Theory of subradiative absorption structure in the interaction between two intense waves in a nonlinear medium

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A theory is constructed of the nonlinear interaction between two electromagnetic fields having arbitrary intensities, in a medium with a homogeneously broadened absorption (amplification) line; the theory agrees well with experiment. Simple approximate expressions are obtained for the positions and widths of the mulitphoton resonances. The existence of a narrow absorption extremum near zero detuning of the field frequencies is predicted. A simple analytic dependence of the nonlinear absorption coefficient on the ratio of the field amplitude is obtained at equal field frequencies. This dependence agrees well with experiment. An analytic equation is derived for the nonlinear absorption on the line wings.

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§1. INTRODUCTION

The interaction of two waves in a nonlinear medium is one of the basic problems of the theory of multimode lasers and laser spectroscopy. This question was first considered by Rautian and Sobel'man,[1] who investigated theoretically the absorption of a weak test wave in the presence of a strong wave that perturbs a two-level system. It was shown that the absorption of the test wave depends in nonmonotonic fashion on the difference $\omega_1 - \omega_2 = \Omega$ between the wave frequencies.[1)] In the symmetrical situation, when the frequency ω_2 of the perturbing wave coincides with the center ω_0 of the absorption line, the maxima of the absorption of the test wave are located at $\Omega = \pm\Omega_R$, where Ω_R is the optical-nutation frequency (the Rabi frequency).

With increase of intensity of the test wave, a structure in its absorption spectrum was observed in Ref. 4 and called there subradiative structure. In addition to the principal maximum at $|\Omega| = \Omega_R$, new maxima appear at $|\Omega_n| = \Omega_R/n$ $(n = 2, 3, 4)$. These maxima are due to multiphoton transitions.[4]

Later experiments[5] have revealed an anomalously strong dependence of the absorption near $\Omega = 0$ on the ratio of the wave amplitudes. When the ratio $\rho = E_1/E_2$ of the test and perturbing wave amplitudes changed from 0.9 to 1.1., the absorption changed by several times. The theory developed in Ref. 4 does not explain this effect, since it is valid in two limiting cases, $\rho \ll 1$ and $\rho \gg 1$. The hopes expressed in Ref. 4 to be able to interpolate the formulas in such a way that they would be valid also at $\rho \approx 1$ were not realized.

In the present paper we develop a theory that describes the subradiative structure at comparable field intensities. We solve the equations for the density matrix of a two-level system in a field of two waves. The amplitudes of the modulation of the level populations at the frequency Ω and at its harmonics $n\Omega$ are obtained in the form of continued fractions that are expanded in series that converge rapidly even at comparable intensities. At $\Omega = 0$, the situation simplifies: both waves have the same frequency and simple analytic expressions are obtained for the density-matrix elements. In particular, the expression for the absorption coefficient of the test wave at $\Omega = 0$ is

$$K_1(0) = \frac{K_0}{2I_1}\left\{1 - \frac{1+I_2-I_1}{[1+2(I_1+I_2)+(I_1-I_2)^2]^{1/2}}\right., \quad (1)$$

where $K_0 = 4\pi d^2 N_0 \omega / \hbar\gamma_{ab}\varepsilon_0^{1/2} c$ is the linear absorption coefficient at the line center, I_1 and I_2 are the dimensionless wave intensities, $I_{1,2} = (dE_{1,2}/\hbar\gamma)^2$ (d is the transition matrix element, γ is the line width, and N_0 is the level-population difference and is determined only by the pumping and by the relaxation). As shown in §4, Eq. (1) describes well the experimental results.[2)]

The theory predicts, at close values of the wave amplitude, the appearance of a new singularity in the absorption structure near zero detuning. At $E_1 < E_2$ the singularity takes the form of a sharp peak with a width less than γ. Thus, at $E_1 = 0.8E_2$ the width of the peak is less than $\gamma/2$. At $E_1 > E_2$ a sharp dip appears. This singularity was observed in experiment.

Simple approximate equations were derived, describing the positions and widths of both the single-photon and multiphoton resonances. In particular, the position of the resonance is described as before by the expression for Ω_R/n, with the following simple approximate expression obtained for Ω_R:

$$\Omega_R = (d/\hbar)(E_1^2 + E_2^2)^{1/2}, \quad (2)$$

which is valid at arbitrary ratios of the wave amplitudes E_1 and E_2.

§2. SOLUTION OF THE DENSITY-MATRIX EQUATIONS IN A BICHROMATIC FIELD

To determine the absorption (amplification) coefficient and other characteristics of the response of a nonlinear medium to the action of a high-frequency electromagnetic field it is necessary to know the polarization (the average dipole moment) of the medium $P = d(\rho_{ab} + \rho_{ba})$. The elements of the density matrices ρ_{ab} and $\rho_{ba} = \rho_{ab}^*$ are obtained in the semiclassical ap-

