STIMULATED RADIATION FROM ATOMS DURING INTERACTION OF CASCADE TRANSITIONS

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The response of a quantum system under the action of several coherent fields which are in resonance with various coupled transitions are investigated by aid of the density matrix and Maxwell's equations. Some features of the system as a source of laser radiation are also studied. It is shown that under some conditions which can be realized in the optical range, nonlinear interaction of allowed transitions via forbidden ones can occur. This interaction results in a change of the dispersion properties of the allowed transitions. Under generation conditions the transition interaction results in a change of the thresholds and generation frequency for each transition, the change depending on the frequency and intensity of the auxillary-transition radiation.

SEVERAL authors [1-3] have shown that when atoms or molecules interact with several microwave fields that are resonant with different coupled transitions, a change in their susceptibilities is observed. The change in the susceptibility can be either due to a redistribution of the particles among the energy levels, or to the existence of a non-linear interaction between the interatomic motions induced by the different fields. The efficiency of interaction is greatly influenced by the relaxation properties of the medium. By now, lasers generating at different transitions of the same substance have already been developed, including those generating at several transitions jointly [4-7]. It is therefore of interest to ascertain what effects can be expected in the interaction of optical transitions and how different kinetic processes become manifest in this case.

In this paper we investigate the optical properties of atoms that are under the influence of several coherent fields, and the singularities of such a system when viewed as a source of coherent emission. The optical transitions are characterized by relaxation properties and selection rules other than those of the microwave ones considered in [1-3]. It turns out as a result that in the optical band a nonlinear coupling can be effectively produced, via the forbidden transitions, between the polarization components at the frequencies of the allowed transitions. This is reflected in the optical properties of the system.

1. RESPONSE OF A QUANTUM SYSTEM TO THE ACTION OF SEVERAL COHERENT FIELDS

Let us consider a quantum system described by a time-independent Hamiltonian H_0 with the eigenvalue scheme shown in Fig. 1. The optical electric-dipole transitions a, b, c, and f are allowed while x and y are forbidden. The relaxation properties of the system are described by the level widths γ_n, by the relaxation-transition probabilities per unit time γ_{mm} (m > n), and by the reciprocal phase-memory times $\gamma_1^2 = \gamma_2^1 = \gamma_a$, $\gamma_1^3 = \gamma_3^1 = \gamma_x$, etc. Each of the levels is filled, from the levels which do not enter in the system under consideration, the excitation probability per unit time being Q_n.

The forced motions in the system under the influence of the field

$$E(r,t) = \mathrm{Re}\,\{E_aU_a(r)e^{i\Omega_at} + E_bU_b(r)e^{i\Omega_bt} + E_cU_c(r)e^{i\Omega_ct} + E_fU_f(r)e^{i\Omega_ft}\}$$

will be described by the aid of a density matrix ρ, which satisfies the equation

$$\frac{\partial}{\partial t}\rho_{mn} + i[H_0 + W(r,t), \rho]_{mn} - R_{mn}(\rho) = 0\ (\hbar = 1), \quad (1.1)$$

where W is the Hamiltonian of the perturbation, and the matrix elements of the operator of relaxation and excitation are

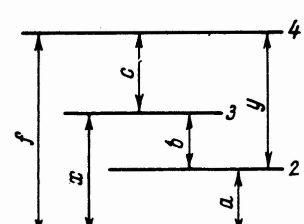

FIG. 1

$$R_{mn}(\rho) = \begin{cases} \sum_k (\rho_{kk}\gamma_{kn} - \rho_{mm}\gamma_{mk}), & m = n \\ -\gamma_n{}^m \rho_{mn}, & m \neq n \end{cases};$$

$$\sum_{k\neq 4,3,2} \rho_{kk}\gamma_{kn} = Q_n, \qquad \sum_k \gamma_{mk} = \gamma_m. \tag{1.2}$$

For convenience we represent the matrix of the perturbation Hamiltonian in the form

$$\{W_{mn}(r, t)\} = \begin{pmatrix} 0 & \gamma_a V_a e^{i\Omega_a t} & 0 & \gamma_f V_f e^{i\Omega_f t} \\ & 0 & \gamma_b V_b e^{i\Omega_b t} & 0 \\ & \text{к.с.} & 0 & \gamma_c V_c e^{i\Omega_c t} \\ & & & 0 \end{pmatrix}, \tag{1.3}$$

where the frequencies Ω_a, Ω_b, Ω_c, and Ω_f are respectively close to the eigenfrequencies ω_a, ω_b, ω_c, and ω_f of the transitions a, b, c, and f.

The optical properties of the medium will be investigated with the aid of the complex polarization that arises at the frequency of each of the transitions. Accurate to the third approximation in the stationary mode, we can seek the matrix of the solution of Eq. (1.1), confining ourselves to resonant terms, in the form

$$\{\rho_{mn}(r, t)\} = \begin{pmatrix} n_1 & \rho_{12} & \rho_{13} & \rho_{14} \\ & n_2 & \rho_{23} & \rho_{24} \\ & \text{к.с.} & n_3 & \rho_{34} \\ & & & n_4 \end{pmatrix}, \quad \rho_{ij} = \overset{*}{\rho}_{ji} \tag{1.4}$$

where

$$\rho_{12} = ae^{i\Omega_a t} + \tilde{a}e^{i\tilde{\Omega}_a t}, \qquad \rho_{23} = be^{i\Omega_b t} + \tilde{b}e^{i\tilde{\Omega}_b t},$$
$$\rho_{34} = ce^{i\Omega_c t} + \tilde{c}e^{i\tilde{\Omega}_c t}, \qquad \rho_{14} = fe^{i\Omega_f t} + \tilde{f}e^{i\tilde{\Omega}_f t},$$
$$\rho_{13} = xe^{i\Omega_x t} + \varkappa e^{i\Omega_\varkappa t}, \qquad \rho_{24} = ye^{i\Omega_y t} + \eta e^{i\Omega_\eta t}; \tag{1.5}$$

$$\tilde{\Omega}_a = \Omega_f - \Omega_b - \Omega_c,\ \tilde{\Omega}_b = \Omega_f - \Omega_a - \Omega_c,\ \tilde{\Omega}_c = \Omega_f - \Omega_a - \Omega_b,$$
$$\tilde{\Omega}_f = \Omega_a + \Omega_b + \Omega_c, \quad \Omega_x = \Omega_a + \Omega_b,$$
$$\Omega_\varkappa = \Omega_f - \Omega_c, \quad \Omega_y = \Omega_b + \Omega_c, \quad \Omega_\eta = \Omega_f - \Omega_a.$$

