

# Atomic quasienergy levels and high-frequency Stark effect in the field of strong polychromatic radiation with equidistant spectrum

G. I. Toptygina and É. E. Fradkin

Leningrad State University

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A method of determining the positions of resonances in the absorption coefficients, without laborious computation of these coefficients, is developed for individual components of strong polychromatic radiation with equidistant spectrum. Computer experiments show that the predicted positions agree well with the resonances on the calculated absorption curves of separate multimode-radiation components.

## 1. INTRODUCTION

Following the detailed investigations of the interaction of an atom with a strong monochromatic radiation, the first step towards consideration of polychromatic radiation was the development<sup>1</sup> of a mathematical formalism describing the interaction between a strong bichromatic field and two-level atoms in the quasiresonance approximation, with arbitrary frequency distances  $\Delta$  between the components of the bichromatic spectrum and with arbitrary component intensity ratio  $E_1^2/E_0^2$  (where  $E_0$  and  $E_1$  are the intensities of the perturbing and probing bichromatic fields). Simple analytic equations for the polarization of the medium were obtained in the following cases: a) both fields are saturating, but one is much weaker than the other ( $1 \ll \kappa E_1^2 \ll \kappa E_0^2$ , where  $\kappa$  is the saturation coefficient); b) for large  $\Delta \gg \gamma [1 + \kappa(E_1^2 + E_0^2)]$ , where  $\gamma = T_1^{-1}$  is the width of the atomic levels; c)  $E_1^2/E_0^2$  is arbitrary but  $\Delta/\gamma \rightarrow 0$ . The latter case was later realized in experiment.<sup>2,3</sup> The results turned out to agree fully with the theory.<sup>4,5</sup> This effect was named “anomalous” absorption.

A subradiation structure<sup>6,7</sup> was observed in 1978 in the absorption spectrum of a separated component of a bichromatic field: Absorption extrema were observed at frequency differences between the components  $\Delta$  of a bichromatic field, obeying the rule

$$\Delta_n = \pm \Delta_R / n, \quad n = 1, 2, 3, \dots \quad (1.1)$$

where  $\Delta_R$  is a characteristic frequency of the “field + atom” system, named the generalized Rabi frequency. As shown in Fig. 1, the maxima for one component corresponds to minima for the other component.<sup>3,8</sup> A theoretical expression for the Rabi frequency of a bichromatic was obtained in Ref. 4:

$$\Delta_R = (d/\hbar) (E_0^2 + E_1^2)^{1/2}, \quad (1.2)$$

where  $E_0^2$  and  $E_1^2$  are the intensities of the bichromatic-field components. The absorption coefficients of the components and the dependences on their frequency differences<sup>1)</sup> were also obtained in Ref. 4 and compared with the experimental dependences.<sup>7</sup> Good agreement was obtained between theory and experiment (see Fig. 1).

Peculiarities in the absorption coefficients of a strong bichromatic field, due to the influence of the strong field on the atomic-system dynamics, manifest themselves in two situations, saturation and nonlinear interference effect (NIEF). The latter causes a redistribution of the absorbed energy among the components (see Fig. 1), and produces negative absorption (amplification) without population in-

version and multiphoton parametric resonances at the frequency differences given by Eq. (1.1).

Sources of intense radiation in the optical band are lasers, a feature of which is an emission spectrum with equidistant or almost-equidistant longitudinal modes. It is therefore of interest to extend these investigations to include polychromatic radiation.

We shall show in the present paper that the extrema of the absorption coefficients of individual components are described by Eq. (1.1) also in the case of strong polychromatic radiation with an equidistant spectrum. We determine the generalized Rabi frequency  $\Delta_R$ . We consider two approaches to the interaction of a strong polychromatic field with a medium. Following Ref. 8, we investigate the connection between the multiphoton resonances (1.1) and the quasienergy level crossings of a “dressed atom” (Ref. 16) (atom + strong radiation). This enables us to obtain the Rabi frequency  $\Delta_R$  without laborious computations of the

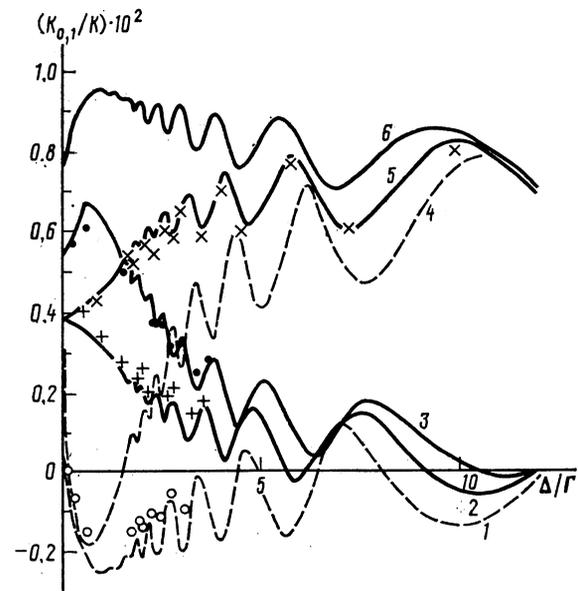


FIG. 1. Plots of the nonlinear relative absorption coefficient ( $K_i/K$ ) of the perturbing field ( $E_0$ ) (4,5,6) and of the test field ( $E_1$ ) (1,2,3) vs the detuning of the test field from the line centers at constant values of the parameters  $\omega_0 = \omega_{ab}$ ,  $\Gamma = \gamma$ ,  $G_0 = 11.1 \Gamma$  and at the following values of  $\rho = E_1/E_0$ :  $\rho = 1.2$  for curves 3 and 4;  $\rho = 1$  for curves 2 and 5;  $\rho = 0.8$  for curves 1 and 6. The experimental curves taken from Ref. 4 are marked on the figure.

TABLE I. Locations of parametric resonances in the absorption spectra of bichromatic-radiation components.

Value of $\Delta$	Number of resonance							
	2	3	4	5	6	7	8	9
$\Delta_{\text{calc}}$	10.8	6.0	4.3	3.3	2.8	2.4	2.0	1.8
$\Delta_c$	10.4	5.8	4.2	3.2	2.7	2.3	2.0	—
$\Delta_{\text{res}}$	9.52	5.83	4.26	3.36	2.78	2.38	2.07	1.84

Remarks. The positions of the resonances for modes 0 and 1 (max and min, respectively) were calculated with a computer ( $\Delta_{\text{calc}}$ );  $\Delta_c$ —experimental values taken from Ref. 4;  $\Delta_{\text{res}}$ —calculated from Eq. (3.9) at the parameter values  $\omega_{ab} = \omega_{ab}$ ,  $\Gamma = \gamma$ ,  $G_0 = 11.1 \Gamma$ ,  $G_1 = 1.1 G_0$ .

absorption coefficient. The quantitative estimates of Ref. 8, however, are obtained in an approximation that is adiabatic in the phase of the total field and is valid in a narrow range. We do not use here the adiabatic approximation, but find the quasienergy levels, and hence also the Rabi frequency  $\Delta_R$ , accurately in the framework of the formulated problem.

