j}{2}|f_j(t)|^2 \right.$$

$$\times ([A, R_j^+]R_j - R_j^+[R_j, A])\rangle$$

$$-\left\langle \sum \frac{\varkappa_j}{2}|f_j(t)|^2 N_j([[A, R_j^+], R_j] + [[A, R_j], R_j^+])\right\rangle.$$

Here the average of an atomic operator in the Heisenberg representation is defined in terms of the density matrix

$$\rho(t) = \mathrm{Tr}_b\{\exp[i\tilde{H}(t-t_0)/\hbar]\rho_{sys}(t_0)$$

$$\otimes \rho_b(t_0)\exp[-i\tilde{H}(t-t_0)/\hbar]\}$$

as

$$\langle A(t)\rangle = \mathrm{Tr}\{A(t_0)\rho(t)\}$$

[initially, at $t = t_0$, the density matrix is equal to the product of the density matrix $\rho_{sys}(t_0)$ of the atomic system and that of the photon thermostat, $\rho_b(t_0)$]. Here

$$\frac{\langle dA(t)\rangle}{dt} = \frac{d\langle A(t)\rangle}{dt} = \mathrm{Tr}\left\{ A(t_0)\left(\frac{i}{\hbar}[\rho, H_{sys}]\right.\right.$$

$$+ \sum \frac{\varkappa_j}{2}|f_j(t)|^2(N_j+1)(2R_j\rho R_j^+ - \rho R_j^+ R_j - R_j^+ R_j\rho) \quad (19)$$

$$\left.\left. + \sum \frac{\varkappa_j}{2}|f_j(t)|^2 N_j(2R_j^+\rho R_j - \rho R_j R_j^+ - R_j R_j^+\rho)\right)\right\}.$$

Since

$$\frac{d\langle A(t)\rangle}{dt} = \mathrm{Tr}\left\{ A(t_0)\frac{d\rho(t)}{dt}\right\}, \quad (20)$$

and the above equations are valid for any atomic operator A, comparison of (19) and (20) yields the following equation for the atomic density matrix:

$$\frac{d\rho}{dt} = \frac{i}{\hbar}[\rho, H_{sys}] + \sum \frac{\varkappa_j}{2}|f_j(t)|^2(N_j+1)$$

$$\times (2R_j\rho R_j^+ - \rho R_j^+ R_j - R_j^+ R_j\rho)$$

$$+ \sum \frac{\varkappa_j}{2}|f_j(t)|^2 N_j(2R_j^+\rho R_j - \rho R_j R_j^+ - R_j R_j^+\rho).$$

$$(21)$$

This equation can easily be generalized to the case that lately has been a topic of intensive investigation,[8-12] where a vacuum field (with $j = l, \theta, q$, or p) or several such fields are in a "squeezed" state.[13] In Refs. [8-12] and other papers the relaxation of an atomic system was considered as being in a squeezed vacuum in resonance with optically allowed transitions. The above reasoning, however, suggests that after the transformed Hamiltonian is written in the form (9') and (10), the general analysis of ordinary one-photon relaxation and of the mechanism of two-photon relaxation proposed in this paper can be conducted along similar lines. Hence in what follows we denote by ω_s the frequencies of the fields of the squeezed vacuum that are in one-photon resonance with optically allowed transitions ($\omega_s \approx \omega_{\alpha'\alpha} > 0$) and in two-photon resonance with optically forbidden transitions ($\omega_s + \nu \approx \omega_{\gamma\alpha} > 0$ for a double resonance and $\omega_s - \nu \approx \omega_{\gamma\alpha} > 0$ and $\nu - \omega_s \approx \omega_{\gamma\alpha} > 0$ for combination resonances). The meaning of the respective operators R_s and parameters $f_s(t)$

remains unchanged. For ideal squeezing the difference from the case of ideal white noise and thermostat consists in replacing condition (15) with

$$\langle b_{(in)s}(t)b_{(in)s'}(t')\rangle = M_s\delta_{ss'}\delta(t-t'),$$

$$\langle b_{(in)s}^+(t)b_{(in)s'}^+(t')\rangle = M_s^*\delta_{ss'}\delta(t-t'),$$