The second-approximation solutions for $\Delta n_a^{(2)} = n_2^{(2)} - n_1^{(2)}$, $\Delta n_b^{(2)} = n_3^{(2)} - n_2^{(2)}$, etc., represented in matrix form, are

$$\begin{pmatrix} \Delta n_a^{(2)} \\ \Delta n_b^{(2)} \\ \Delta n_c^{(2)} \\ \Delta n_f^{(2)} \end{pmatrix} = \begin{pmatrix} \Delta n_a \\ \Delta n_b \\ \Delta n_c \\ \Delta n_f \end{pmatrix} + \begin{pmatrix} \alpha_a & \alpha_b & \alpha_c & \alpha_f \\ \beta_a & \beta_b & \beta_c & \beta_f \\ s_a & s_b & s_c & s_f \\ \varphi_a & \varphi_b & \varphi_c & \varphi_f \end{pmatrix} \begin{pmatrix} \delta n_a |V_a|^2 \\ \delta n_b |V_b|^2 \\ \delta n_c |V_c|^2 \\ \delta n_f |V_f|^2 \end{pmatrix}, \tag{1.6}$$

where $\Delta n_c = n_4^{(0)} - n_3^{(0)}$, $\Delta n_f = n_1^{(0)}$, etc. are the zeroth-approximation solutions, which describe the fraction contributed to the formation of the difference of the populations in each of the transitions by the sources of excitation of the different levels. For example, when $Q_1 = Q_3 = 0$ we have

$$\Delta n_a = \frac{\gamma_{43}\gamma_{32}(\gamma_1 - \gamma_{21})}{\gamma_3\gamma_2\gamma_1}\frac{Q_4}{\gamma_4} + \frac{\gamma_1 - \gamma_{21}}{\gamma_1}\frac{Q_2}{\gamma_2},$$

$$\Delta n_b = \frac{\gamma_{43}(\gamma_2 - \gamma_{32})}{\gamma_3\gamma_2}\frac{Q_4}{\gamma_4} - \frac{Q_2}{\gamma_2}, \qquad \Delta n_c = \frac{\gamma_3 - \gamma_{43}}{\gamma_3}\frac{Q_4}{\gamma_4},$$

$$\Delta n_f = \frac{\gamma_3\gamma_2(\gamma_1 - \gamma_{41}) - \gamma_{43}\gamma_{32}\gamma_{21}}{\gamma_3\gamma_2\gamma_1}\frac{Q_4}{\gamma_4} - \frac{\gamma_{21}}{\gamma_1}\frac{Q_2}{\gamma_2}; \tag{1.7}$$

The values of δn_μ are

$$\delta n_\mu = \frac{\Delta n_\mu}{|D_\mu|^2}, \qquad D_\mu = \frac{\omega_\mu - \Omega_\mu}{\gamma_\mu} + i = \delta\Omega_\mu + i,$$

and the coefficients α, β, s, and φ describe the contribution of each of the fields to the redistribution of the particles in the allowed transitions, and are completely expressed in terms of the relaxation constants. Thus,

$$\beta_a = 2\frac{\gamma_a}{\gamma_2}, \qquad \beta_b = -2\gamma_b\frac{\gamma_2 + \gamma_3 - \gamma_{32}}{\gamma_3\gamma_2}, \tag{1.8}$$

$$\beta_c = 2\gamma_c\frac{(\gamma_4 - \gamma_{43})(\gamma_2 - \gamma_{32})}{\gamma_4\gamma_3\gamma_2}, \qquad \beta_f = -2\gamma_f\frac{\gamma_{43}(\gamma_2 - \gamma_{32})}{\gamma_4\gamma_3\gamma_2}.$$

Depending on the relation between the relaxation constants, the role of each of the fields in the redistribution of the particles among the levels under consideration can be different. Whereas the field that is resonant with the transition under consideration always decreases the inverted population (α_a, β_b, s_c, $\varphi_f < 0$), interaction with other fields may contribute to the occurrence of inversion. For the transition b, for example, we note that the action of the field E_c is proportional to the factor $1 - \gamma_{43}/\gamma_4$, whereas the analogous action of the excitation source Q_4 is proportional to γ_{43}/γ_4. The action of both factors is proportional to $\gamma_2 - \gamma_{32}$ and contributes to the creation of inversion if $\gamma_2 > \gamma_{32}$. Under these conditions, the action of the field E_f is favorable only when $\Delta n_f < 0$, and in the opposite case it worsens the conditions for the formation of the inversion $\Delta n_b^{(2)}$.

It also follows from the second-approximation solutions that the interaction between the oscillations at the frequencies of the allowed transitions produces in the system oscillations with combination frequencies close to the natural frequencies of the forbidden transitions. The amplitudes x, κ, y, and η of these oscillations are proportional to the product of the field amplitudes at the combining frequencies, and have three resonances. These are the resonances of the combining transitions and the "combination" resonance corresponding to the requirement that the frequency of the combination oscillation be close to the natural frequency of the forbidden transition.

The combination oscillations x, κ, y, and η, which are not accompanied by emission of photons of suitable frequency, ensure the "reactive" coupling between the oscillations of the allowed transitions via the forbidden ones, and introduce additional resonances in the third-approximation solutions:

$$b^{(3)} = \frac{1}{D_b}\left(\Delta n_b^{(2)} V_b + \frac{\gamma_a}{\gamma_b} x V_a^* - \frac{\gamma_c}{\gamma_b} y V_c^*\right),$$

$$\tilde{b}^{(3)} = \frac{1}{D_b}\left(\frac{\gamma_a}{\gamma_b} \varkappa V_a^* - \frac{\gamma_c}{\gamma_b} \eta V_c^*\right). \quad (1.9)$$

The remaining solutions have an analogous construction.