On the other hand, using a computer solution of a system of nonlinear algebraic equations that follow from the equation for the density matrix, we calculate the spectra of the absorption coefficients of individual components for three-, four-, and five-mode emission with an equidistant spectrum as functions of the intermode distance  $\Delta$ . These calculations can be called computer experiments. Good agreement is obtained between the computer-generated positions of the resonances on the absorption-coefficient curves of individual field components and those calculated from Eq. (1.1), in which  $\Delta_R$  is calculated from the intersection of the quasienergy levels (see Tables I and II below). We hope that our computations will stimulate physical experiments in this field.

## 2. CALCULATION OF THE QUASIENERGY OF AN "ATOM + MULTIMODE FIELD" SYSTEM

2.1. We consider the interaction between a two-level atom and multimode electromagnetic radiation with equidistant spectrum in the semiclassical formalism.

The equations for the probability amplitudes  $C_a$  and  $C_b$  of a two-level atomic system are:

$$i\hbar(dC_a/dt) = V_{ab}C_b \exp(i\omega_{ab}t),$$

$$i\hbar(dC_b/dt) = V_{ba}C_a \exp(-i\omega_{ab}t),$$
(2.1)

where

$$V_{ab} = V_{ba}^* = -1/2\hbar \left\{ \left[ \sum_{l=0}^{k-1} G_l \exp(-il\Delta t) \right] \times \exp[-i(p_0 + \omega_{ab})t] + \text{c.c.} \right\}$$

is the matrix element of the Hamiltonian of the interaction with the field in the dipole approximation;  $p_l = \omega_l - \omega_{ab}$ ;  $G_l = (dE_l/\hbar)\exp(i\varphi_l)$ .

We substitute  $V_{ab}$  and  $V_{ba}$  in (2.1). In accordance with the "rotating field" approximation, we discard the high-frequency exponentials  $\exp(\pm 2i\omega_{ab}t)$  and make the following change of variables:

$$C_a = S_a \exp[-i(p_0/2)t], \quad C_b = S_b \exp[i(p_0/2)t].$$
(2.2)

We transform the obtained system into a second-order differential equation with periodic coefficients for  $S_a$ , equivalent to the system (2.1).

Using the substitution

$$S_a = T[G(t)]^{\hbar}, \quad G(t) = \sum_{l=0}^{k-1} G_l \exp(-il\Delta t)$$
(2.3)

we obtain:<sup>2)</sup>

TABLE II. Locations of parametric resonances in the absorption spectra of four-mode radiation.

Mode number	Type of Resonance	Number of resonance						
		2	4	6	7	9	10	12
0	min	10.3	5.6	3.7	2.9	—	2.2	1.7
1	max	10.3	5.6	3.8	2.9	2.3	—	1.7
2	min	11.3	5.7	3.8	3.0	2.3	—	1.7
3	min	9.0	5.1	3.6	2.8	—	2.1	1.7
$\Delta_{\text{res}}$	—	10.7	5.27	3.5	3.0	2.33	2.1	1.75

Remarks. The resonance positions on the individual modes were calculated on a computer, the values of  $\Delta_{\text{res}}$  were calculated from Eq. (3.8) at the following parameter values:  $\omega_1 = \omega_{ab} - \Delta/2$ ,  $\Gamma = \gamma$ ,  $G_0 = 11.1 \Gamma$ ,  $G_1 = 1.2 G_0$ ,  $G_2 = 0.8 G_0$ ,  $G_3 = 0.7 G_0$ .

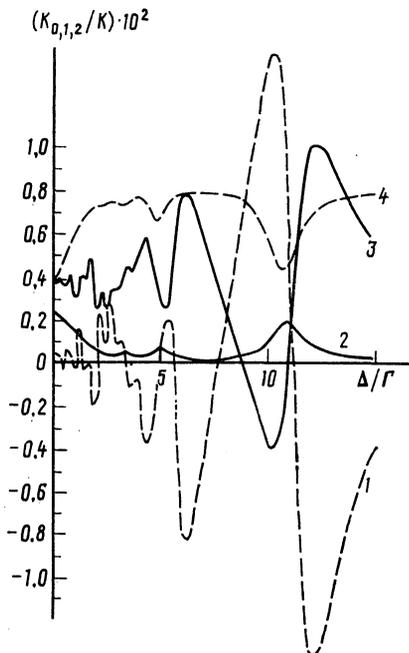


FIG. 2. Nonlinear absorption coefficients of three-mode radiation components vs the frequency difference between the fields in the cases of synchronized and nonsynchronized radiation (curves 2 and 3—absorption coefficients of zeroth and second harmonic; 1 and 4—of the first harmonic; 1 and 3—synchronized radiation; 2 and 4—nonsynchronized radiation; the parameter values are;  $\omega_1 = \omega_{ob}$ ,  $\Gamma = \gamma$ ,  $|G_0| = |G_1| = |G_2| = 11.1 \Gamma$ ; for nonsynchronized radiation  $G_0 = |G_0|$ ,  $G_1 = |G_1| \exp(-i\pi/2)$ ,  $G_2 = |G_2|$ ).

$$d^2T/dt^2 + QT = 0,$$

$$Q = \frac{(|G|^2 + p_0^2)}{4} + \frac{ip_0 G}{2G} + \frac{1 G}{2G} - \frac{3 G^2}{4 G^2}. \quad (2.4)$$

Equation (2.4) is a second-order differential equation with a periodic coefficient. It is called the Hill equation and has been thoroughly investigated in the mathematical literature.<sup>17</sup>

We examine now the dependence of the upper-level population  $|C_a|^2$  on the phase constants  $\varphi_l$ . Using a time-origin shift

$$t = t' + (\varphi_1 - \varphi_0)/\Delta,$$

we can show that the functions  $|G(t')|$  and  $Q(t')$  depend on the phase combinations

$$\chi_l = \varphi_l - l\varphi_1 + (l-1)\varphi_0$$

( $l = 1, 2, \dots$ ) (see Fig. 2). Consequently the functions  $|T(t')|^2$  and, ultimately  $|C_a|^2$ , also depend on the phases  $\chi_l$ . In the bichromate case we have  $l = 1$ ,  $\chi_1 = 0$ , and there is no dependence on the initial phases. This agrees with theoretical calculations,<sup>18</sup> where the dependence of the upper-level population on the experimental constant phase difference  $\varphi_1 - \varphi_0$  was of the components was attributed to a contribution of high-frequency terms neglected in the equations for the probability amplitudes in the rotating-field approximation. According to the foregoing, the theory developed in Ref. 19 in the rotating-field approximation, which leads to a dependence of the upper-level population  $|C_a|^2$  on the initial

component phase difference  $\varphi_1 - \varphi_0$  of the bichromate is incorrect.