$$(22)$$

and, in deriving the Ito equation, instead of

$$dB_s(t)dB_s(t) = dB_s^+(t)dB_s^+(t) = 0$$

we must assume that

$$dB_s(t)dB_{s'}(t) = M_s\delta_{ss'}dt, \quad dB_s^+(t)dB_{s'}^+(t) = M_s^*\delta_{ss'}dt,$$

where the parameters M_s and N_s obey the condition $|M_s|^2 \leqslant N_s(N_s + 1)$, with the equality attained for squeezed light exiting from an ideal degenerate parametric amplifier.[14] The following result can easily be obtained:

$$\frac{d\rho}{dt} = \frac{i}{\hbar}[\rho, H_{sys}] + \sum \frac{\varkappa_j}{2}|f_j(t)|^2(N_j+1)$$

$$\times (2R_j\rho R_j^+ - \rho R_j^+ R_j - R_j^+ R_j\rho)$$

$$+ \sum \frac{\varkappa_j}{2}|f_j(t)|^2 N_j(2R_j^+\rho R_j - \rho R_j R_j^+ - R_j R_j^+\rho)$$

$$- \sum \frac{\varkappa_s}{2}|f_s(t)|^2 M_s(2R_s^+\rho R_s^+ - \rho R_s^+ R_s^+ - R_s^+ R_s^+\rho)$$

$$(23)$$

$$- \sum \frac{\varkappa_s}{2}|f_s(t)|^2 M_s^*(2R_s\rho R_s - \rho R_s R_s - R_s R_s\rho),$$

where in the sums over j we have included the summation over s.

4. THE RELAXATION OPERATOR FOR THREE-LEVEL MODELS OF AN ATOM

It has proved expedient to write the equation for the transformed atomic density matrix $\tilde{\rho}$ in terms of the relaxation operator $\hat{\Gamma}$ as follows:

$$i\hbar\left(\frac{\partial}{\partial t} + \hat{\Gamma}\right)\tilde{\rho} = [\tilde{H}_s, \tilde{\rho}]. \quad (24)$$

Here the Hamiltonian $\tilde{H}_s$ of the atomic system may include, in addition to $\tilde{H}_{sys}$, the interaction of the atom with resonant (classical) electromagnetic fields. We assume that the appropriate unitary transformation[5] [similar to (4)-(9)] have been applied to the initial density matrix and the additional terms in the initial Hamiltonian that describe this resonant interaction with classical electromagnetic fields.

The simplest model of an atom allowing for spontaneous relaxation in an external field contains three levels, say E_a, E_b, and E_c, that form two adjoining optically allowed transitions E_b-E_a and E_b-E_c and an optically forbidden (two-photon) transition E_c-E_a. Three different configurations are possible here, Λ, V, and θ, and in each configuration the frequency ν of the coherent wave (1) can be either lower or higher than the frequency ω_{ca} of the optically forbidden transition. Below for these six three-level models of an atom (Fig. 2) we list the expressions for the matrix elements of the relaxation operator $(\hat{\Gamma}\rho)_{\alpha\alpha'}$. The matrix elements of the other terms in Eq. (24) are well-known (see, e.g., Refs. 5 and 15).
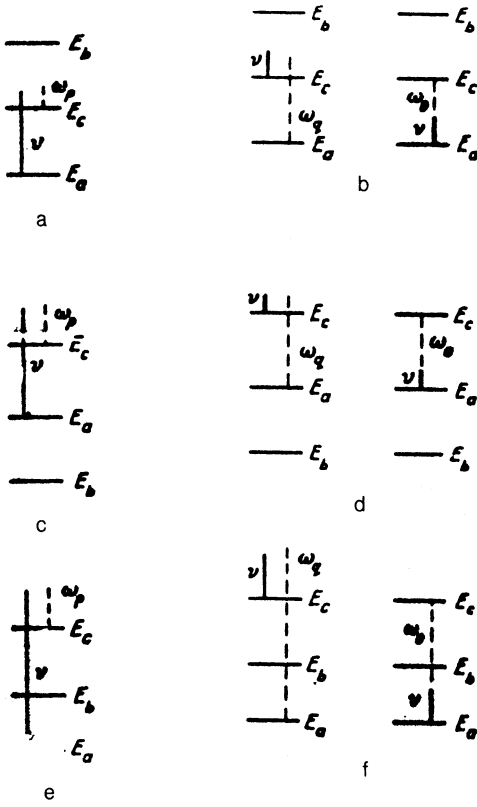
FIG. 2. Three-level models of an atom and two-photon relaxation transitions in which the photons of the monochromatic and noise fields participate.