Thus, the solution for each transition consists of two components. The first is proportional to the intensity of the resonant field and describes oscillations having the same frequency and phase as this field. Besides the ordinary oscillations, it receives contributions also from the combination oscillations. The second component is due exclusively to the combination processes and does not depend at all on the intensity of the field which is resonant to the given transition. It describes oscillations whose frequency and phase are in general not equal to the frequency and phase of the resonant field[1)].

2. DISPERSION PROPERTIES OF THE RESPONSE

By way of an example let us consider the complex amplitude P_b of the polarization in the transition b, belonging to the first spatial harmonic. For simplicity we consider the case $\Omega_a + \Omega_b + \Omega_c = \Omega_f$ and $k_a + k_b + k_c = k_f$, when both components of the solution (1.9) for the transition b describe oscillations at the same frequency Ω_b. We assume that the fields are essentially inhomogeneous only along one coordinate axis:

$$V_a = A \sin k_a z, \quad V_b = B \sin k_b z,$$
$$V_c = C \sin k_c z, \quad V_f = F \sin k_f z,$$

where $A = -E_a d_a / 2\gamma_a$, $C = -E_c d_c / 2\gamma_c$ etc., $d_a = d_{12}$ is the matrix element of the dipole moment of the transition, and k is the projection of the wave vector on the z axis.

With the aid of the third-approximation formulas we can obtain

$$\frac{1}{N d_b^*} P_b = \frac{B}{D_b}\Big[\Delta n_b + \frac{1}{2}(\beta_a \delta n_a + \Lambda) \times |A|^2 + \frac{1}{2}(\beta_c \delta n_c + \Sigma)|C|^2 + \frac{1}{2}\beta_f \delta n_f |F|^2 + \frac{3}{4}\beta_b \delta n_b |B|^2\Big] + \frac{1}{4}\frac{\Theta}{D_b} A^* C^* F; \quad (2.1)$$

$$\Lambda = \frac{1}{D_x^a}\left(\frac{\gamma_a}{\gamma_b}\frac{\Delta n_b}{D_b} - \frac{\Delta n_a}{D_a}\right), \quad \Sigma = \frac{1}{D_y^c}\left(\frac{\gamma_c}{\gamma_b}\frac{\Delta n_b}{D_b} - \frac{\Delta n_c}{D_c}\right),$$

$$\Theta = \frac{1}{D_x^a}\left(\frac{\gamma_f}{\gamma_b}\frac{\Delta n_c}{D_c^*} - \frac{\gamma_c}{\gamma_b}\frac{\Delta n_f}{D_f}\right) + \frac{1}{D_y^c}\left(\frac{\gamma_f}{\gamma_b}\frac{\Delta n_a}{D_a^*} - \frac{\gamma_a}{\gamma_b}\frac{\Delta n_f}{D_f}\right),$$

$$D_x^a = \frac{\omega_x - \Omega_x + i\gamma_x}{\gamma_a} = \delta\Omega_a + \frac{\gamma_b}{\gamma_a}\delta\Omega_b + i\frac{\gamma_x}{\gamma_a},$$

$$D_y^c = \delta\Omega_c + \frac{\gamma_b}{\gamma_c}\delta\Omega_b + i\frac{\gamma_y}{\gamma_c} \quad (2.2)$$

(N—density of the atoms of the working component of the medium). The numerical coefficients in formula (2.1) are due to the operation of separating the first spatial Fourier component of the polarization, and vary with the character of the spatial inhomogeneity of the fields. The coefficients Λ, Σ, and Θ describe the contribution of the combination processes to the formation of the polarization in the transition b. The corresponding formulas for the other transitions have a form which is symmetrical to (2.1).

It follows from (2.2) that $\Lambda = \Lambda(\delta\Omega_b)$, $\Sigma = \Sigma(\delta\Omega_b)$ and $\Theta = \Theta(\delta\Omega_b)$, meaning that the contributions made to different sections of the spectral contour of the transition are different. The conditions for the manifestation of the effect are as follows:

$$|\Lambda| \gtrsim \max\{|\beta_a \delta n_a|, |\Delta n_b|\},$$

$$|\Sigma| \gtrsim \max\{|\beta_c \delta n_c|, |\Delta n_b|\}, \quad |\Theta| \gtrsim |\Delta n_b|. \quad (2.3)$$

When conditions (2.3) are satisfied in fields E_a, E_c and E_f that noticeably perturb their own transitions, that is, when $|A|^2, |C|^2, |F|^2 \sim 1$, one can observe a distortion of the dispersion properties of the transition b; this distortion increases with increasing intensity of these fields. The required fields $|Ed/\gamma| \sim 1$ are attained in laser cavities when the threshold is exceeded by several times (see, for example, formula (12) of [9]). At small relative detunings, the conditions for the appearance of combination processes take the form

$$\frac{\gamma_a}{\gamma_x}\left|\frac{\gamma_a}{\gamma_b} - \frac{\Delta n_a}{\Delta n_b}\right| \gtrsim \max\left\{1, 2\frac{\gamma_a}{\gamma_2}\left|\frac{\Delta n_a}{\Delta n_b}\right|\right\}, \quad (2.4)$$

$$\frac{\gamma_c}{\gamma_y}\left|\frac{\gamma_c}{\gamma_b} - \frac{\Delta n_c}{\Delta n_b}\right| \gtrsim \max\left\{1, \left|\beta_c \frac{\Delta n_c}{\Delta n_b}\right|\right\}, \quad (2.5)$$

$$\frac{\gamma_f}{\gamma_b}\left(\frac{\gamma_a}{\gamma_x}\frac{\Delta n_c}{\Delta n_b} + \frac{\gamma_c}{\gamma_y}\frac{\Delta n_a}{\Delta n_b}\right) - \left(\frac{\gamma_a}{\gamma_x}\frac{\gamma_c}{\gamma_b} + \frac{\gamma_c}{\gamma_y}\frac{\gamma_a}{\gamma_b}\right)\left|\frac{\Delta n_f}{\Delta n_b}\right| \gtrsim 1. \quad (2.6)$$

It follows from (2.4)—(2.6) that in the optical band the effectiveness of the combination oscillations is determined not only by the ratio of the relaxation constants, but also by the ratio of the differences of the populations in the interacting transitions, which can be appreciable in the optical band. With respect to relaxation, it is necessary above all that the relaxation constants of the combination processes be smaller than or close to the widths of the spontaneous-emission lines at

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1)The possibility of a similar effect in a two-level system was noted by Kuznetsova and Rantian[8].

the corresponding transitions ($\gamma_x \lesssim \gamma_a$, $\gamma_y \lesssim \gamma_c$).