It is interesting to note that in a polychromate field with an arbitrary number of components, the phases of which are synchronized, i.e., the condition  $\varphi_l = \varphi_0 + l\varphi$ , is met, and the phase combinations  $\chi_l = 0$ . Thus, the interaction of the atoms with the synchronized radiation does not depend on the constant phases of the fields.

2.2. We expand the periodic function  $Q$  contained in (2.4) in a Fourier series and separate the zeroth term. As a result, (2.4) takes the form

$$d^2T/dt^2 + T \left[ Q_0 + \sum_{n=-\infty}^{\infty} Q_n \exp(in \Delta t) \right] = 0. \quad (2.5)$$

According to the Floquet-Lyapunov theorem, Hill's equation has a solution in the form

$$T = \exp(\mu t) \sum_{n=-\infty}^{\infty} A_n \exp(-im \Delta t), \quad (2.6)$$

where  $\mu$ , called the characteristic exponent of the Hill equation,<sup>3</sup> takes on two values  $\mu_1$  and  $\mu_2$ .

2.3. We obtain the characteristic exponents  $\mu_{1,2}$  by a method described in Refs. 20–22. We must generalize here the case of the even real function  $Q$  considered in these references to include our situation, in which  $Q$  is a complex periodic function having no definite parity. We substitute the solution (2.6) in Hill's equation and equate coefficients of like powers of the exponentials:

$$A_m [(\mu - im\Delta)^2 + Q_0] + \sum_{n=-\infty}^{\infty} A_{m+n} Q_n = 0.$$

Dividing the equation by the coefficients of  $A_m$ , we obtain an infinite system of homogeneous linear algebraic equations:

$$A_m + \sum_{n=-\infty}^{\infty} A_{m+n} Q_n [(\mu - im\Delta)^2 + Q_0]^{-1} = 0. \quad (2.7)$$

This system has a solution only if its principal determinant  $D(i\mu)$  is zero. From the condition  $D(i\mu) = 0$  we can determine the value of  $\mu$ .

Consider the convergence of the principal determinant  $D(i\mu)$  of the system (2.7). An infinite determinant converges absolutely if: 1) the product of the diagonal terms converges absolutely and 2) the sum of the off-diagonal elements converges absolutely. The condition 1) is met in our case, since the product of the diagonal elements is equal to unity.

To verify the satisfaction of condition 2), we must prove the convergence of the series

$$\sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} |Q_n [(\mu - im\Delta)^2 + Q_0]^{-1}| = \sum_{n=-\infty}^{\infty} |Q_n| \sum_{m=-\infty}^{\infty} |(\mu - im\Delta)^2 + Q_0|^{-1}. \quad (2.8)$$

The sum over  $n$  converges, since it is made up of coefficients of a Fourier series of a function that is differentiable more than once. The sum over  $m$  also converges, since the denomi-

nator contains  $m^2$  and the numerator unity. Thus,  $D(i\mu)$  converges absolutely for all finite values of  $\mu$ , provided only that the denominators  $(\mu - im\Delta)^2 + Q_0$  differ from zero.

The determinant  $D(i\mu)$  is a function of complex variable. Let us examine its parity. Replacement of  $\mu$  by  $-\mu$  is equivalent to replacement of  $m$  by  $-m$ , and since  $m \in (-\infty, \infty)$  it follows that  $D(i\mu) = D(-i\mu)$ , i.e.,  $D(i\mu)$ , is an even function with period  $\Delta$  relative to  $i\mu$ .

We omit the details of the calculation of the  $\mu_{1,2}$  that satisfy the condition  $D(i\mu) = 0$ , since they are described in the cited references. For the sought  $\mu$  we have the relation

$$\cos(2\pi i\mu\Delta^{-1}) = 1 - D_0 [1 - \cos(2\pi\Delta^{-1}Q_0^{1/2})] \equiv B, \quad (2.9)$$

where  $D_0 = D(i\mu)|_{\mu=0}$ .

The left-hand side of (2.9) contains the cosine of a complex quantity. We use therefore Euler's formula and solve the quadratic equation for  $\exp(2\pi\mu\Delta^{-1})$ :

$$\exp(2\pi\mu\Delta^{-1}) = B \pm i(1 - B^2)^{1/2}. \quad (2.10)$$

Expressions (2.10) are the exact values of the multipliers of the Hill equation (2.4).

**2.4.** In Sec. 2.3 we found the characteristic exponents  $\mu_1$  and  $\mu_2$  and the periodic functions  $\psi(t)$  contained in the solution (2.6) of the Hill equation (2.4). We can therefore express, on the basis of equations (2.2), (2.3), and (2.6) the solutions of Eqs. (2.1) in the form

$$\begin{aligned} C_a &= [G(t)]^{1/2} \exp[-i(p_0/2)t] \{ \exp(\mu_1 t) \psi_1(t) + \exp(\mu_2 t) \psi_2(t) \}, \\ C_b &= [G^*(t)]^{1/2} \exp[i(p_0/2)t] \{ \exp(\mu_1 t) \psi_3(t) + \exp(\mu_2 t) \psi_4(t) \}. \end{aligned} \quad (2.11)$$

The connection between the function pairs  $\psi_1(t)$  and  $\psi_3(t)$ ;  $\psi_2(t)$  and  $\psi_4(t)$  is determined by Eqs. (2.1).

From (2.1) follows the obvious stationarity condition

$$d(|C_a|^2 + |C_b|^2)/dt = 0. \quad (2.12)$$

Substitution of the functions (2.11) in (2.12) leads to the conclusion that the equality (2.12) holds only if  $\mu_1$  and  $\mu_2$  are pure imaginary. Using this fact in (2.9) and (2.10), we find that  $B$  is real and

$$B^2 \equiv \{1 - D_0 [1 - \cos(2\pi\Delta^{-1}Q_0^{1/2})]\}^2 \leq 1. \quad (2.13)$$

It follows in this case from (2.10) that  $\mu_1 = -\mu_2 = i\beta = i\Delta(2\pi)^{-1} \arccos B$ .