We write the matrix elements of the relaxation operator in a form that reflects the "closed" nature of the three-level system (i.e., the fact that the number of three-level atoms does not change):

$$(\hat{\Gamma}\rho)_{aa} = (\Gamma_a^{(b)} + \Gamma_a^{(c)})\rho_{aa} - \Gamma_b^{(a)}\rho_{bb} - \Gamma_c^{(a)}\rho_{cc},$$
$$(\hat{\Gamma}\rho)_{bb} = (\Gamma_b^{(a)} + \Gamma_b^{(c)})\rho_{bb} - \Gamma_a^{(b)}\rho_{aa} - \Gamma_c^{(b)}\rho_{cc},$$

$$(\hat{\Gamma}\rho)_{cc} = (\Gamma_c^{(a)} + \Gamma_c^{(b)})\rho_{cc} - \Gamma_a^{(c)}\rho_{aa} - \Gamma_b^{(c)}\rho_{bb},$$

$$(\hat{\Gamma}\rho)_{ba} = (\Gamma_{ba}^{(ba)} + \Gamma_{ba}^{(bc)} + \Gamma_{ba}^{(ca)} + \Gamma_{ba})\rho_{ba},$$

$$(\hat{\Gamma}\rho)_{cb} = (\Gamma_{cb}^{(cb)} + \Gamma_{cb}^{(ca)} + \Gamma_{cb}^{(ba)} + \Gamma_{cb})\rho_{cb},$$

$$(\hat{\Gamma}\rho)_{ca} = (\Gamma_{ca}^{(ca)} + \Gamma_{ca}^{(cb)} + \Gamma_{ca}^{(ba)} + \Gamma_{ca})\rho_{ca}.$$

$$\Gamma_a^{(c)} = 2\Gamma_{ba}^{(ca)}, \quad \Gamma_c^{(a)} = 2\Gamma_{cb}^{(ca)}, \quad \Gamma_b^{(c)} = 2\Gamma_{ba}^{(cb)}, \quad \Gamma_c^{(b)} = 2\Gamma_{ca}^{(cb)}$$
$$\Gamma_a^{(b)} = 2\Gamma_{ca}^{(ba)}, \quad \Gamma_b^{(a)} = 2\Gamma_{cb}^{(ba)}.$$

In the formulas referring to the off-diagonal elements of the density matrix, the order of the indices in each pair of lower (upper) indices is unimportant. We write the constants $\Gamma_{\alpha\beta}$, $\alpha \neq \beta$ (i.e., Γ_{ca}, Γ_{cb}, and Γ_{ba}) in the unified form

$$\Gamma_{\alpha\beta} = \varkappa_v |\mathscr{E}|^2 [\Pi_\alpha(v) - \Pi_\beta(v)]^2 (N_v + 1/2)/\hbar^2. \quad (25)$$

and the remaining constants for each possible configuration of the three-level system as follows:

1. Λ-configuration $E_a < E_c < E_b$ with $v > \omega_{ca}$ (Fig. 2a):

$$\Gamma_{ba}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba} + 1/2), \quad \Gamma_{ba}^{(bc)} = \varkappa_{bc} |d_{bc}/\hbar|^2 (N_{bc} + 1)/2,$$

$$\Gamma_{ba}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 (N_p + 1)/2\hbar^2,$$

$$\Gamma_{bc}^{(bc)} = \varkappa_{bc} |d_{bc}/\hbar|^2 (N_{bc} + 1/2), \quad \Gamma_{bc}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba} + 1)/2.$$

$$\Gamma_{bc}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 N_p/2\hbar^2.$$

$$\Gamma_{ca}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 (N_p + 1/2)/\hbar^2,$$

$$\Gamma_{ca}^{(cb)} = \varkappa_{bc} |d_{bc}/\hbar|^2 N_{bc}/2, \quad \Gamma_{ca}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 N_{ba}/2,$$

