Photon frequency redistribution function in the resonance radiation transfer theory in the presence of a laser field

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Using the atom-photon density matrix formalism in the resonant radiation transfer theory, we obtain explicit expressions for the photon frequency redistribution function in the presence of a laser field. We show that the presence of the laser field significantly changes not only the shape of the redistribution function, but also its structure. We also show that the four-wave interaction, which occurs in this case, has an important effect on the reabsorption of the radiation.

1. INTRODUCTION

The problem of the transfer of resonant radiation in the presence of a laser field has been studied in a number of papers.¹⁻⁴ The problem was solved in Refs. 2 and 3 by using the approximation that the saturation parameter is small: $\Omega/\gamma \ll 1$, where Ω is the Rabi frequency for the amplitude of the laser field strength and γ is the spontaneous relaxation rate. The assumption that this parameter is small was not made in Ref. 4, but this paper did not obtain an explicit expression for the frequency redistribution function of the noise photons (we call it simply the rescattering function in what follows) which is very important and convenient for a practical description of radiative transfer. Keldysh's diagram technique for nonequilibrium Green's functions,³ which was used in Ref. 4, had earlier been applied successfully in Refs. 6 to 8 for a description of radiation (or phonon) transfer. An important difference between the problem of radiative transfer in the presence of a laser field and the usual radiative transfer problem is that the structure of the radiation is significantly more complicated-anomalous correlators appear corresponding to four-wave interaction processes. Moreover, the single-loop approximation turns out to be insufficient for the description of the polarization and mass operators.⁴ Because of these purely technical difficulties no analytical expression was found in Ref. 4 for the rescattering function, although the basic processes were studied.

In the present paper we use Scully and Lamb's atomphoton density matrix formalism⁹ to describe the transfer processes and obtain the rescattering function. This description was developed in Ref. 10 in a form which is closest to the present problem; in those papers the problem was studied of a quantum description of several modes of a resonator in which a resonant gaseous medium was present. Note that the description used is close to the perturbation theory method for quantum fields in the density matrix framework for compound systems.¹¹ This approach has been used to calculate the rescattering function in Refs. 12 and 13.

2. BASIC EQUATIONS

We write the Hamiltonian of this system in the form

$$\dot{H} = \dot{H}_0 + \dot{H}_r + \dot{H}_i + \dot{H}_c. \tag{1}$$

where the first two terms describe the Hamiltonian of the unperturbed atomic subsystem and of the free radiation field (in rad/s):

$$\hat{H}_{o} = -\Delta_{L}\sigma_{z}, \quad \hat{H}_{r} = \sum_{k} v_{k}a_{k}^{+}a_{k}.$$
(2)

Here a_k^+ and a_k are the creation and annihilation operators for photons with wavevector **k** and frequency ω_k and the σ are the usual Pauli matrices. The atom and photon energies are reckoned from the energy of the quanta of the monochromatic electromagnetic laser wave with frequency ω_L : $v_k = \omega_k - \omega_L$; $\Delta_L = \omega_L - \omega_0$, where ω_0 is the transition frequency in the atomic subsystem. The last two terms in Eq. (1) describe the interaction of the atomic subsystem with the laser wave and the quantized radiation field. Using the rotating wave approximation we write them in the form

$$\hat{H}_i = \sigma^- V + \sigma^+ V^*, \tag{3}$$

$$\hat{H}_{c} = g \sum_{k} (\sigma^{+} a_{k} + \sigma^{-} a_{k}^{-}), \qquad (4)$$

where we have $V = -\mu E_L/2\hbar$, E_L is the amplitude of the laser wave field strength, μ the dipole moment matrix element for the $1 \rightarrow 2$ transition, and $g = \mu (2\pi\omega_k/\hbar L^3)^{1/2}$ is the coupling constant (vacuum Rabi frequency), which we assume to be the same for all modes, and L^3 is the quantization volume.

The equation for the atom-photon density matrix has the form

$$i\dot{\rho}_{\mathrm{at.ph.}} = [H;\rho_{\mathrm{at.ph.}}] + i\Gamma(\rho_{\mathrm{at.ph.}}), \qquad (5)$$

where $\Gamma_{at.ph.}$ is an operator describing relaxation processes. Contracting in (5) over the photon variables we get an equation for the atomic density matrix ρ and, on the other hand, taking the trace over the atomic states we have an equation for the photon field operator *P*. As a first step we follow Ref. 10 and write the atom-photon density matrix in factored form: $\rho_{at.ph.} = P\rho$. We neglect here effects connected with radiation capture and also with the saturation of the atomic subsystem by quantized fields (it is natural to assume them to be small) and we arrive then at the results of Ref. 10 and particularly of Ref. 4. In that case we have for the atomic density matrix the usual equation:

$$i\dot{\rho} = [H_0 + H_i; \rho] + i\Gamma(\rho). \tag{6}$$

Assuming that the amplitudes of all fields change little during the characteristic lifetime of the atomic subsystem we can find a stationary solution of Eq. (6) and afterwards obtain an equation for the photon field operator P. It was

shown in Refs. 4 and 10 that apart from the usual absorption and emission processes of the quantized field there are processes connected with four-wave interactions: $\mathbf{k} + \bar{\mathbf{k}} = 2\mathbf{k}_L$, $\omega_k + \omega_{\bar{k}} = 2\omega_L$, i.e., two photons of the pump wave are transformed into two photons of the quantized field. The equation for the two-mode photon field operator then has the form¹⁰

$$\dot{P} = [A_{k}(a_{k}^{+}Pa_{k}^{-}-Pa_{k}a_{k}^{+}) + B_{k}(a_{k}Pa_{k}^{+}-a_{k}^{+}a_{k}^{+}P)
+ C_{k}(a_{k}^{+}a_{\bar{k}}^{+}P-a_{\bar{k}}^{+}Pa_{k}^{+})
+ D_{k}(Pa_{\bar{k}}^{+}a_{k}^{+}-a_{\bar{k}}^{+}Pa_{k}^{+}) + \text{h.c.}] + (k \leftrightarrow \bar{k}).$$
(7)