In accord with Ref. 16, the quasienergy levels are

$$W_\alpha + \hbar \epsilon_{n_\alpha} = W_\alpha + [\beta + p_\alpha/2 \pm n_\alpha \Delta] \hbar,$$

$$W_\alpha + \hbar \epsilon_{m_\alpha} = W_\alpha + [-\beta + p_\alpha/2 \pm m_\alpha \Delta] \hbar.$$

For the upper states we have  $\alpha = a$  and  $p_\alpha = -p_0$  and for the lower  $\alpha = b$  and  $p_\alpha = p_0$ ;  $n_\alpha$  and  $m_\alpha$  are positive integers. Therefore the levels  $a$  and  $b$  each split in a multimode field into two systems of sublevels. By analogy with the high-frequency Stark effect,<sup>23</sup> the quasienergy level splitting effect described above should be referred to as a high-frequency Stark effect generalized to include the case of polychromatic radiation.

The conditions for the quasienergy-level crossing are the same for both states:<sup>8</sup>

$$W_\alpha + [\beta + p_\alpha/2 \pm n_\alpha \Delta] \hbar = W_\alpha + [-\beta + p_\alpha/2 \pm m_\alpha \Delta] \hbar, \quad \alpha = a, b.$$

It follows hence that

$$(m_\alpha - n_\alpha) \Delta \equiv n \Delta = \pm 2\beta, \quad n = 1, 2, 3, \dots \quad (2.14)$$

Comparing (2.14) with (1.1) we can conclude that (2.14) is a generalization of the resonance condition (1.1) to include polychromatic radiation. The resonances in the absorption coefficients of the individual modes are due to transitions that begin and end at the quasienergy level-crossing points of the upper and lower states. The frequency  $2\beta$  is called the generalized Rabi frequency.

Our definition of the generalized Rabi frequency differs from that introduced in Ref. 24. The point is that if  $\beta$  depends on  $\Delta$  Eq. (2.14) must be regarded as an equation for  $\Delta$ . We shall show below that  $D_0$  depends weakly on  $\Delta$  and that in most cases the characteristic exponents are solutions of a quadratic equation, so that two sequences of resonance frequencies can be obtained.<sup>4)</sup> The resonance frequencies of these sequences for  $n = 1$  were in fact called in Ref. 24 generalized Rabi frequencies. A quantitative estimate of the generalized Rabi frequency was obtained in Ref. 24 only with allowance for the diagonal terms of an infinite determinant of an algebraic-equations system equivalent to the equations for the density matrix. Comparison with our results shows that this approximation is inadequate.

Our definition of the generalized Rabi frequency follows directly from the formalism that describes the quasienergy levels, and has a clearer physical meaning. In addition, under our definition there exists for any set of fields only one generalized Rabi frequency which is the difference of the characteristic exponents (2.10) of the Hill equation and depends, in the general case, on the amplitudes and constant phases of the fields, on the intermode spacing  $\Delta$ , on the position  $p_0$  of the spectrum relative to the line center, and on the dipole moment  $d$  of the transition.

### 3. ANALYSIS OF THE EXPRESSIONS FOR THE GENERALIZED RABI FREQUENCY AND THE PARAMETRIC RESONANCES

**3.1.** The expression (2.10) for  $\mu$  contains the determinant  $D_0$  which depends on the Fourier components of the function  $Q$  contained in Eq. (2.4). Let us calculate the Fourier components of the function  $Q$ . We put  $y = \exp(-i\Delta t)$  and transform  $G(t)$  into

$$G(y) = \sum_{l=0}^{k-1} G_l y^l = G_{k-1} \prod_{j=1}^{k_s} (y - a_j)^{s_j} = \mathcal{P}(y), \quad (3.1)$$

where  $k_s$  is the number of the different roots of the polynomial  $\mathcal{P}(y)$  of degree  $k - 1$ ,  $s_j$  is the multiplicity of the  $j$ th root, and  $a_j$  is the root of the polynomial.

By definition,

$$\begin{aligned} Q_n &= (2\pi)^{-1} \int_{-\pi}^{\pi} Q \exp(-in\Delta t) d(\Delta t), \quad n \geq 0; \\ Q_{-n} &= (2\pi)^{-1} \int_{-\pi}^{\pi} Q \exp(in\Delta t) d(\Delta t), \quad n > 0. \end{aligned} \quad (3.2)$$

We make in the integrals (3.2) the change of variable  $x = \exp(i\Delta t)$  and transform to integration on the complex plane along the contour  $|x| = 1$ . As a result we obtain

$$Q_n = 1/4 \sum_{\substack{l=0 \\ 0 \leq n \leq k-1}}^{k-1-n} G_l G_{l+n}^* + (p_0^2/4) \delta_{n0} + (p_0 \Delta/2) \sum_{\substack{j=1 \\ |a_j| < 1}}^{k_s} s_j a_j^n$$

$$\begin{aligned}
& + (\Delta^2/2) \sum_{\substack{j=1, \\ |a_j| < 1}}^{k_s} s_j n a_j^n + (\Delta^2/4) \sum_{\substack{j=1, \\ |a_j| < 1}}^{k_s} s_j^2 (n+1) a_j^n \\
& + (\Delta^2/2) \sum_{j=1}^{k_s} \sum_{\substack{m=1, \\ |a_m| < 1, \\ m \neq j}}^{k_s} s_j s_m a_m^{n+1} (a_m - a_j)^{-1}, \quad n \geq 0;
\end{aligned} \tag{3.3}$$

$$\begin{aligned}
Q_{-n} = & 1/4 \sum_{\substack{l=0 \\ 1 \leq n \leq k-1}}^{k-1-n} G_l * G_{l+n} - (p_0 \Delta/2) \sum_{\substack{j=1, \\ |a_j| > 1}}^{k_s} s_j a_j^{-n} \\
& + (\Delta^2/2) \sum_{\substack{j=1, \\ |a_j| > 1}}^{k_s} s_j n a_j^{-n} \\
& + (\Delta^2/4) \sum_{\substack{j=1, \\ |a_j| > 1}}^{k_s} s_j (n-1) a_j^{-n} \\
& + (\Delta^2/2) \sum_{j=1}^{k_s} \sum_{\substack{m=1, \\ m \neq j, \\ |a_m| > 1}}^{k_s} s_j s_m a_j^{-1} a_m^{-n}, \quad n > 0.
\end{aligned} \tag{3.4}$$