2. Λ-configuration $E_a < E_c < E_b$ with $v < \omega_{ca}$ (Fig. 2b):

$$\Gamma_{ba}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba} + 1/2), \quad \Gamma_{ba}^{(bc)} = \varkappa_{bc} |d_{bc}/\hbar|^2 (N_{bc} + 1)/2,$$

$$\Gamma_{ba}^{(ca)} = (\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 N_q + \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 N_\theta)/2\hbar^2,$$

$$\Gamma_{bc}^{(bc)} = \varkappa_{bc} |d_{bc}/\hbar|^2 (N_{bc} + 1/2), \quad \Gamma_{bc}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba} + 1)/2,$$

$$\Gamma_{bc}^{(ca)} = [\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 (N_q + 1) + \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 (N_\theta + 1)]/2\hbar^2,$$

$$\Gamma_{ca}^{(ca)} = [\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 (N_q + 1/2)$$
$$+ \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 (N_\theta + 1/2)]/\hbar^2,$$

$$\Gamma_{ca}^{(bc)} = \varkappa_{bc} |d_{bc}/\hbar|^2 N_{bc}/2, \quad \Gamma_{ca}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 N_{ba}/2.$$

3. V-configuration $E_b < E_a < E_c$ with $v > \omega_{ca}$ (Fig. 2c):

$$\Gamma_{ab}^{(ab)} = \varkappa_{ab} |d_{ab}/\hbar|^2 (N_{ab} + 1/2). \quad \Gamma_{ab}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 N_{cb}/2.$$

$$\Gamma_{ab}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 (N_p + 1)/2\hbar^2.$$

$$\Gamma_{cb}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 (N_{cb} + 1/2), \quad \Gamma_{cb}^{(ab)} = \varkappa_{ab} |d_{ab}/\hbar|^2 N_{ab}/2,$$

$$\Gamma_{cb}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 N_p/2\hbar^2, \quad \Gamma_{ca}^{(ca)}$$
$$= \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 (N_p + 1/2)/\hbar^2,$$

$$\Gamma_{ca}^{(cb)} = \varkappa_{bc} |d_{bc}/\hbar|^2 (N_{cb} + 1)/2, \quad \Gamma_{ca}^{(ab)} = \varkappa_{ab} |d_{ab}/\hbar|^2 (N_{ab} + 1)/2.$$

4. V-configuration $E_b < E_a < E_c$ with $v < \omega_{ca}$ (Fig. 2d):

$$\Gamma_{cb}^{(ab)} = \varkappa_{ab} |d_{ab}/\hbar|^2 (N_{ab} + 1/2), \quad \Gamma_{ab}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 N_{cb}/2,$$

$$\Gamma_{ab}^{(ca)} = (\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 N_q + \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 N_\theta)/2\hbar^2,$$

$$\Gamma_{cb}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 (N_{cb} + 1/2), \quad \Gamma_{cb}^{(ab)} = \varkappa_{ab} |d_{ab}/\hbar|^2 N_{ab}/2,$$

$$\Gamma_{cb}^{(ca)} = [\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 (N_q + 1) + \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 (N_\theta + 1)]/2\hbar^2,$$

$$\Gamma_{ca}^{(ca)} = [\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 (N_q + 1/2)$$
$$+ \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 (N_\theta + 1/2)]/\hbar^2$$

$$\Gamma_{ca}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 (N_{cb} + 1)/2, \quad \Gamma_{ca}^{(ab)} = \varkappa_{ab} |d_{ab}/\hbar|^2 (N_{ab} + 1)/2,$$

5. θ-configuration $E_a < E_b < E_c$ with $v > \omega_{ca}$ (Fig. 2e):

$$\Gamma_{ba}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba} + 1/2), \quad \Gamma_{ba}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 N_{cb}/2,$$

$$\Gamma_{ba}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 (N_p + 1)/2\hbar^2,$$

$$\Gamma_{cb}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 (N_{cb} + 1/2), \quad \Gamma_{cb}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba} + 1)/2,$$

$$\Gamma_{cb}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 N_p/2\hbar^2,$$

$$\Gamma_{ca}^{(ca)} = \varkappa_p |\mathscr{E}\Pi_{ac}(-v)|^2 (N_p + 1/2)/\hbar^2,$$

$$\Gamma_{ca}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 (N_{cb} + 1)/2, \quad \Gamma_{ca}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 N_{ba}/2,$$