The expression for the photon occupation numbers is determined by the relation $n = \langle a^+ aP \rangle$. Moreover, it has been shown in Refs. 10 and 4 that in this case there appear new correlators: $\langle a_k a_{\bar{k}} \rangle = \langle a_k a_{\bar{k}} P \rangle$ and $\langle a_k^+ a_{\bar{k}}^+ \rangle = \langle a_k^+ a_{\bar{k}}^+ P \rangle$. Following Ref. 10 we shall call them combination tone operators. Their appearance is connected with four-wave interaction processes. From Eq. (7) we get the equation of motion for the photon occupation numbers $\dot{n} = \langle a^+ a\dot{P} \rangle$ (and similarly for the combination tone operators):

$$\frac{d}{dt}n_{k} = (A_{k} - B_{k})n_{k} + (C_{k} - D_{k}) \langle a_{k}a_{\bar{k}} \rangle + A_{k} + \text{h.c.}, \qquad (8)$$

$$\frac{d}{dt} \langle a_k a_{\bar{k}} \rangle = (A_k + A_{\bar{k}} - B_k - B_{\bar{k}}) \langle a_k a_{\bar{k}} \rangle + (C_k - D_k) n_{\bar{k}} + (C_{\bar{k}} - D_{\bar{k}}) n_k + C_k + C_{\bar{k}}.$$
(9)

It is clear from these expressions that the coefficients B and A describe the absorption (amplification) of photons, C and D describe four-wave coupling of modes, while A and C are spontaneous sources for photon occupation numbers and for combination tone operators, respectively. The structure of these coefficients has been well studied: the absorption and four-wave coupling coefficients are the same as the corresponding coefficients for classical test signals, symmetrically tuned in frequency from the pump wave, while the expression for A is described by the resonance fluorescence spectrum of a two-level atom. The spontaneous source for the combination tone was studied in detail in papers by the authors of Ref. 10, who showed, in particular, that this quantity is extremely important for the description of squeezed states in the resonance four-wave interaction mechanism.¹⁴

It has been noted earlier⁴ that Eqs. (6), (8), and (9) are not sufficient for describing the transfer of resonance radiation in the presence of a laser field: the appropriate equations must be linear in the photon occupation numbers (and in the combination tone operators, if wave synchronism is satisfied). Equations (8) and (9) contain spontaneous sources in zeroth order in those quantities and for V = 0, for instance, they do not even describe the usual radiative transfer.

It is thus necessary to supplement Eqs. (8) and (9) with expressions for spontaneous sources which are linear in the photon occupation numbers and in the combination tone operators. This problem is not solved by just taking into account in the simplest way the saturation of the atomic subsystem by the quantized fields (this part of the problem is completely solved in Ref. 4). The coherence introduced by the laser field significantly changes the emission spectrum of the atom and cannot be represented, as is done in the description of radiative transfer with a complete mixing in frequency, in the form of the product of the population of the excited level and the contour of the emission lines.^{15,16}

3. EQUATIONS FOR THE OPERATORS OF THE ATOMIC AND PHOTON SUBSYSTEMS

To solve our problem we must generalize the photon operator P from the single- or two-mode case, considered in Ref. 10, to a continuous photon distribution.

In the single-mode case we have

$$P(\mathbf{k}) = \sum_{n} P_{n} | n \rangle \langle n |, \quad \sum_{n} P_{n} = 1.$$
(10)

In the two-mode case we have

$$P(\mathbf{k}, \ \overline{\mathbf{k}}) = \sum_{n_{\mathbf{k}}, \ n_{\mathbf{k}}} \left(P_{n_{\mathbf{k}}n_{\overline{k}}} \mid n_{\mathbf{k}} \right) \langle n_{\overline{k}} \mid \\ + P_{n_{\mathbf{k}}} P_{n_{\overline{k}}} \mid n_{\mathbf{k}} \rangle \langle n_{\mathbf{k}} \mid \mid n_{\overline{k}} \rangle \langle n_{\overline{k}} \mid).$$

$$(11)$$

We define the multimode photon operator \mathscr{P} as the product of two-mode operators (11) or single-mode (10) operators, if for a given wavevector there are no waves satisfying the wave synchronism condition (note that we use the resonance approximation):

$$\mathscr{P} = \prod_{\mathbf{k},\bar{\mathbf{k}}} P(\mathbf{k},\bar{\mathbf{k}}).$$
(12)

In the present case we assume that coherence (i.e., offdiagonal matrix elements) occurs only for those pairs of modes which satisfy wave synchronism. Coherence for modes which do not satisfy this condition or coherence between more than two modes corresponds to correlators of an order higher than the second in the photon creation and annihilation operators and that goes beyond the framework of our considerations.

The single-mode photon field operator is obtained by summing over all other modes, i.e., $P(\mathbf{k}) = \operatorname{Sp}_{\mathbf{k}' \neq \mathbf{k}}(\mathscr{P})$.