We write for  $Q_0$  a separate equation

$$\begin{aligned}
Q_0 = & 1/4 \left( \sum_{l=0}^{k-1} |G_l|^2 + p_0^2 \right) + (p_0 \Delta/2) \sum_{\substack{j=1, \\ |a_j| < 1}}^{k_s} s_j + (\Delta^2/4) \sum_{\substack{j=1, \\ |a_j| < 1}}^{k_s} s_j^2 \\
& + (\Delta^2/2) \sum_{j=1}^{k_s} \sum_{\substack{m=1, \\ m \neq j, \\ |a_m| < 1}}^{k_s} s_j s_m a_m (a_m - a_j)^{-1}.
\end{aligned} \tag{3.5}$$

Note that in Eqs. (3.3)–(3.5) the symbol

$$\sum_{\substack{j=1, \\ |a_j| \leq 1}}^{k_s}$$

must be taken to mean summation over only those numbers  $j$  for which  $|a_j| \geq 1$ . In addition, it follows from (3.3) and (3.4) that the series

$$\sum_{n=-\infty}^{\infty} |Q_n|$$

of (2.8) converges if  $|a_j| \neq 1$ . Our equations for  $Q_{\pm n}$  are therefore not valid when the root (3.1) of the polynomial  $\mathcal{P}(y)$  (3.1) has a unity modulus. In these cases the Fourier coefficients must be calculated in a different manner, but everything said above is valid here, too.

The simplest expressions for  $Q_0$  (at arbitrary  $k$ ) can be obtained in two cases:

1) all the roots of the polynomial  $\mathcal{P}(y)$  (3.1) are larger than unity in absolute value, and then

$$Q_0 = 1/4 \left[ \sum_{l=0}^{k-1} |G_l|^2 + p_0^2 \right], \tag{3.6}$$

2) all the roots of the polynomial  $\mathcal{P}(y)$  are smaller than unity in absolute value, and then

$$Q_0 = 1/4 \left[ \sum_{l=0}^{k-1} |G_l|^2 + p_{k-1}^2 \right], \tag{3.7}$$

for in this case

$$\sum_{j=1}^{k_s} s_j = k-1,$$

i.e., to the degree of the polynomial  $\mathcal{P}(y)$ , and

$$\sum_{\substack{j=1, \\ |a_j| < 1}}^{k_s} s_j^2 + 2 \sum_{j=1}^{k_s} \sum_{\substack{m=1, \\ m \neq j, \\ |a_m| < 1}}^{k_s} s_j s_m a_m (a_m - a_j)^{-1} = (k-1)^2.$$

**3.2.** In cases (3.6) and (3.7)  $Q_0$  is real and positive. Recognizing that the parameter  $B$  (2.9) is real, we can state in these cases that  $D_0$  is a real number. The region of validity of this statement can apparently be substantially expanded.

Computer calculations of the determinant  $D_0$  for a bichromatic field have shown that  $D_0$  is close to unity. Assume that this situation obtains also in the case of several fields with equidistant spectra. We put  $D_0 = 1$  and determine on the basis of (2.9), (2.10), and (2.14) the positions of the resonances of the absorption coefficients:

$$\Delta_{res} = 2Q_0^{1/2}/n, \quad n=1, 2, 3, \dots \tag{3.8}$$

The approximate values  $\beta = Q_0^{1/2}$  require no computer calculation, and agree well with the curves of Figs. 1 and 2 (see also Fig. 4 below), as demonstrated in Tables I and II. They permit a relatively simple calculation of the positions of the resonances on the absorption-coefficient curves of the polyharmonic radiation component, in sufficiently large ranges of the parameters.

Let us examine the quasienergy level-crossing scheme. We take the particular case of three fields with parameters  $G_1 = 11.1 \Gamma$ ,  $G_0 = G_2 = 0.25 G_1$ ,  $\Gamma = \gamma$ , and  $p_0 = -\Delta$ . At  $k = 3$  the polynomial  $\mathcal{P}(y)$  is of second degree and its roots are therefore easy to determine:

$$a_{1,2} = -G_1(2G_2)^{-1} \pm \{ [G_1(2G_2)^{-1}]^2 - G_0 G_2^{-1} \}^{1/2}.$$

In the example considered we have

$$\beta = 1/2 \left[ \sum_{l=0}^2 |G_l|^2 + 2\Delta^2 a_m (a_m - a_j)^{-1} \right]^{1/2}, \quad |a_m| < 1, \quad |a_j| > 1$$

and  $2a_m(a_m - a_j)^{-1} = -0.155$ . It follows from this expression that  $\beta$  depends on  $\Delta$ . Thus, for  $\Delta = 0$  we have  $2\beta = 11.77 \Gamma$ , and for  $\Delta = 11 \Gamma$  we have  $2\beta \approx 11 \Gamma$ . The quasienergy levels are correspondingly not straight lines but parabolas, as shown in Fig. 3.

In Fig. 3 are separated the quasienergy level-crossing points with  $n = 2$  (see the dash-dot line). The resonant transitions are those beginning at the upper-state quasienergy level crossing points and terminate at lower-level quasienergy crossing points. Thus, the transition