Further, if the population difference in the transition under consideration is much smaller than in the transition combining with it, then a relatively large change takes place in the susceptibility, owing to the incoherent interaction of the transitions, which changes only the difference in the populations. In order for the combinations of the first type ($\frac{1}{2}\Lambda B|A|^2$, $\frac{1}{2}\Sigma B|C|^2$) to be noticeable, it is necessary that the line width of the spontaneous emission in the additional transition be close to the width of the level which is common with the transition under consideration ($2\gamma_a \sim \gamma_2$, $2\gamma_c \sim \gamma_3$). To observe the combination effect of the second type ($\frac{1}{4}\Theta A^*C^*F$) at this ratio of the population difference, it is necessary that the line widths for the additional transitions be larger than or close to the line widths of the spontaneous emission for the transition under consideration.

If the ratio of the population differences is inverted, the principal factor that must be compared with the magnitude of the combination contributions is the proper population difference in the transition under consideration. In this case it is necessary to satisfy the condition $\gamma_a, \gamma_c, \gamma_f \gtrsim \gamma_b$, and this requirement must be more stringent for the combination process of the second type.

Obviously, the conditions for the observation of the effects connected with the combination processes are easiest to realize in gases. In this case the most favorable situation is one in which the broadest of all the levels of two transitions under consideration is the common level. In gases, however, additional effects connected with the motion of the particles can arise if the Doppler width of the spectral line is much larger than the dispersion width ($ku/\gamma \gg 1$). An investigation of these effects is beyond the scope of the present work, but the results can be used for estimates for long-wave transitions of heavy inert gases ($ku/\gamma \sim 1$)[2)].

Let us consider a model with characteristics that are typical for this case: $\gamma_4 = \gamma_1 = 8 \times 10^{-7}$ sec^{-1}, $\gamma_{41} = \gamma_{43} = 0.1 \times 10^7$ sec^{-1}, $\gamma_3 = 10^8$ sec^{-1}, $\gamma_{32} = 0.8 \times 10^7$ sec^{-1}, $\gamma_2 = 10^7$ sec^{-1}, $\gamma_{21} = 0.5 \times 10^7$ sec^{-1}, $\gamma_a = \gamma_y = 4 \times 10^7$ sec^{-1}, $\gamma_b = 5 \times 10^7$ sec^{-1}, $\gamma_c = \gamma_x = 9 \times 10^7$ sec^{-1}, $\gamma_f = 8 \times 10^7$ sec^{-1}, $Q_4/\gamma_4 \approx 10^{-10}$, $Q_2/\gamma_2 \approx 10^{-11}$, and $Q_3 = Q_1 = 0$. With the aid of formulas (1.7) and (1.8) we get

$$\Delta n_a = 0{,}9\frac{Q_2}{\gamma_2} - 5\cdot 10^{-3}\frac{Q_4}{\gamma_4} = 8{,}5\cdot 10^{-12},$$

$$\Delta n_b \approx -\frac{Q_2}{\gamma_2} = -10^{-11}, \quad \Delta n_f = \Delta n_c \approx \frac{Q_4}{\gamma_4} = 10^{-10},$$

$$\beta_a \approx 8, \quad \beta_b \approx -10, \quad \beta_c \approx 0{,}4, \quad \beta_f \approx -2,$$

$$|\Lambda| \ll \beta_a \Delta n_a, \quad |\Sigma| \sim |\beta_c \Delta n_c|,$$

$$|\Theta| \sim |\beta_c \Delta n_c|, |\beta_a \Delta n_a|, |\beta_f \Delta n_f| \quad (|\delta\Omega_{a,b,c,f}| \ll 1).$$

Thus, under the considered conditions, the excitation of level 4 exerts a noticeable influence on the formation of inversion in transition a, and has practically no effect on the population difference in transition b. Excitation of level 2 exerts no noticeable influence on the population difference of the transition f. The effect of the field E_f turns out to be the largest in the change of the population difference of the transition b, producing an increase in the latter. Fields E_c and E_a of equal intensity increase $\Delta n_b^{(2)}$ in approximately equal fashion. However, the action of the field E_c is manifest not only in an increase in the difference of the populations of the transition b, but also in the change of its dispersion properties, whereas the action of the field E_a under the same conditions only changes the magnitude of the population difference.

Figure 2 shows the imaginary (solid) and real (dashed) parts of the factor $(\beta_c \delta n_c + \Sigma)/\Delta n_b D_b$, describing the ratio of the weight of the contribution to the polarization by the fields E_c and E_b to the intrinsic characteristics of the transition at the center of the line. Curves 1 correspond to $\delta\Omega_c = -0.4$. On curves 2 we show for comparison the case $\delta\Omega_c = 0$. It is seen from curves 1 that the contour of the contribution due to the field E_c is antisymmetrical, and has a positive maximum and negative minima. Since the contribution of the field E_c can even change sign in different parts of the contour of the transition. The maxima and

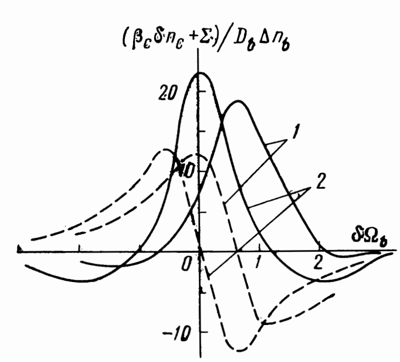

FIG. 2. Dispersion properties of the contribution of the field E_c to the polarization in the transition b. The real part is shown by the dashed line, the imaginary one by the solid line; curves: 1–$\delta\Omega_c$ = –0.4, 2–$\delta\Omega_c$ = 0.