The equations for \mathcal{P} have the form

$$i\dot{\mathscr{P}} = [H_r, \mathscr{P}] + \operatorname{Sp}_{a}([H_r, \rho_{\mathrm{at. ph.}}]), \qquad (13)$$

$$i\dot{\mathscr{P}} = [H_r, \mathscr{P}] + g \operatorname{Sp}_{\mathfrak{a}} \left(\sum_{\mathbf{k}} (\sigma^+ F_k^- + \sigma^- F_k^+) - \mathrm{h.c.} \right), \quad (14)$$

where we have followed Ref. 10 and introduced auxiliary operators $F_k^+ = a_k^+ \rho_{\text{at.ph.}}$ and $F_k^- = a_k \rho_{\text{at.ph.}}$ and the subscript of the trace sign indicates summation over atomic states. In the present case these operators are functions of an infinite number of variables (since the atom-photon density matrix contains a continuous photon distribution).

The equation for the F_k^- operator has the form

$$i\dot{F}_{k} = [\hat{V}_{j}, F_{k}] + \omega_{k}F_{k} + g\sigma^{-}\rho_{\text{at. ph.}} + [\hat{H}_{c}, F_{k}] + i\Gamma(F_{k}),$$
(15)

where $\hat{V}_j = \hat{H}_0 + \hat{H}_i$. We get the single-mode (or twomode) operator by contracting over all photons with wavevectors $\mathbf{k}' \neq \mathbf{k} (\mathbf{k}' \neq \mathbf{\bar{k}}, \mathbf{\bar{k}} + \mathbf{k} = 2\mathbf{k}_L)$:

$$G_k = \operatorname{Sp}_{k' \neq k}(F_k). \tag{16}$$

Note that these operators differ from the purely two- or single-mode operators Φ_k which were considered in Ref. 10, since they contain corrections connected with the rescattering of the radiation from other modes or with the parametric action of these modes on the given one. These corrections are important for calculating the rescattering function.

The equation for the atomic density matrix has the form

$$i\rho = [\hat{V}_{i}, \rho] + \operatorname{Sp}_{ph.}([H_{e}, \rho_{at. ph.}]) + i\Gamma(\rho).$$
(17)

One can easily transform this equation to the form

$$i\dot{\rho} = [\hat{\nu}_{j}, \rho] + \sum_{k} g(\sigma^{+}G_{k}^{-} + \sigma^{-}G_{k}^{+} - \text{h.c.}).$$
 (18)

To solve the problem posed in the present paper we must obtain equations for the atomic density matrix which are linear in the photon occupation numbers and in the combination tone operators. This means that we must use in Eq. (18) instead of the G_k operators the Φ_k operators, since taking these corrections into account in this case would exceed our accuracy. The equations for the components of the atomic density matrix thus have the form

$$i\dot{\rho}_{22} = -i\gamma_{1}\rho_{22} + V^{*}\rho_{12} - \rho_{21}V + \sum_{k} g(\langle \Phi_{k12}^{+} \rangle - \langle (\Phi_{k}^{-}) \rangle_{21}^{+} \rangle),$$

$$i\dot{\rho}_{11} = i\gamma_{1}\rho_{22} + V\rho_{21} - \rho_{12}V^{*} + \sum_{k} g(\langle \Phi_{k21}^{+} \rangle - \langle (\Phi_{k}^{+}) \rangle_{12}^{+} \rangle),$$

$$i\dot{\rho}_{12} = \Delta^{*}\rho_{12} + V(\rho_{22} - \rho_{11}) + \sum_{k} g(\langle \Phi_{k22}^{+} \rangle - \langle (\Phi_{k}^{-}) \rangle_{11}^{+} \rangle),$$

$$i\dot{\rho}_{24} = -\Delta\rho_{24} + V^{*}(\rho_{11} - \rho_{22}) + \sum_{k} g(\langle \Phi_{k11}^{-} \rangle - \langle (\Phi_{k}^{+}) \rangle_{22}^{+} \rangle),$$

(19)

where $\langle \Phi_k \rangle$ equals $\langle \mathbf{k} | \Phi_k | \mathbf{k} \rangle$ or $\langle \mathbf{k} | \Phi_k | \overline{\mathbf{k}} \rangle$. Assuming the factorization of the atom-photon density matrix in the third and fourth terms of Eq. (15), we get from Eqs. (15) and (16) the evolution equation for the Φ_k^- operator of Ref. 10:

$$i \Phi_k^- = \mathbf{M} \Phi_k^- + \mathbf{S}^-, \tag{20}$$

We understand here by Φ_k^- the column of the projection of that operator on the atomic subsystem states:

$$\mathbf{\Phi}_{k}^{-} = \begin{vmatrix} \mathbf{\Phi}_{22k}^{-} \\ \mathbf{\Phi}_{11k}^{-} \\ \mathbf{\Phi}_{12k}^{-} \\ \mathbf{\Phi}_{21k}^{-} \end{vmatrix}; \quad \mathbf{M}(\mathbf{v}) = \begin{vmatrix} \mathbf{v} - i \mathbf{\gamma}_{1} & 0 & V^{*} & -V \\ i \mathbf{\gamma}_{1} & \mathbf{v} & -V^{*} & V \\ V & -V & \mathbf{v} + \Delta^{*} & 0 \\ -V^{*} & V^{*} & 0 & \mathbf{v} - \Delta \end{vmatrix},$$

$$S^{-} = g \begin{vmatrix} a_{\bar{k}} a_{\bar{k}} P(\rho_{12}^{(0)} - a_{\bar{k}} P a_{\bar{k}}^{+} \rho_{21}^{(0)} \\ - a_{\bar{k}} P a_{\bar{k}} \rho_{12}^{(0)} + a_{\bar{k}} a_{\bar{k}}^{+} P \rho_{21}^{(0)} \\ - a_{\bar{k}} P a_{\bar{k}} \rho_{11}^{(0)} + a_{\bar{k}} a_{\bar{k}}^{+} P \rho_{22}^{(0)} \\ - a_{\bar{k}} P a_{\bar{k}} \rho_{22}^{(0)} + a_{\bar{k}} a_{\bar{k}} P \rho_{11}^{(0)} \end{vmatrix},$$