$$W_a + \beta \hbar/2 \rightarrow W_b + \beta \hbar/2$$

with frequency

$$\omega = (W_a - W_b)/\hbar = \omega_{ab}$$

will be resonant and will lead to a maximum or minimum at

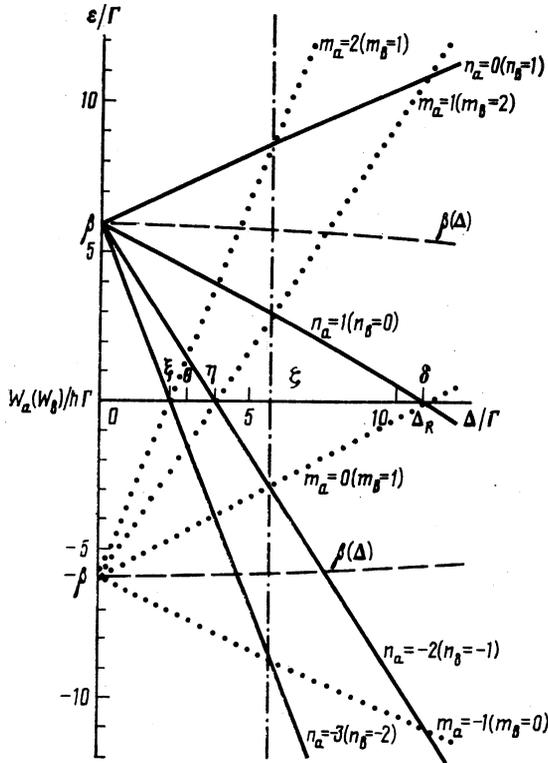


FIG. 3. Quasienergy level-crossing diagram for the upper and lower states of a two-level atom. The subscripts "a" and "b" label the upper and lower levels; the numbers on the axes correspond to the case of three fields. The quasienergy levels  $\varepsilon_{na} = \beta + \Delta/2 \pm n_a \Delta$  and  $\varepsilon_{nb} = \beta - \Delta/2 \pm n_b \Delta$  are shown by solid lines, and  $\varepsilon_{ma} = -\beta + \Delta/2 \pm m_a \Delta$  and  $\varepsilon_{mb} = -\beta - \Delta/2 \pm m_b \Delta$  by dotted ones. A dashed line shows the dependence of  $\beta$  on  $\Delta$ . The Greek letters on the  $\Delta/\Gamma$  axis denote the crossing points of quasienergy levels with different values of  $n$  and with corresponding values of  $\Delta/\Gamma = 2\beta/n$ ; at the point  $\delta$ :  $n = 1$ ,  $\Delta/\Gamma = 2\beta \equiv \Delta_R$ , at the point  $\zeta$ :  $n = 2$ ,  $\Delta/\Gamma = \beta$ , at the point  $\eta$ :  $n = 3$ ,  $\Delta/\Gamma = 2\beta/3$ , at the point  $\theta$ :  $n = 4$ ,  $\Delta/\Gamma = \beta/2$ , and at the point  $\xi$ :  $n = 5$ ,  $\Delta/\Gamma = 2\beta/5$ .

the first harmonic whose frequency coincides with  $\omega_{ab}$ . Other similar transitions can also be considered. The transition

$$W_a + \beta\hbar/2 \rightarrow W_b - \beta\hbar/2$$

at the frequency  $\omega = \omega_{ab} + \beta$  and those similar to it cause a resonance in the second component having this frequency. The transition

$$W_a - \beta\hbar/2 \rightarrow W_b + \beta\hbar/2$$

at the frequency  $\omega = \omega_{ab} - \beta$  gives rise to a resonance in the zeroth harmonic. One can consider transitions that cause resonances in combination tones, etc.

The resonances are numbered from the line wing towards the center, so that by increasing  $n$ , i.e., by decreasing  $\Delta$ , we obtain the quasienergy level crossings and consequently also the resonances on the absorption-coefficient curves of individual components of multimode radiation with equidistant spectrum with ever increasing number  $n$ , until these resonances merge (i.e., until the sum of the half-widths of two neighboring resonances exceeds the distance between resonances).

Since the quasienergy levels in our case are parabolas whose branches are oppositely directed (the coefficient of  $\Delta^2$  is negative), the quasienergy levels with numbers  $n_a = m_a = 0, 1, 2, \dots$  intersect. Resonance transitions can therefore exist at  $n = 0$ . If  $\Delta^2$  has a positive coefficient, the first few

resonances may be absent. An example of the absence of a resonance at  $n = 1$  is contained in Table I. In the three-field case chosen by us a zero resonance is possible at  $\Delta_0 = 29.9 \Gamma$ . The error in the determination of the resonances by Eq. (3.8), compared with the resonance positions of the resonances on the computer-calculated absorption-coefficient curves does not exceed 3% for  $n > 1$  and 9% for  $n = 1$ .

**3.3.** We consider two examples of radiation containing equidistant monochromatic strong fields. We shall use the approximate equation (3.8) in the analysis of the resonance positions.

a)  $k = 2$ . Two mode field—example of radiation synchronized at any instant of time. In the case of a bichromate  $\mathcal{P}(y)$  is a polynomial of first degree, and its root is  $a_1 = -G_0/G_1$ . If  $|a_1| > 1$ , i.e.,  $G_0 > G_1$ , we have

$$\Delta_{\text{res}} = 2Q_0^{1/2}/n = \left( \sum_{l=0}^1 |G_l|^2 + p_0^2 \right)^{1/2} / n,$$

and the condition obtained for resonance to occur at  $p_0 = 0$  coincides with the result of our earlier paper.<sup>4</sup> In the same paper we have compared the experimental data with those obtained with a computer and from Eq. (1.2) at  $G_0 > G_1$ . If  $|a_1| < 1$ , i.e.,  $G_0 < G_1$ , then

$$2Q_0^{1/2} = \left( \sum_{l=0}^1 |G_l|^2 + p_l^2 \right)^{1/2}.$$

Table I shows the case  $p_0 = 0$  and consequently  $|p_1| = \Delta$ . The condition (3.8) for quasienergy level crossing is here an equation for  $\Delta$ . Its solution is

$$\Delta_{\text{res}} = \left[ \sum_{l=0}^1 |G_l|^2 (n^2 - 1)^{-1} \right]^{1/2}. \quad (3.9)$$

It follows from (3.9) that at  $G_0 < G_1$  there is no first resonance, and the numbering of the resonances begins with the second (see Table I). The error in the position of resonances as given by (3.9) obtained from (3.8) does not exceed 5% for  $n \geq 3$ . The error for  $n = 2$  is 12%.

b)  $k = 4$ . For the four fields in the example given in Table II, the root  $|a_1| < 1$  and the roots  $a_2$  and  $a_3$  are complex conjugates, with  $|a_2| = |a_3| > 1$ . We have accordingly

$$2Q_0^{1/2} = \left[ \sum_{l=0}^3 |G_l|^2 + p_l^2 + 4\Delta^2 a_l \operatorname{Re}(a_1 - a_2)^{-1} \right]^{1/2},$$

$p_1 = -(\Delta/2)$  and  $\Delta_{\text{res}} = 439.8597/(n^2 - 0.15)^{1/2}$ . The error in the calculation of the resonance positions does not exceed 6%. The position of the first resonance, calculated from the approximate equation, is  $22.7 \Gamma$ .

The examples given here have shown that our assumption that  $D_0$  is equal to unity is valid in a rather wide range of the parameters of our problem, and leads to good agreement between the compared resonance positions on the absorption-coefficient curves of individual polychromatic-radiation harmonics calculated from Eq. (3.8), on the one hand, and those obtained by computer solution of the equations for the density matrix.