[2)]Thus, for example, for the $3s_2 - 3p_4$ and $3p_4 - 2s_2$ transitions, at which generation was attained, the respective values of ku/γ are 3 and 7.

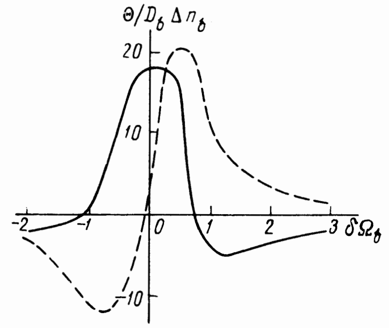

FIG. 3. Dispersion properties of the contribution of the field E_a, E_c, and E_f to the polarization in the transition b. The real part is represented by the dashed line, the imaginary one by the solid line; $\delta\Omega_a = 1$, $\delta\Omega_c = -0.4$, $\delta\Omega_a + \delta\Omega_b + \delta\Omega_c = \delta\Omega_f$.

minima are the results of a superposition of the resonances referred to above. In this case the position of the maximum Im $\Sigma/\Delta n_b D_b$ is close to the point $\delta\Omega_b = -(\gamma_c/\gamma_b)\,\delta\Omega_c$, tending to $\delta\Omega_b = 0$ as $\delta\Omega_c \to 0$. As follows from curves 2, the intensity of the maximum and of the minima increases in this case.

The dispersion properties of the factor $\Theta/\Delta n_b D_b$ as a function of $\delta\Omega_b$ are also described by a complicated contour. The solid and dashed lines of Fig. 3 are respectively the imaginary and real parts of this factor for the case $\delta\Omega_b = 1$, $\delta\Omega_c = -0.4$, $\delta\Omega_a + \delta\Omega_b + \delta\Omega_c = \delta\Omega_f$. Thus, the dispersion properties of the transition b in the model under consideration can experience noticeable changes when the field intensity is increased. Therefore the effects noted can lead to a distortion of the properties of the interacting optical signals and exert an influence on the operation of lasers.

3. FEATURES OF GENERATION IN THE CASE WHEN COMBINATION PROCESS DO NOT APPEAR

Let us consider the self-consistent problem of the emission from atoms in a laser cavity. In view of the fact that we are investigating only the case of generation near threshold, we assume that the radiation is single-mode in each of the transitions. To investigate the interaction between the radiation and the "active" medium in the optical resonator, we obtain from Maxwell's equation in the stationary state the equation

$$\frac{\Omega_\alpha^2 - k_\alpha^2}{\Omega_\alpha^2} - i\frac{\Delta k_\alpha}{k_\alpha} = -8\pi P_\alpha \frac{1}{E_\alpha} \quad (c = 1), \qquad (3.1)$$

where Ω_α is the generation frequency, k_α the wave vector resolved by the interferometer, Δk_α the width of the transmission band of the interferometer relative to radiation of frequency $\Omega_\alpha \approx k_\alpha$ ($|\Omega_\alpha - k_\alpha| \lesssim \Delta k_\alpha$), E_α the amplitude of the field intensity of the radiation at this frequency, and P_α the amplitude of the first spatial harmonic of the complex polarization at the frequency Ω_α. Thus, knowing P_α and equating real and imaginary parts of (3.1), we obtain the conditions imposed on the frequency and generation power by the rates of excitation and by the relaxation characteristics of the quantum system and the Q of the interferometer.

We confine ourselves to an investigation of the case of interaction of electric-dipole cascade transitions $4 \to 3 \to 2 \to 1$ ($F = 0$). We consider generation in the transition b when the conditions (2.3)—(2.5) are not satisfied, that is, the field interaction merely redistributes the particles over the levels. The expression in the curly brackets in formula (2.1) will in this case be pure real and will describe the saturated population difference.

A. If the fields E_a and E_c are specified and the field E_b is generated, then, substituting (2.1) in (3.1), we obtain formulas for the frequency and amplitude of the generated field:

$$(\Omega_b^2 - k_b^2)/\Omega_b^2 = \delta\Omega_b \Delta k_b/k_b, \qquad (3.2)$$

$$|B|^2 = K_b'(1 - \Pi_b'/\Delta n_b), \qquad (3.3)$$

where

$$\Pi_b' = \Pi_b - {}^1/_2\,\beta_a \delta n_a |A|^2 - {}^1/_2\,\beta_c \delta n_c |C|^2,\ \Pi_b = |D_b|^2 \Delta\tilde{k}_b,$$
$$\Delta\tilde{k}_b = \Delta k_b \gamma_b / 4\pi N \Omega_b |d_b|^2,\ K_b' = K_b = -{}^4/_3 |D_b|^2 / \beta_b.$$

Putting in (3.2) $\Omega_b + k_b \approx 2\Omega_b$ and $(\omega_b - k_b)/\gamma_b = \delta k_b$ we get

$$(1 + \Delta k_b/2\gamma_b)\,\delta\Omega_b = \delta k_b. \qquad (3.4)$$

We see therefore that in our case the generation frequencies do not depend on the power and are closer to the natural frequency of the system having the higher Q (atomic or resonator). When $k_b = \omega_b$ we get $\Omega_b = \omega_b$. It is seen from (3.3) that Π_b' is the threshold value of Δn_b. We shall henceforth take the threshold value of Δn to mean always the zeroth-approximation Δn corresponding to the threshold excitation rates.