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where the $\rho_{jk}^{(0)}$ are the stationary solutions of the equations for the atomic density matrix (6), the exact expressions of which are given in the Appendix; we have $\Delta = \Delta_L + i\gamma_2$ and γ_1 and γ_2 are the longitudinal and transverse relaxation rates (in the radiative regime we have $\gamma_1 = 2\gamma_2 = \gamma$). Assuming that the radiation field changes slowly during the characteristic lifetime of the atomic subsystem we find from the set (20) a stationary solution for the Φ_k operators. The explicit expressions which are important in what follows for the description of the dynamics of the atomic and photon subsystems are given in the Appendix. These expressions determine for the two-mode case the kinetic coefficients in Eqs. (7) to (9).¹⁰ If we write the expression for S^- as a sum:

$$S^{-} = S_{r}^{-} + S_{a}^{-}, \tag{21}$$

where S_r^- contains the elements of the column proportional to $a_k a_k^+ P$ and $a_k P a_{\bar{k}}$, and S_a^- those proportional to $a_k P a_k^+$ and $a_{\bar{k}} a_k P$, the solution Φ_k^- corresponding to S_r^- describes the A_k and C_k coefficients in Eqs. (7) to (9) and the solution corresponding to S_a^- describes the B_k and D_k coefficients. These coefficients have the form (cf. Refs. 4 and 10)

$$A_{k} = Ng^{2}i(\rho_{21}^{(0)} m_{23} - \rho_{22}^{(0)} m_{33}), \ C_{k} = -Ng^{2}i(\rho_{12}^{(0)} m_{23} + \rho_{22}^{(0)} m_{43}),$$

$$B_{k} = -Ng^{2}i(\rho_{21}^{(0)} m_{13} + \rho_{11}^{(0)} m_{33}), \ D_{k} = Ng^{2}i(\rho_{11}^{(0)} m_{43} - \rho_{12}^{(0)} m_{13}),$$
(22)

where N is the number of atoms interacting with the radiation.

4. EQUATIONS FOR THE ATOMIC SUBSYSTEM

Using the explicit form (A1) to (A4) of the Φ_k operators we can write Eqs. (19) for the components of the atomic density matrix to first order in the quantities n_k and $\langle a_k a_{\bar{k}} \rangle$ in the form

$$i\dot{\rho}_{22} = -i\gamma_{1}\rho_{22} + V \cdot \rho_{12} - \rho_{21}V$$

$$+\gamma \int \frac{dv_{k}}{2\pi} (n_{k} [\rho_{21}^{(0)} m_{13}^{22} + (\rho_{11}^{(0)} - \rho_{22}^{(0)}) m_{33}]$$

$$- \langle a_{k}a_{k} \rangle [\rho_{12}^{(0)} m_{14}^{24} + (\rho_{11}^{(0)} - \rho_{22}^{(0)}) m_{34}] - \text{h.c.}), \qquad (23)$$

(23)

$$\begin{split} \dot{\rho}_{21} &= -\Delta \rho_{21} + V^* (\rho_{11} - \rho_{22}) \\ &- \gamma \int \frac{d\nu_k}{2\pi} (2n_k [\rho_{21}^{(0)} m_{12}^{22} + (\rho_{11}^{(0)} - \rho_{22}^{(0)}) m_{32}] \\ &- 2 \langle a_k a_k^* \rangle [\rho_{12} m_{12}^{22} + (\rho_{11}^{(0)} - \rho_{22}^{(0)}) m_{42}]), \end{split}$$

$$(24)$$

where $m_{jk}^{ln} = m_{jk} + m_{ln}$. The explicit form of the expressions for the m_{jk} can be found in the Appendix.

These expressions are the same as the results of Ref. 4 in which equations for the atomic subsystems were obtained which were accurate up to first order in the occupation numbers.

5. EQUATIONS FOR THE ATOM-PHOTON OPERATORS

For a correct description of the radiative transfer we must evaluate the A and C coefficients in Eq. (7) for the photon field operator to first order in the occupation numbers. This equation follows from (14) when we sum it over all modes except those whose dynamics is described by the P operator. For the solution of the problem we need thus find expressions for the G_k operators, which determine the dynamics of the P operator after Eq. (14) has been summed over the other modes, to the same accuracy.

We use as zeroth approximation for the G_k^- quantities– Φ_k the stationary solution of the set (20) with the inhomogeneous column S_r^- which determines the spontaneous sources A and C in that approximation [see (22)]. We then write G_k^- in the form

$$G_{k} = \widetilde{\Phi}_{k} + \chi_{k} + \overline{\Phi}_{k}, \qquad (25)$$

where

$$\chi_{h}^{-} = \sum_{\mathbf{k}'} \theta(\mathbf{k}', \overline{\mathbf{k}}')$$

is the stationary solution of Eq. (15) in the second order of perturbation theory in the coupling constant with zeroth approximation $\tilde{\Phi}_k^-$. The equation for the quantity θ has the form

$$i\dot{\theta} = M(v)\theta + g(\sigma^{+}\psi^{--} + \sigma^{-}\psi^{-+} - \xi^{--}\sigma^{+} - \xi^{-+}\sigma^{-}).$$
 (26)