6. θ-configuration $E_a < E_b < E_c$ with $v < \omega_{ca}$ (Fig. 2f):

$$\Gamma_{ba}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba}+1/2), \quad \Gamma_{ba}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 N_{cb}/2,$$

$$\Gamma_{ba}^{(ca)} = [\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 N_q + \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 N_\theta]/2\hbar^2,$$

$$\Gamma_{cb}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 (N_{cb}+1/2), \quad \Gamma_{cb}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 (N_{ba}+1)/2,$$

$$\Gamma_{cb}^{(ca)} = [\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 (N_q+1) + \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 (N_\theta+1)]/2\hbar^2,$$

$$\Gamma_{ca}^{(ca)} = [\varkappa_q |\mathscr{E}\Pi_{ca}(-v)|^2 (N_q+1/2)$$

$$+ \varkappa_\theta |\mathscr{E}\Pi_{ca}(v)|^2 (N_\theta+1/2)]/\hbar^2,$$

$$\Gamma_{ca}^{(cb)} = \varkappa_{cb} |d_{cb}/\hbar|^2 (N_{cb}+1)/2, \quad \Gamma_{ca}^{(ba)} = \varkappa_{ba} |d_{ba}/\hbar|^2 N_{ba}/2,$$

5. DISCUSSION

At present the most effective methods of studying atomic relaxation are those based on the photon echo phenomenon.[15,16] In analyzing two-photon relaxation it has proved expedient to employ echo phenomena that manifest themselves against the background of nonresonant illumination, since if a two-photon transition $E_c \to E_a$ is in some way excited in advance ($\tilde\rho_{ca} \neq 0$), any probing nonresonant wave with an electric field strength

$$E_0 = \mathscr{E}_0 \exp\{i(k_0 z - \omega_0 t)\} + \text{c.c.} \tag{26}$$

experiences Raman scattering, that is, radiation appears at $|\omega_0 \pm \omega_{ca}|$.[5] If the duration and intensity of (26) are sufficient for the spontaneous processes discussed above to manifest themselves, the Raman scattering signals will strongly depend on the respective relaxation parameters. A simple example is the Raman scattering of light in coherent excitation of three-level systems. References 3 and 17 show that when two light pulses (of duration τ_1 and τ_2 and separated by a time interval τ)

$$E_1 = \mathscr{E}_1 \exp\{i(k_1 z - \omega_1 t - \varphi_1)\} + \text{c.c.}, \quad 0 \leqslant t - z/c \leqslant \tau_1,$$
$$E_2 = \mathscr{E}_2 \exp\{i(k_2 z - \omega_2 t - \varphi_2)\} + \text{c.c.}, \quad \tau_1 + \tau \leqslant t - z/c \leqslant \tau_1 + \tau_2 + \tau,$$

act on a gaseous medium in conditions where the first pulse is in resonance with the optically allowed transition $E_a \to E_b$ and the second is in resonance with the adjacent optically allowed transition $E_c \to E_b$ (the V-configuration of the three-level system; $\omega_1 \approx \omega_{ab}$ and $\omega_2 \approx \omega_{cb}$), this induces specific Raman scattering on the nonresonance wave (26). Raman scattering at the Stokes frequency $\omega_- = \omega_2 - \omega_1 - \omega_0$ and the anti-Stokes frequency $\omega_+ = \omega_2 - \omega_1 + \omega_0$ emerges, after the pulses E_1 and E_2 have passed, at time

$$t - \frac{z}{c} = t_p = \frac{\omega_2}{\omega_2 - \omega_1}\left(\tau + \frac{\tau_2}{2}\right) + \frac{2\omega_2 - \omega_1}{2(\omega_2 - \omega_1)}\tau_1$$

and is determined by the following value $E_\pm$ of the electric field strength of the anti-Stokes (upper sign) and Stokes signals:

$$E_\pm = \varepsilon_\pm (t - z/c) \exp[i(k_\pm z - \omega_\pm t - \varphi_\pm)] + \text{c.c.}, \tag{27}$$

$$\varepsilon_\pm (t) = e_\pm' (t)\mathscr{E}_0 \Pi_{\mp\omega_0} \exp[-\gamma_{ab}\tau - \gamma_{ca}(t_p - \tau)]. \tag{28}$$