In the absence of the fields E_a and E_c, the proper generation threshold is $\Pi_b = |D_b|^2 \Delta\tilde{k}_b$. The presence of additional fields in the system can raise or lower the threshold, depending on the ratio of the relaxation constants and of the population differences in the additional transitions. From the formula for Π_b' and from (1.8) it follows that when $\Delta n_a > 0$, $\Delta n_c > 0$ and $\gamma_2 > \gamma_{23}$ the fields E_a and E_c lower the threshold of generation in the transition b. Comparing the experimental values of K_b' and Π_b' with the calculated values, we can determine the unknown relaxation constants from the known characteristics. If the fields that are resonant to the transitions a and c are homogeneous, then the coefficients $1/2$ in the formula for Π_b' must be replaced by unity. The coefficient $4/3$ in the formula for K_b' is also connected with the inhomogeneity of the generated field along the resonator.

B. Assume that the resonator has several natural frequencies that are close, for example, to the natural frequencies of the atomic transitions a, b, and c. Then, with increasing excitation power, the first to begin to generate is the transition in which the condition $\Delta n = |D|^2\Delta\tilde{k}$ is satisfied most rapidly. The radiation produced by one of the transitions facilitates the excitation of generation by the other transitions. When all three fields generate simultaneously, their frequencies are described as before by formulas such as (3.4), and the amplitudes must satisfy the following system of equations:

$$\begin{aligned}
&{}^3/_4\, \alpha_a\delta n_a|A|^2 + {}^1/_2\, \alpha_b\delta n_b|B|^2 + {}^1/_2\, \alpha_c\delta n_c|C|^2 \\
&\quad = |D_a|^2\,\Delta\tilde{k}_a - \Delta n_a, \\
&{}^1/_2\, \beta_a\delta n_a|A|^2 + {}^3/_4\, \beta_b\delta n_b|B|^2 + {}^1/_2\, \beta_c\delta n_c|C|^2 \\
&\quad = |D_b|^2\,\Delta\tilde{k}_b - \Delta n_b, \\
&\quad {}^1/_2 s_b\delta n_b|B|^2 + {}^3/_4 s_c\delta n_c|C|^2 = |D_c|^2\Delta\tilde{k}_c - \Delta n_c.
\end{aligned} \tag{3.5}$$

For each of the transitions, the solution can be represented in the form (3.3). Thus, under conditions when the fields E_a and E_c generate, the formulas for the threshold population difference Π and for the coefficients $K^{a,c}$ for the transition b are

$$\begin{aligned}
\Pi_b{}^{a,c} &= \Pi_b + \frac{2}{3}\left(\frac{\beta_c}{s_c} - \frac{2}{3}\frac{\alpha_c\beta_a}{s_c\alpha_a}\right)(\Delta n_c - |D_c|^2\Delta\tilde{k}_c) \\
&\quad + \frac{2}{3}\frac{\beta_a}{\alpha_a}(\Delta n_a - |D_a|^2\Delta\tilde{k}_a),
\end{aligned} \tag{3.6}$$

$$\begin{aligned}
K_b{}^{a,c} &= \frac{9}{2}\cdot\alpha_a s_c|D_b|^2\left[\frac{3}{2}\alpha_a\left(s_b\beta_c - \frac{9}{4}s_c\beta_b\right)\right. \\
&\quad \left. + \beta_a\left(\frac{3}{2}s_c\alpha_b - s_b\alpha_c\right)\right]^{-1}.
\end{aligned} \tag{3.7}$$

If $\gamma_1 > \gamma_{21}$ and $\gamma_2 > \gamma_{32}$, then it turns out that the coefficients s_b, β_c, α_b, β_a, and α_c are positive. Analysis of (3.7) shows that under these conditions $K^{a,c} > K_b$. Thus, if the following conditions are satisfied for the transitions c and a

$$\Delta n_c - |D_c|^2\,\Delta\tilde{k}_c > 0, \qquad \Delta n_a - |D_a|^2\,\Delta\tilde{k}_a > 0,$$

then generation by the transition b can be excited even if the proper threshold is not exceeded, that is, when $\Delta n_b - |D_b|^2\Delta\tilde{k}_b < 0$).

For the other elements of the cascade, the formulas for the threshold values of Δn, under conditions of generation by the remaining transitions, are

$$\begin{aligned}
\Pi_c{}^{b,a} &= |D_c|^2\Delta\tilde{k}_c + \left(\frac{3}{2}\frac{\beta_b}{s_b} - \frac{2}{3}\frac{\beta_a\alpha_b}{\alpha_a s_b}\right)^{-1}(\Delta n_b - |D_b|^2\Delta\tilde{k}_b) \\
&\quad + \left(\frac{\alpha_b}{s_b} - \frac{9}{4}\frac{\beta_b\alpha_a}{s_b\beta_a}\right)^{-1}(\Delta n_a - |D_a|^2\,\Delta\tilde{k}_a),
\end{aligned} \tag{3.8}$$

$$\begin{aligned}
\Pi_a{}^{b,c} &= |D_a|^2\,\Delta\tilde{k}_a + \frac{3\alpha_b s_c - 2s_b\alpha_c}{{}^9/_2\beta_b s_c - 2\beta_c s_b}(\Delta n_b - |D_b|^2\,\Delta\tilde{k}_b) \\
&\quad + \frac{3\beta_b\alpha_c - 2\alpha_b\beta_c}{{}^9/_2\beta_b s_c - 2\beta_c s_b}(\Delta n_c - |D_c|^2\,\Delta\tilde{k}_c).
\end{aligned} \tag{3.9}$$

If we consider the conditions of excitation in the presence of generation by only one of the elements of the cascade, then it is necessary to set equal to zero in (3.6), (3.8), and (3.9) those coefficients which make independent the corresponding pair of equations in the system (3.5). It follows from (3.6), (3.8), and (3.9) that if $\Pi_\epsilon^\delta < \Pi_\epsilon$ and $\Pi_\epsilon > \Delta n_\epsilon > \Pi_\epsilon^\delta$, then $\Pi_\delta^\epsilon > \Pi_\delta$ and has the meaning of the threshold population difference in the transition δ, for which joint generation is still realized. It can be shown, however, that even if $\Pi_\delta^\epsilon > \Pi_\delta$ at fixed $\Delta n_\delta > \Pi_\delta^\epsilon$, the generation power in the transition δ in the presence of generation by the transition ϵ is larger than in the absence of generation by the transition ϵ. The latter is connected with the fact that $K_\delta^\epsilon > K_\delta$. This is also seen directly from (3.5). At all transitions, the generation is excited simultaneously only under the condition that the relation $\Delta n = |D|^2\Delta\tilde{k}$ is satisfied in each of the transitions simultaneously. In the opposite case, for fixed Δn_α, it is possible to excite or stop the generation by some transitions by selectively varying the resonator Q for the other transitions.