The quantities ψ^{--} , ψ^{-+} , ξ^{--} , and ξ^{-+} are defined as follows:

$$\psi^{--} = a_{l} a_{k'} \rho_{\text{at. ph.}}, \quad \psi^{-+} = a_{k} a_{k'} \rho_{\text{at. ph.}},$$

$$\xi^{-+} = a_{k} \rho_{\text{at. ph.}} a_{k'} +, \quad \xi^{--} = a_{k} \rho_{\text{at. ph.}} a_{k'},$$

and we have for them the following equations:

$$i\psi^{--} = M(v+v)\psi^{--}$$

$$+g\{\sigma^{-}(a_{k}a_{k}^{+}P\langle\Phi_{k},-\rangle+n_{k'}\Phi_{k}^{-})-n_{k'}\tilde{\Phi}_{k}^{-}\sigma^{-}$$

$$+\sigma^{+}\tilde{\Phi}_{k}^{-}\langle a_{\bar{k}},a_{k'}\rangle-(a_{k}Pa_{\bar{k}}\langle\Phi_{k'},-\rangle+\langle a_{k'}a_{\bar{k}'}\rangle\tilde{\Phi}_{k}^{-})\sigma^{+}\},$$

$$(27)$$

$$i\psi^{-*} = M(v-v')\psi^{-+} + g\{\sigma^{+}n_{k'}\widetilde{\Phi}_{k}^{-}$$

$$+\sigma^{-}(\langle a_{\bar{k}'}^{+}+a_{k'}^{+}\rangle\Phi_{h}^{-}+a_{k}a_{k}^{+}P\langle\Phi_{k'}^{+}\rangle\rangle)$$

$$-(a_{k}Pa_{\bar{k}}\langle\Phi_{k'}^{+}\rangle+n_{k'}\widetilde{\Phi}_{k}^{-})\sigma^{+}-\langle a_{k'}^{+}+a_{\bar{k}'}^{+}+\rangle\Phi_{k}^{-}\sigma^{-}\}.$$

$$(28)$$

$$i\tilde{z}^{-+} = M(v-v')\tilde{z}^{-+}+g\{\sigma^{+}n_{k'}\widetilde{\Phi}_{k'}^{-}-$$

$$+\sigma^{-}(\langle a_{\bar{k}'}^{+}a_{k'}^{+}\rangle\widetilde{\Phi}_{k}^{-}+a_{k}a_{k}^{+}P\langle(\Phi_{k'}^{-})^{+}\rangle) \\ -(a_{k}Pa_{\bar{k}}\langle(\Phi_{k'}^{-})^{+}\rangle+n_{k'}\widetilde{\Phi}_{k}^{-})\sigma^{+}-\langle a_{k'}^{+}a_{\bar{k}'}^{+}\rangle\widetilde{\Phi}_{k}^{-}\sigma^{-}\},$$

$$(29)$$

$$i\xi^{--} = M(v+v')\xi^{--} + g\{\sigma^{+}\langle a_{\overline{k}}, a_{k'}\rangle \widetilde{\Phi}_{k}^{-} \\ +\sigma^{-}(n_{k'}\widetilde{\Phi}_{k}^{-} + a_{k}a_{k}^{+}P\langle (\Phi_{k'}^{+})^{+}\rangle) \\ -(a_{k}Pa_{\overline{k}}\langle (\Phi_{k'}^{+})^{+}\rangle + \langle a_{k'}a_{\overline{k}}, \rangle \widetilde{\Phi}_{k}^{-})\sigma^{+} - n_{k'}\widetilde{\Phi}_{k}^{-}\sigma^{-}\}.$$
(30)

Here $\langle \Phi_{k'}^- \rangle$ equals $\langle \mathbf{k}' | \Phi_{k'}^- | \mathbf{k}' \rangle$ or $\langle \mathbf{k}' | \Phi_{k'}^- | \mathbf{\bar{k}}' \rangle$, where $\Phi_{k'}^-$ is the solution of the set (20) for the mode \mathbf{k}' ; $\langle \Phi_{k'}^- \rangle$ is thus proportional to n_k , or to $\langle a_k, a_{\bar{k}}, \rangle$.

The quantity $\overline{\Phi}_k^-$, which is the third term in Eq. (25) for G_k^- is, like $\widetilde{\Phi}_k^-$ a solution of the set (20) with the same column S_r^- but the components of the atomic density ma-

trix $\rho_{jk}^{(0)}$ are replaced by $\bar{\rho}_{jk}$. These are the stationary solutions of the perturbation theory set of Eqs. (23) and (24) with zeroth approximation $\rho_{jk}^{(0)}$. The explicit expressions for them have the form

$$\bar{\rho}_{22} = \frac{i(V^{*}\Delta I_{12}^{*} - V\Delta^{*}I_{21}^{*} - I_{22}|\Delta|^{2})}{\gamma_{1}|\Delta|^{2} + 4|V|^{2}\gamma_{2}},$$

$$\bar{\rho}_{12} = -(2V\bar{\rho}_{22}^{*} + I_{12})/\Delta^{*}, \quad \bar{\rho}_{21} = \bar{\rho}_{12}^{*},$$

(31)

where the I_{jk} are the integral terms in the expressions for ρ_{jk} of Eqs. (23) and (24).

The expression for G_k^- obtained in this way makes it possible to describe expressions for the spontaneous sources A and C with the required accuracy.