#### 4. CALCULATION OF THE ABSORPTION COEFFICIENT OF AN INDIVIDUAL HARMONIC OF POLYCHROMATIC RADIATION

**4.1. Formulation of problem.** Multimode radiation with an equidistant spectrum is incident on a medium consisting of two-level atoms with a homogeneously broadened transition line.<sup>5)</sup> The characteristics of the medium are the transition half width  $\Gamma = T_2^{-1}$ , the level widths  $\gamma_a$  and  $\gamma_b$ , the transition dipole moment  $d$ , and the transition frequency  $\omega_{ab}$ . To simplify the equations we assume equal level widths,  $\gamma_a = \gamma_b = \gamma = T_1^{-1}$ . The radiation is characterized by an intermode distance  $\Delta$ , a zeroth-harmonic frequency  $\omega_0$  (the frequency of the  $\omega_l$ th harmonic is  $\omega_l = \omega_0 + l\Delta$ ), and radiation component intensities  $E_l$  and phases  $\varphi_l$ .

No synchronism conditions are imposed on the field-component phases in the solution of the equations for the density-matrix elements. The radiation can either be synchronized or not.

The difference between the half width  $\Gamma$  of the homogeneously broadened transition and  $\gamma$  makes it possible to take phenomenologically into account the dephasing processes in the model of homogeneous absorption-line broadening. We denote by  $\lambda_a$  and  $\lambda_b$  the stationary pumps to the levels  $a$  and  $b$ , respectively (i.e., the number of atoms excited per unit time in a unit volume into a given state).

The equations for the density matrix element of a two-level system are, in the rotating-field approximation,

$$\begin{aligned} \sigma &= \rho_{ab} \exp(i\omega_0 t), \\ N &= \rho_{aa} - \rho_{bb}, \quad \lambda = \lambda_a - \lambda_b. \end{aligned} \quad (4.1)$$

We introduce the dimensionless quantities  $\tau = \Gamma t$ ,  $f = \Delta/\Gamma$ ,  $\kappa = \gamma/\Gamma$ ,  $f_0 = (\omega_{ab} - \omega_0)/\Gamma$ . The system of equations takes then the form

$$\begin{aligned} d\sigma/d\tau + (1 + if_0)\sigma &= -(i/2)(V^*/\Gamma)N, \\ dN/d\tau + \kappa N &= \lambda/\Gamma - i[(V/\Gamma)\sigma - (V^*/\Gamma)\sigma^*], \end{aligned} \quad (4.2)$$

where

$$V = \sum_{l=0}^{k-1} G_l \exp(ilf\tau).$$

**4.2.** We express the stationary solution of the system in terms of Fourier series

$$\begin{aligned} N &= N_0 + \left( \sum_{n=1}^{\infty} N_n \exp(-inf\tau) + \text{c.c.} \right), \\ \sigma &= \sum_{n=-\infty}^{\infty} \sigma_n \exp(-inf\tau), \end{aligned} \quad (4.3)$$

and take  $N_0$  to be a real quantity.

We substitute the series (4.3) in the system (4.2) and equate coefficients of like arguments of the exponentials. We substitute in turn the expressions obtained for  $\sigma_{\pm n}$  in the equation for  $N_n$ . After gathering like terms we obtain

$$\sum_{j=1}^{k-1} N_{n+j} B_{n,j} F_n^{-1} + N_n + \sum_{j=1}^{k-1} N_{n-j} D_{n,j} F_n^{-1} = \lambda (\gamma F_0)^{-1} \delta_{n0}, \quad (4.4)$$

where

$$\begin{aligned} F_n &= 1 - inf\kappa^{-1} + \sum_{m=0}^{k-1} |G_m|^2 (2\Gamma\gamma)^{-1} \{ (1+i[f_0 - (n+m)f])^{-1} \\ &\quad + (1-i[f_0 + (n-m)f])^{-1} \}, \\ B_{n,j} &= \sum_{m=j}^{k-1} G_m G_{m-1}^* (2\Gamma\gamma)^{-1} \{ (1+i[f_0 - (n+m)f])^{-1} \\ &\quad + (1-i[f_0 + (n-m+j)f])^{-1} \}, \\ D_{n,j} &= \sum_{m=j}^{k-1} G_{m-j} G_m^* (2\Gamma\gamma)^{-1} \{ (1+i[f_0 - (n+m-j)f])^{-1} \\ &\quad + (1-i[f_0 + (n-m)f])^{-1} \}. \end{aligned}$$

Since  $N_{-j} = N_j^*$  in (4.4) at  $n = 0$ , and the coefficients of  $N_{\pm j}$  are complex conjugate, this equation is real.

The system (4.4) is an infinite inhomogeneous set of linear algebraic equations. Let us prove that it has a solution. We construct the principal determinant of the system. This is an infinite ribbon-like determinant with a ribbon width  $2k - 1$  terms, and with all diagonal terms equal to unity. Obviously, the determinant is not equal to zero, since all the rows of the ribbon determinant are linearly independent.

Let us prove that the infinite determinant converges. The condition 1) (see Sec. 2 above) is met, since the aforementioned product is in this case also equal to unity. The condition 2) is met if the series

$$\sum_{n=-\infty}^{\infty} \sum_{j=1}^{k-1} |B_{n,j} + D_{n,j}|$$

converges. This is indeed the case, since the relations  $|B_{n,j}|, |D_{n,j}| < 1/n^2$  are satisfied if  $n$  is large enough.

The infinite determinant of the system (4.4) is thus nonzero and converges. The system has therefore a solution.

The system (4.4) was solved on a computer. The number  $n$  is chosen to satisfy a given computation accuracy, after which a solution is obtained for a finite system of  $2n + 1$  linear algebraic equations with  $2n + 1$  unknowns; the system contains equations with numbers from  $-n$  to  $+n$ . Equations with numbers larger than  $n$  and smaller than  $-n$  were not considered. In the chosen equations, values of  $N_m$  with numbers larger than  $n$  and smaller than  $-n$  were assumed to be zero. The solutions  $\sigma_{\pm l}$  at the field frequencies  $l = 0, 1, 2, \dots, k - 1$  depend on  $N_l$  having the same number. A value  $n = 20$  to  $25$  was necessary to calculate  $N_l$  for  $k = 2$  to  $6$ .

**4.3.** Knowing  $N_n$ , we can determine  $\sigma_m$  and determine with the latter the absorption coefficient of the harmonic numbered  $m$  in the presence of other strong fields:

$$K_m = -8\pi\omega_{ab} d^2 (\epsilon_0^{1/2} c \hbar \Gamma)^{-1} \text{Im} \{ \sigma_m \Gamma (G_m^*)^{-1} \}. \quad (4.5)$$

A computer was used to determine the relative absorption coefficient  $K_m/K$ , where  $K$  is the linear absorption coefficient at the line center,

$$K = d^2 \lambda 4\pi\omega_{ab} (\hbar \Gamma \gamma \epsilon_0^{1/2} c)^{-1},$$

$d$  is the matrix element of the transition,  $\epsilon_0$  is the dielectric constant of the medium without allowance for the resonance-transition contribution, and  $c$  is the speed of light in vacuum.