Let us consider the limiting case, when $\gamma_{mn} \ll \gamma_n, \gamma_m$ and the excitation is realized only at the level 4. Under these conditions $\Delta n_b \equiv \Delta n_a \equiv 0$ and the lower levels are filled essentially as a result of induced transitions. If the resonator bandwidth is sufficiently large, then we get from (3.4) that $|D_a| = |D_b| = |D_c| = 1$, and from formulas (3.8), (3.5), and (3.9) it follows that the threshold rates of excitation of the level 4 are related like

$$\begin{aligned}
&Q_4 : Q_4{}^c : Q_4{}^{c,b} \\
&\quad = \Delta\tilde{k}_c : \left(\Delta\tilde{k}_c + \frac{3}{2}\frac{\gamma_3+\gamma_4}{\gamma_4}\Delta\tilde{k}_b\right) : \left[\Delta\tilde{k}_c + \frac{3}{2}\frac{\gamma_3+\gamma_4}{\gamma_4}\Delta\tilde{k}_b\right. \\
&\quad \left. + \left(\frac{9}{4}\frac{\gamma_3+\gamma_4}{\gamma_4}\frac{\gamma_3+\gamma_2}{\gamma_3} - \frac{\gamma_2}{\gamma_3}\right)\Delta\tilde{k}_a\right].
\end{aligned}$$

Thus, it is easiest to excite the cascade if $\gamma_4 \gg \gamma_3 \gg \gamma_2$, and then

$$Q_4:Q_4^c:Q_4^{c,b} = \Delta\tilde{k}_c:(\Delta\tilde{k}_c + {}^3/_2\,\Delta\tilde{k}_b) :(\Delta\tilde{k}_c + {}^3/_2\,\Delta\tilde{k}_b + {}^9/_4\Delta\tilde{k}_a).$$

The numerical coefficients are connected as before with the inhomogeneity of the generated fields. As follows from the formula, this factor raises the excitation threshold.

4. INTERACTION OF TRANSITIONS WITH PARTICIPATION OF COMBINATION PROCESSES

In this case the expression in the curly brackets of (2.1) becomes complex and does not represent simply the saturated population difference. The solution of the system (3.1) for the general case now has too complicated a form. We therefore confine ourselves to an analysis of the interactions of only two transitions, b and c.

We consider the case when the generation is realized only in the transition b, and the field E_c is specified ($E_a = E_f = 0$). It follows from (3.1) and (2.1) that

$$\left(1+\frac{\Delta k_b}{2\gamma_b}\right)\delta\Omega_b = \delta k_b - \frac{1}{2}\frac{\Delta k_b}{2\gamma_b}\Delta\tilde{k}_b^{-1}\,\mathrm{Im}\,\Sigma\,|C|^2, \quad (4.1)$$

$$^3/_4\,\beta_b\delta n_b|B|^2 + {}^1/_2(\beta_c\delta n_c - \mathrm{Im}\,\{D_b^*\,\Sigma\})\,|C|^2 = \Delta\tilde{k}_b|D_b|^2 - \Delta n_b. \quad (4.2)$$

The effect is connected with the fact that an additional phase shift due to the combination processes is observed during the passage of the radiation through the medium. This phase shift is proportional to the intensity of the field combining with the given field. In addition, the field E_c gives rise to a "hump" on the plot of the grain of the transition if $\Delta n_c < \Delta n_b$, and to a "dip" of $\Delta n_c < \Delta n_b$, at a frequency close to the resonance of the combination process

$$\delta\Omega_b = \left(\delta k_b - \frac{1}{2}\frac{\Delta k_b}{2\gamma_b}\Delta\tilde{k}_b^{-1}\varkappa_c|C|^2\delta\Omega_c\right) \times\left[1+\frac{\Delta k_b}{2\gamma_b}\left(1-\frac{1}{2}\Delta\tilde{k}_b^{-1}\varkappa_b|C|^2\right)\right]^{-1} \quad (4.3)$$

where

$$\varkappa_c = \frac{1}{|D_y^c|^2}\left[\left(1+\frac{\gamma_y}{\gamma_c}\right)\delta n_c - \frac{\gamma_c}{\gamma_b}\delta n_b\right],$$

$$\varkappa_b = \frac{1}{|D_y^c|^2}\left[\left(1+\frac{\gamma_y}{\gamma_b}\right)\delta n_b - \frac{\gamma_b}{\gamma_c}\delta n_c\right]. \quad (4.4)$$

under analogous conditions, the formula for $\delta\Omega_c$ has a symmetrical form.

If $|\delta\Omega_b|, |\delta\Omega_c| \ll 1$, then we find from (4.3) and (4.4) that when $\Delta n_c \gg \Delta n_b > 0$ the sign of the pulling is opposite to the sign of $\delta\Omega_c$, and when $\Delta n_c \ll \Delta n_b$ the signs are the same. The magnitude of the effect depends essentially on the ratio γ_c/γ_y and on Δn_c and Δn_b. If $\gamma_c \sim \gamma_b \sim \gamma_y$, as is frequently the case in gases, and if $|C|^2 \sim 1$ and $\Delta n_c \sim \Delta n_b$, then, recognizing that $\Delta\tilde{k}^{-1}\Delta n \sim 1$, we find that the order of magnitude of the pulling is $(\Delta k/2\gamma_b)\,\delta\Omega_c$ and can be appreciable.