6. RESCATTERING FUNCTION

We can now write the expression for the spontaneous source A for the photon occupation numbers (C for the combination tone operators) as a sum

$$A = A_0 + A_1 + A_2, \quad C = C_0 + C_1 + C_2, \quad (32)$$

where A_0 and C_0 are proportional to $\overline{\Phi}_k^-$ and are given by Eqs. (22), a result obtained before.¹⁰ The quantities A_1 and C_1 (A_2 and C_2) which are, respectively, proportional to $\chi_k^$ and $\overline{\Phi}_k^-$ describe spontaneous sources associated with the rescattering from other modes. One can interpret the first of these as a characteristic connected with parametric (combination) processes and the second we connect with the saturation of the atomic subsystem by scattered photons.

We now introduce the rescattering functions for the quantities A and C as follows:

$$A_{k} = A_{0} + Ng^{2} \int (n_{k} \cdot R(n_{n} \cdot) + \langle a_{n} \cdot a_{\overline{n}} \cdot \rangle R(\langle a_{k} \cdot a_{\overline{n}} \cdot \rangle)$$

+ $\langle a_{k} \cdot a_{\overline{n}} \cdot \rangle R(\langle a_{k} \cdot a_{\overline{n}} \cdot \rangle)) \frac{dv'}{2\pi},$ (33)

$$C_{k} = C_{6} + Ng^{2} \int (n_{k'}Q(n_{k'}) + \langle a_{k'}a_{\overline{k'}}\rangle Q(\langle u_{k'}a_{\overline{k'}}\rangle) + \langle a_{k'}^{+}a_{\overline{k'}}^{+} + \rangle Q(\langle a_{k'}^{+}a_{\overline{k'}}^{+} \rangle)) \frac{dv'}{2\pi}.$$
(34)

Like Eq. (31) we write the rescattering functions as a sum:

$$R = R_1 + R_2, \quad Q = Q_1 + Q_2.$$

The explicit expressions for the rescattering functions in the spontaneous source for the photon occupation numbers have the form

$$R_{1}(n_{k'}) = i\gamma \left(\left[m_{24}(v) \rho_{21} - m_{32}^{31}(v) \rho_{22} \right] \right] \times \left(m_{13}^{23}(v) m_{44}(v-v') + m_{14}^{24}(v-v') m_{33}(v) \right) + \left[m_{23}(v) \rho_{21} - m_{33}(v) \rho_{22} \right] \left(m_{13}^{23}(v) m_{14}^{24}(v-v') + 2m_{12}^{22}(v-v') m_{33}(v) \right) - \left[m_{24}^{44}(-v') \rho_{12} - m_{44}(-v') (\rho_{11} - \rho_{22}) \right] \times \left(m_{13}^{23}(v) m_{24}(v-v') + m_{21}^{22}(v-v') m_{33}(v) \right) + \left[- m_{11}^{23}(-v') \rho_{12} + m_{41}(-v') (\rho_{11} - \rho_{22}) \right] \times \left(m_{13}^{23}(v) m_{34}(v-v') + m_{31}^{32}(v-v') m_{33}(v) \right) + \left[m_{34}(v) \rho_{22} - m_{22}^{21}(v) \rho_{21} \right] \left(- m_{13}^{23}(v) m_{23}^{13}(v+v') + 2m_{21}^{22}(v+v') m_{34}(v) \right) + \left[m_{14}^{24}(v') \rho_{21} + m_{44}(v') (\rho_{11} - \rho_{22}) \right] \right]$$

$$\times (m_{13}^{23}(v)m_{23}(v+v')+m_{22}^{21}(v+v')m_{34}(v)) + [(m_{14}^{21}(v')+m_{22}^{21}(v))\rho_{21}-m_{32}^{31}(v)\rho_{22}+m_{34}(v')(\rho_{11}-\rho_{22})] \times (m_{13}^{23}(v)m_{33}(v+v')+m_{13}^{23}(v+v')m_{34}(v))-c.c.), R_{2}(n_{k'}) = \gamma ((m_{33}(v)+2V^{*}m_{23}(v)/\Delta) \times (V\Delta^{*}[\rho_{21}m_{12}^{22}(v')+(\rho_{11}-\rho_{22})m_{32}(v')] + |\Delta|^{2}[\rho_{24}m_{13}^{23}(v')+(\rho_{11}-\rho_{22})m_{33}(v')]-c.c.]/ (4|V|^{2}\gamma_{2}+|\Delta|^{2}\gamma_{1}) +im_{23}(v)[\rho_{24}m_{13}^{23}(v')+(\rho_{11}-\rho_{22})m_{33}(v')]/\Delta+c.c.).$$

Here the ρ_{jk} are the stationary solutions of the equations for the atomic density matrix (6) (before we used the notation $\rho_{jk}^{(0)}$). Explicit expressions for the other functions in Eqs. (33) and (34) can be found in Ref. 17.

The expressions (35) given here for the rescattering function in the spontaneous source for the photon occupation numbers are the main characteristic in the description of the transfer, since they describe the action of quantities with an arbitrary angular distribution and therefore give the main contribution, in contrast to the contribution from the combination tone operators whose angular distribution is strongly restricted by the wave synchronism condition.