Here $e_\pm'$ is independent of the field strength (26) and for small areas of the exciting pulsed fields E_1 and E_2 is given in Ref. 3 ($e_\pm' = e_\pm /\mathscr{E}_0$). It is assumed that the nonresonant field (26) is switched on no later than when the second exciting pulse has ceased to act.

If there are no vacuum photons ($N_{ab} = N_{cb} = N_{ca} = 0$) and the Stark shifts of levels are ignored, the relaxation constants γ_{ab} and γ_{ca} have the following form:

$$\gamma_{ab} = \gamma_{ab}^{(0)} + \gamma_{ab}^{(1)}, \quad \gamma_{ca} = \gamma_{ca}^{(0)} + \gamma_{ca}^{(1)} + \gamma_{ca}^{(2)},$$

$$\gamma_{ab}^{(1)} = \omega_{ab}^3 |d_{ab}|^2/\hbar c^3, \quad \gamma_{ca}^{(1)} = \gamma_{ab}^{(1)} + \omega_{cb}^3 |d_{cb}|^2/\hbar c^3,$$

$$\gamma_{ca}^{(2)} = 2\pi I_0 [\omega_+^3 |\Pi_{ca}(-\omega_0)|^2 + \omega_-^3 |\Pi_{ca}(\omega_0)|^2)/\hbar c^4,$$

where the constants $\gamma_{ab}^{(0)}$ and $\gamma_{ca}^{(0)}$ are determined by atomic collisions and are independent of the intensity of the light fields, $\gamma_{ab}^{(1)}$ and $\gamma_{ca}^{(1)}$ describe ordinary spontaneous relaxation, $\gamma_{ca}^{(2)}$ corresponds to the present mechanism of two-photon spontaneous relaxation with a photon from the monochromatic wave (26) participating in the process, and I_0 is the intensity of wave (26) averaged over the period of rapid oscillations, $I_0 = c|\mathscr{E}_0|^2/2\pi$.

It is easy to see that studying $\log(I_\pm /I_0)$ as a function of I_0 in experiments makes it possible to avoid determining $2\pi(\omega_+^3 |\Pi_{ca}(-\omega_0)|^2 + \omega_-^3 |\Pi_{ca}(\omega_0)|^2)/\hbar c^4$ and to verify the importance of the two-photon relaxation mechanism. Experiments involving monochromatic waves of the form (26) at different frequencies ω_0 and other echo effects should make it possible to separately estimate the parameters $|\Pi_{ca}(-\omega_0)|$ and $|\Pi_{ca}(\omega_0)|$ of the operator of two-photon interaction. When the Stark shifts of levels cannot be ignored, an additional relaxation constant $\Gamma_{\alpha\beta}$ emerges, whose study provides information about $|\Pi_\alpha(\omega_0) - \Pi_\beta(\omega_0)|$.

As for the order of magnitude of $\gamma_{ca}^{(2)}$, for the $4^2S_{1/2} \to 3^2S_{1/2}$ two-photon transition in sodium, with a 10^{12} s^{-1} detuning of the nonresonance wave from the $3^2P_{1/2}^0$ level, we find $\gamma_{ca}^{(2)} \approx 4 \times 10^{-5} I_0$ s^{-1} in the esu system, that is, two-photon spontaneous relaxation manifests itself in megawatt fields. Two-photon relaxation may also prove significant at lower intensities of the external monochromatic field if noise fields are employed at frequencies of the vacuum photons emitted in the relaxation process.

Note that two-photon relaxation depends on the type of two-photon transition and the polarization of the monochromatic field, since linearly and circularly polarized waves scatter differently.

The above method also makes it easy to describe other two-photon relaxation processes in nonresonant light fields, say, in a nonresonant noise electromagnetic field [which essentially is determined by the constants in (25)] or in a squeezed field.

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