The effect is connected with the fact that an additional phase shift due to the combination processes is observed during the passage of the radiation through the medium. This phase shift is proportional to the intensity of the field combining with the given field. In addition, the field E_c gives rise to a "hump" on the plot of the gain of the transition if $\Delta n_c < \Delta n_b$, and to a "dip" of $\Delta n_c < \Delta n_b$, at a frequency close to the resonance of the combination process ($\delta\Omega_b \approx -(\gamma_c/\gamma_b)\,\delta\Omega_c$). Since the generation frequency at a fixed excitation level can be determined also by the condition that the gain per pass be equal to the losses due to reflection from the mirrors, then the result can be understood from this point of view.

The solution of Eq. (4.2) for the amplitude of the field generated in the transition b in the presence of a field E_c can be represented as before in the form (3.3), with the following values of the coefficients:

$$\left[\frac{\partial|B|^2}{\partial(\Delta n_b/\Pi_b'')}\right]_{\Delta n_b/\Pi_b''\approx 1} \equiv K_b'' = K_b\left[1-\frac{\gamma_c}{2\gamma_b}\,\mathrm{Im}\frac{D_b^*}{D_y^cD_b}|C|^2\right], \quad (4.5)$$

$$\Pi_b'' = \left[\Pi_b - \frac{1}{2}\left(\beta_c + \mathrm{Im}\frac{D_b^*\,D_c^*}{D_y^c}\delta n_c|C|^2\right)\right] \times\left[1-\frac{1}{2}\frac{\gamma_c}{\gamma_b}\,\mathrm{Im}\frac{D_b^*}{D_y^cD_b}|C|^2\right]^{-1}. \quad (4.6)$$

From (4.5) and (4.6) it follows, under the conditions $|\delta\Omega_b|, |\delta\Omega_c| \ll 1$ and $\Delta n_c > 0$, that the combination process lowers the generation threshold, but at the same time the value of K_B'' is decreased by an approximate factor $[1-(1/2)(\gamma_c^2/\gamma_b\gamma_y)|C|^2]^{-1}$. Information concerning these characteristics is contained by their variation with the value E_c, since these characteristics are connected with the relaxation constants. However, as follows directly from (4.2), at fixed $\Delta n_b > \pi_b' > \Pi_b''$, in spite of the fact that $K_b'' < K_b'$, the appearance of the combination process increases the power generated in the transition b if $\Delta n_c > (\gamma_c/\gamma_b)\Delta n_b$, and decreases it in the opposite case. If the field E_c is represented in the form of a traveling wave, then the coefficient $\frac{1}{2}$ is front of $|C|^2$ in the obtained formulas should be replaced by unity.

When $|\delta\Omega_c| \gtrsim 1$ the generation threshold may rise (if $\mathrm{Im}(D_b^* D_c^*/D_y^c) + \beta_c < 0$ and $\Delta n_c < 0$, and also as a result of the possible increase of $|\delta\Omega_b|$) solely as a result of the combination phenomena. This can lead to an interruption of the generation and can serve as an experimental confirmation of the appearance of combination processes.

By solving (4.2) together with the equation symmetrical to it for the field E_c, it is easy to obtain a solution for the case of combined generation in the system of both fields. This solution is too complicated, however and we state, without presenting the solution, that generation at the center of the line is realized only if both natural frequencies of the resonator are tuned to the centers of the lines of the corresponding transitions. Then the generation is realized at the center of the line in both transitions. In the opposite case, scanning of the proper frequency of the resonator at one of the transitions leads to a change in the generation frequency in the other transition. Other characteristics of generation of both transitions are likewise interrelated. The larger the ratio γ_c/γ_y or γ_b/γ_y, the stronger the coupling.

5. CONCLUSION

In this paper we did not consider effects connected with the motion of the particles that interact with the field. In the case of Doppler contours, the combination processes apparently cause the dips to have an asymmetrical form with singularities corresponding to the combination processes. We plan to consider this case in the future. The effects considered in Sec. 4 have, when considered by themselves, the same order of magnitude as the effects connected with the formation of the dips, since they are due to the same cause—relative deformations, of comparable magnitude, of the contours of the spectral lines of the atomic transitions. Thus, it is seen from Fig. 2, that a noticeable distortion of the contour of the spectral line of transition b can be attained at relatively weak fields (if the accuracy of the perturbation-theory methods is satisfactory). For example, when $|C|^2 \approx 0.025$ and $\delta\Omega_c = 0.4$ we have

$$\max\{\mathrm{Im}[\tfrac{1}{2}(\beta_c\delta n_c + \Sigma)/D_b\Delta n_b]\} \approx \tfrac{1}{4},$$

and its position does not coincide with the natural frequency of the transition. Since the gain for some transitions is so large that the resonator can become broadband, such a change in the dispersion properties of the transition leads to a noticeable change in the generation frequency.

Substituting the third-approximation solutions in the equations for the diagonal elements, which represent in the stationary regime the balance of the probabilities of the transitions due to different kinetic processes, and setting the corresponding values of Δn equal to 0 and ∓ 1, we can verify that the contributions due to the combination oscillations are connected with two-quantum transitions. Thus, the combination oscillation x is coupled to the transitions between levels 1 and 3 via the intermediate level 2, and κ is coupled via the intermediate level 4. On going from the lower state to the upper one, the photons $\hbar\Omega_a$ and $\hbar\Omega_b$ are absorbed in one act in the former case and in the latter case the quantum $\hbar\Omega_f$ is absorbed and the quantum $\hbar\Omega_c$ is emitted. The efficiency of the processes is determined by the presence of resonant intermediate levels. Such a phenomenon whereby the efficiency of third-harmonic generation is greatly enhanced if the medium has a transition that resonates with the second harmonic, was noted in [10]. Thus, under the conditions explained above, the contribution of these processes becomes noticeable. Thus, with increasing intensity of the field E_c an increase takes place in the probability of participation of the quanta of the field E_b in the two-photon process, which becomes manifest in the dispersion characteristics of the medium in the form of an additional resonance at a frequency connected with the energy conservation law in the two-photon transition.

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