For $V \rightarrow 0$ (i.e., without a laser field) Eqs. (35) for the rescattering function change, apart from a normalizing factor $(2\pi)^2$, into the well known result:¹⁸

$$R_{1} = \frac{i\gamma}{|\nu'+\Delta|^{2}} \left(\frac{1}{\nu-\nu'+i0} - \frac{1}{\nu+\Delta} \right) + \text{c.c.},$$

$$R_{2} = \frac{2i\gamma_{2}\gamma}{\gamma_{1}|\nu'+\Delta|^{2}(\nu+\Delta)} + \text{c.c.},$$
(36)

and in weak fields, $V \ll \gamma$, agree with the results of Ref. 2. In strong fields, $V \gg |\Delta|$, the rescattering function has the following asymptotic form:

$$R \approx i\gamma ((\alpha_{+}(\nu)/2 + \alpha_{-}(\nu)/2 - \alpha_{0}(\nu))^{2} (\alpha_{+}(\nu - \nu') + \alpha_{-}(\nu - \nu')) - \frac{1}{2} \alpha_{0}(\nu) + \alpha_{+}(\nu)/2 + \alpha_{-}(\nu)/2] [(\alpha_{+}(\nu) - \alpha_{-}(\nu)) (\alpha_{+}(\nu - \nu') - \alpha_{-}(\nu - \nu'))] - \alpha_{-}(\nu) (\alpha_{+}(\nu - \nu') + \alpha_{-}(\nu - \nu'))] - [\alpha_{+}(\nu) - \alpha_{-}(\nu)] [(\alpha_{+}(\nu) - \alpha_{-}(\nu)) (\alpha_{0}(\nu - \nu') + \alpha_{+}(\nu - \nu')/2] + \alpha_{-}(\nu - \nu')/2 + \alpha_{0}(\nu + \nu') + \alpha_{+}(\nu + \nu')/2 + \alpha_{-}(\nu + \nu')/2) + (\alpha_{+}(\nu + \nu') - \alpha_{-}(\nu + \nu')) (\alpha_{+}(\nu) + \alpha_{-}(\nu) - 2\alpha_{0}(\nu))] - c.c.),$$

where

$$\alpha_{\circ}(\mathbf{v}) = \frac{1/2}{\mathbf{v} - i\gamma_{2}}, \quad \alpha_{\pm}(\mathbf{v}) = \frac{1/2}{\mathbf{v} \pm \Omega - i\Gamma}, \quad \Gamma = \frac{1}{2}(\gamma_{1} + \gamma_{2}),$$

while $\Omega \approx 2V$ is the Rabi frequency.

It is clear from Eq. (36) that the rescattering function contains two kinds of terms: one of them is proportional to $\delta(\nu - \nu')$ and describes coherent rescattering of photons, while the second describes incoherent rescattering. In the strong collision regime, i.e., for $\gamma_2 \ge \gamma$, the second type is the deciding one and one can say that there is a complete mixing in frequency. In the purely radiative regime the rescattering relaxation is coherent. The presence of a laser field leads to the appearance of a number of new effects. Firstly, there appear new channels for coherent rescattering: the first one is proportional to $\delta(\nu)$ and describes coherent scattering of the laser field, the second one is proportional to $\delta(\nu - \nu')\delta(\nu)$ and is connected with interference effects. And secondly, even in the purely radiative regime the relaxation of the rescattering function contains an incoherent component. In weak fields, $V \ll \gamma$, this part of the rescattering function has a bell-shaped form. The same features are also found in the rescattering function for the spontaneous sources for the combination tone operators. We show in Figs. 1 and 2 how the incoherent components of the rescattering functions $R(n_{k'})$ and $Q(n_{k'})$ change with increasing intensity in a purely radiative regime and for $\Delta_L = 0$.

We note that in strong fields, $V \ge \gamma$, the rescattering function ceases to be positive (see Fig. 1). This is connected with the features of the interaction of resonance radiation with the atomic subsystem in the presence of a strong laser field: in particular, in some spectral regions we have amplification rather than absorption, which leads effectively to a decrease in the saturation of the atomic subsystem, i.e., the correction to the population of the upper level becomes negative. In that case it is clear, for instance, that $R_2(n_k)$ becomes negative. Parametric effects lead to $R_1(n_k)$ also not being positive.

CONCLUSION

The application of the operator method has turned out to be an extremely convenient procedure in the description of the transfer of resonant radiation in the presence of a laser field. This is connected with the fact that in that description the problem of evaluating the rescattering function, and also other kinetic coefficients, is reduced to a problem in linear algebra, in contrast to the cumbersome procedure of the estimating and calculating of integrals in Keldysh's diagram technique of Ref. 4.

In the present paper we did not take into account the thermal motion of the atoms. However, it has been shown, for instance, in Ref. 4 that taking the thermal motion into account may significantly change not only the absolute magnitude and width of the lines, but also the sign of the effect in various spectral regions, so that this problem requires additional studies.

Notwithstanding the simplified nature of the proposed model the problem may be studied experimentally. The problem considered by us is close to the problem of resonant fluorescence of a two-level atom and the first term in Eq. (33) is described by the Mollow spectrum.¹⁹ This spectrum has been observed in experiments on the scattering of a laser wave by an atomic beam.²⁰ The second term in Eq. (33) and accordingly the rescattering function we have obtained can be measured in an experiment on the scattering of a test signal by an atomic beam in the presence of a laser wave.

Finding how the rescattering function depends on the direction of the photons is connected both with the Doppler effect and with the polarization structure of the radiation. In the present paper we assumed a unique polarization structure of the radiation. The generalization to the case of various polarizations makes the problem considerably more complicated: a significantly larger set of combinations tones



FIG. 1. Spectral behavior of the rescattering function $R(n_{k'})$ for a laser wave in exact resonance, $\Delta_L = 0$, and a radiative regime: $\gamma_1 = \gamma$, $\gamma_2 = \gamma/2$. The parameter V/γ equals: a: 0.01, b: 0.5, c: 1, d: 3.