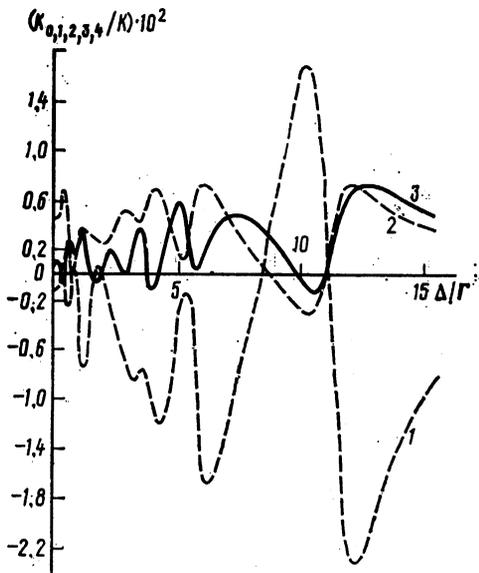


FIG. 4. Dependences of the nonlinear absorption coefficients on the components of five-mode synchronized radiation from the following parameter values:  $\omega = \omega_{ab}$ ,  $\Gamma = \gamma$ ,  $G_2 = 11.1$ ,  $\Gamma$ ,  $G_1 = G_3 = 1.2$ ,  $G_2$ ,  $G_0 = G_4 = 0.8$ ,  $G_2$ , curve 1— $K_2/K$ ; 2— $K_{1,3}/K$ ; 3— $K_{0,4}/K$ .

In addition to the absorption coefficient we can, knowing  $\sigma_n$  and meaning also  $\rho_{ab}$  (4.1), find the polarization (the response of the medium to the action of a multicomponent field):

$$P = d(\rho_{ab} + \rho_{ba}) = d \sum_{n=-\infty}^{\infty} \{ \sigma_n \exp[-i(\omega_0 + n\Delta)t] + \sigma_n^* \exp[i(\omega_0 + n\Delta)t] \}. \quad (4.6)$$

It follows from (4.6) that the response of the medium is formed not only at the frequencies of the fields themselves, but also at the combination frequencies  $\omega_0 \pm n\Delta$ .

Note the following circumstance: the spatial structure plays an essential role for optical fields. The spatial dependence was not specified in the expression for the perturbation. This makes it possible to use expressions (4.4)–(4.6) to solve a large number of problems involving the interaction of fields with a medium having a homogeneously broadened absorption line (a detailed explanation of this question is contained in Ref. 4).

As shown by the computer calculations (Figs. 1, 2, 4) the plot of the absorption coefficient of a component of multimode radiation with an equidistant spectrum consists of a set of resonances whose positions obey the conditions (2.14). Figure 2 shows absorption-coefficient plots of the harmonics of synchronized and not synchronized three-mode radiation. It can be seen that the absorption coefficients of the individual harmonics of complex radiation depend on the specific set of field phases.

The positions of the resonances on the absorption-coefficient curves, calculated with a computer and from Eq. (3.8), are given in Tables I and II. The type of resonance—maximum or minimum—is also indicated. All the absorption-coefficient resonances of an individual component of

complex radiation satisfying the condition (2.14) are of the same type.

The absorption-coefficient curves have regions of negative absorption (gain) of individual multimode-radiation components (see Figs. 1, 2, and 4). The greatest gain is observed on these curves in the central component of the five-mode radiation (see Fig. 4). The component without population inversion is amplified by energy transfer from other radiation components. The most typical is the gain of the central component at the expense of the sideband components (see Figs. 2 and 4).

## 5. CONCLUSION

Let us summarize our main results.

1. A Hill equation was derived for the probability amplitudes of a two-level system acted upon by multimode radiation with equidistant spectrum, and the characteristic exponents of this equation were determined. A connection is found between the characteristic exponents of Hill's equation and the "dressed-atom" quasienergy, an expression for which contains both the parameters of the system itself and the characteristics of the radiation.

The quasienergy level splitting is treated as a manifestation of the high-frequency Stark effect due to the action of the multimode radiation on the atomic system.

2. We have shown that transitions starting at the crossing point of the upper-state quasienergy levels and terminating at the lower-state quasilevel crossing points are resonant and cause maxima or minima to appear on the plots of the absorption coefficients of the monochromatic radiation of the components.

3. The density-matrix formalism was used to calculate the absorption coefficients of individual components of polychromatic radiation. The computer-calculated resonance positions on the curve were compared in detail with the resonance positions given by the quasienergy-level crossing. This comparison has shown good agreement between the two approaches to the problem on hand.

In conclusion, the authors thank V. I. Perel' for a helpful discussion of the results.

<sup>1</sup>Individual details of bichromate-component absorption processes are explained in theoretical papers.<sup>9–15</sup>

<sup>2</sup>The coefficient  $Q(t)$  becomes infinite at  $G(t) = 0$ . We shall therefore assume that  $G_l = |G_l| \exp(i\varphi_l)$  ( $l = 0, 1, 2, \dots$ ) are chosen to make  $G(t) \neq 0$  at any instant of time  $t$ . It will be shown in Sec. 4 that the solutions are continuous in the parameters  $G_l$  and in the detunings  $\Delta$ . We seek therefore the solutions of (2.4) under the condition  $G(t) \neq 0$  and extend it to include the set of instants  $t$  at which  $G(t_n) = 0$ .

<sup>3</sup>It is known that the characteristic exponents of the Hill equation are determined accurate to  $n\Delta$ , i.e., if  $\mu$  is a characteristic exponent of Hill's equation, then  $\mu \pm n\Delta$  is also a characteristic exponent of this equation for any integer  $n$ .

<sup>4</sup>The asymmetry of the absorption profile of a strong test wave in the presence of a strong perturbing wave shifted relative to the line center was described by us in Refs. 9 and 12.

<sup>5</sup>The problem of three-mode interaction was first solved Yankauskas<sup>25</sup> for an amplitude-modulated signal. A different approach to the solution of the problem was used later in Ref. 26. The polarization of a medium by a strongly modulated signal was investigated also in Refs. 27 and 28. Also noteworthy are Refs. 29 and 30, in which the action on a medium by strong multicomponent radiation with an equidistant spectrum was considered. In Ref. 29 was calculated the polarization of a medium by the action of multimode radiation in the case of large frequency spacing between the modes,  $\Delta > G_l$ . In Ref. 30 was considered the case when the

amplitude of one strong field was much larger than the amplitudes of several other strong fields.

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