FIG. 2. Spectral behavior of the rescattering function $Q(n_{k^*})$ for the combination tone operators. The conditions and the parameters are the same as for Fig. 1.



appear which is connected with the possibility of four-wave interactions involving different polarizations.²¹ The procedure of evaluating the rescattering function remains as before in that case. In view of the considerable complications this problem is of practical interest only in the context of describing the interaction between radiation with actual atomic subsystems and taking into account their more complex many-level structure.

APPENDIX

The stationary solution of Eq. (20) for the operator Φ_k^- has the form

$$\begin{split} \Phi_{22k}^{-} = a_{k}a_{k}^{+}P\left(m_{21}\rho_{21}^{(0)} - m_{31}\rho_{22}^{(0)}\right) + a_{k}Pa_{k}^{+}\left(m_{11}\rho_{21}^{(0)} + m_{31}\rho_{11}^{(0)}\right) \\ -a_{k}Pa_{\bar{k}}\left(m_{21}\rho_{21}^{(0)} + m_{41}\rho_{22}^{(0)}\right) - a_{\bar{k}}a_{k}P\left(m_{11}\rho_{12}^{(0)} - m_{41}\rho_{11}^{(0)}\right), \\ (A1) \\ \Phi_{11k}^{-} = -a_{k}a_{k}^{+}P\left(m_{21}\rho_{22}^{(0)} - m_{32}\rho_{22}^{(0)}\right) - a_{k}Pa_{k}^{+}\left(m_{12}\rho_{21}^{(0)} + m_{32}\rho_{11}^{(0)}\right) \\ + a_{k}Pa_{\bar{k}}\left(m_{22}\rho_{12}^{(0)} + m_{42}\rho_{22}^{(0)}\right) + a_{\bar{k}}a_{k}P\left(m_{12}\rho_{12}^{(0)} - m_{42}\rho_{11}^{(0)}\right), \\ (A2) \\ \Phi_{12k}^{-} = a_{k}a_{k}^{+}P\left(m_{23}\rho_{21}^{(0)} - m_{33}\rho_{22}^{(0)}\right) + a_{k}Pa_{k}^{+}\left(m_{13}\rho_{21}^{(0)} + m_{33}\rho_{11}^{(0)}\right) \\ - a_{k}Pa_{\bar{k}}\left(m_{23}\rho_{12}^{(0)} + m_{43}\rho_{22}^{(0)}\right) - a_{\bar{k}}Pa_{k}^{+}\left(m_{13}\rho_{21}^{(0)} - m_{43}\rho_{11}^{(0)}\right), \\ (A3) \\ \Phi_{21k}^{-} = -a_{k}a_{k}^{+}P\left(m_{24}\rho_{21}^{(0)} - m_{34}\rho_{22}^{(0)}\right) - a_{k}Pa_{k}^{+}\left(m_{14}\rho_{21}^{(0)} + m_{34}\rho_{11}^{(0)}\right) \\ + a_{k}Pa_{\bar{k}}\left(m_{24}\rho_{12}^{(0)} + m_{44}\rho_{22}^{(0)}\right) + a_{\bar{k}}a_{k}P\left(m_{14}\rho_{12}^{(0)} - m_{44}\rho_{11}^{(0)}\right), \\ (A4) \end{split}$$

where we have written $m_{jk} = M_{jk}/\det M$, while M_{jk} is the determinant of the corresponding minor of the M(v) matrix. For these quantities we have the following expressions:

$$\begin{split} M_{11} = v (v - \Delta) (v + \Delta^{*}) - 2 |V|^{2} (v - i\gamma_{2}), \\ M_{13} = -V (v - \Delta) (v + i\gamma_{1}), \\ M_{12} = (v - \Delta) (v + \Delta^{*}) i\gamma_{1} + 2 |V|^{2} (v - i\gamma_{2}). \\ M_{14} = -V^{*} (v + \Delta^{*}) (v + i\gamma_{1}), \\ M_{21} = 2 |V|^{2} (v - i\gamma_{2}), \quad M_{22} = -2 |V|^{2} (v - i\gamma_{2}) \\ + (v - \Delta) (v + \Delta^{*}) (v - i\gamma_{1}), \\ M_{23} = -V (v - \Delta) (v - i\gamma_{1}), \quad M_{24} = -V^{*} (v + \Delta^{*}) (v - i\gamma_{1}), \\ M_{31} = -V^{*} v (v - \Delta), \end{split}$$

$$M_{33} = v [(v - \Delta) (v - i\gamma_1) - 2|V|^2], \quad M_{34} = 2 (V^*)^2 v.$$

$$M_{41} = -Vv(v + \Delta^{*}),$$

$$M_{43} = 2V^{2}v, \quad M_{44} = v[(v + \Delta^{*})(v - i\gamma_{1}) - 2|V|^{2}],$$

$$M_{32} = M_{31}, \quad M_{42} = M_{41};$$

$$\det M = vf(v), f(v) = (v - \Delta)(v + \Delta^{*})(v - i\gamma_{1}) - 4|V|^{2}(v - i\gamma_{2}).$$

For the components $\rho_{jk}^{(0)}$ of the atomic density matrix which is the stationary solution of Eq. (6) we have the well known expressions (see, e.g., Ref. 11):

$$\rho_{22}^{(0)} = \frac{2|V|^2 \gamma_2}{\gamma_1 |\Delta|^2 + 4|V|^2 \gamma_2}, \quad \rho_{12}^{(0)} = \frac{V \Delta \gamma_1}{\gamma_1 |\Delta|^2 + 4|V|^2 \gamma_2},$$
$$\rho_{11}^{(0)} = 1 - \rho_{22}^{(0)}, \quad \rho_{21}^{(0)} = (\rho_{12}^{(0)})^{\circ}.$$

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