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proximation from the solution of the following density-matrix equations:

$$\frac{\partial\rho_{ab}}{\partial t}+(i\omega_0+\gamma_{ab})\rho_{ab}=iV(t)(\rho_{aa}-\rho_{bb}),$$
$$\frac{\partial\rho_{aa}}{\partial t}+\gamma_a\rho_{aa}=\lambda_a+iV(t)(\rho_{ab}-\rho_{ba}), \tag{3a}$$
$$\frac{\partial\rho_{bb}}{\partial t}+\gamma_b\rho_{bb}=\lambda_b-iV(t)(\rho_{ab}-\rho_{ba}),$$

where

$$V(t)=-dE(t)/\hbar=-({}^1/{}_2G_1e^{i\omega_1t}+{}^1/{}_2G_2e^{i\omega_2t})+\text{c.c.},$$
$$G_i=(dE_i/\hbar)e^{-i\varphi_i}.$$

The difference between γ_{ab} and $(\gamma_a+\gamma_b)/2$ makes it possible to describe phenomenologically the dephasing collisions in the model of homogeneous absorption-line broadening; λ_a and λ_b represent the pumping of the atoms to the levels a and b per unit time.

We seek the stationary solutions for the elements of the atomic density matrix in a two-frequency radiation field. Just as the known stationary solution for a monochromatic field, these solutions will not depend on the initial conditions. We change over to the slow variable $\sigma=\rho_{ab}\exp(i\omega_2t)$. The system (3a) changes accordingly in the "rotating field" approximation into the following system of equations

$$\frac{\partial\sigma}{\partial t}+[i(\omega_0-\omega_2)+\gamma_{ab}]\sigma=-\frac{i}{2}(G_2^*+G_1^*e^{-i\Omega t})(\rho_{aa}-\rho_{bb}),$$
$$\frac{\partial\rho_{aa}}{\partial t}+\gamma_a\rho_{aa}=\lambda_a-\frac{i}{2}[(G_2+G_1e^{i\Omega t})\sigma-(G_2^*+G_1^*e^{-i\Omega t})\sigma^*], \tag{3b}$$
$$\frac{\partial\rho_{bb}}{\partial t}+\gamma_b\rho_{bb}=\lambda_b+\frac{i}{2}[(G_2+G_1e^{i\Omega t})\sigma-(G_2^*+G_1^*e^{-i\Omega t})\sigma^*].$$

In Ref. 4, a system of equations similar to (3b) was solved under the conditions $\omega_2=\omega_0$ and $\gamma_a=\gamma_b$ in the zeroth order in the small parameter $\Omega/|G_2|\ll 1$ at $\rho\ll 1$ or $\rho\gg 1$ $(\rho=|G_1|/|G_2|)$. In the present paper the system (3b) is solved without any additional approximations whatever.[3)]

We seek the solution of the system (3b) in series form:

$$\sigma=\sum_{n=-\infty}^{\infty}\sigma_ne^{-in\Omega t},\quad \rho_{aa}=\sum_{n=0}^{\infty}a_ne^{-in\Omega t}+\text{c.c.},\quad \rho_{bb}=\sum_{n=0}^{\infty}b_ne^{-in\Omega t}+\text{c.c.}. \tag{4}$$

Substitution of the series (4) in Eq. (3b) leads to an infinite system of linear algebraic equations:

$$[\gamma_{ab}+i(\omega_0-\omega_2-n\Omega)]\sigma_n=-\frac{i}{2}(G_2^*d_n+G_1^*d_{n-1}),\quad n\geqslant 1, \tag{5a}$$

$$[\gamma_{ab}+i(\omega_0-\omega_2+n\Omega)]\sigma_{-n}=-\frac{i}{2}(G_2^*d_n^*+G_1^*d_{n+1}^*),\quad n\geqslant 0, \tag{5b}$$

$$(\gamma_a-in\Omega)a_n=\lambda_a\delta_{n0}-\frac{1}{2}[G_2\sigma_n-G_2^*\sigma_{-n}^*+G_1\sigma_{n+1}-G_1^*\sigma_{-n+1}^*], \tag{5c}$$

$$(\gamma_b-in\Omega)b_n=\lambda_b\delta_{n0}+\frac{i}{2}[G_2\sigma_n-G_2^*\sigma_{-n}^*+G_1\sigma_{n+1}-G_1^*\sigma_{-n+1}^*], \tag{5d}$$

where $d_n=a_n-b_n$ $(n=1,2,3,\ldots)$; $d_0=d_0^*=2(a_0-b_0)$; δ_{n0} is the Kronecker symbol; at $n=0$ it is necessary to substitute in the left-hand sides of the equations $2a_0$ in place of a_n and $2b_0$ in place of b_n.

Substituting in (5c) and (5d) the expressions obtained from (5a) and (5b) for σ_n and σ_{-n}, we obtain a_n and b_n and their difference. We then obtain the following recurrence relation[4)]:

$$d_{n+1}G_2^*G_1B_{n+1}+d_nF_n+d_{n-1}G_2G_1^*B_n=\gamma(\lambda_a/\gamma_a-\lambda_b/\gamma_b)\delta_{n0}, \tag{6}$$

where

$$n\geqslant 0,\quad d_{-1}=d_1^*,\quad \gamma=2\gamma_a\gamma_b/(\gamma_a+\gamma_b),$$

$$F_n=\frac{2(\gamma_a-in\Omega)(\gamma_b-in\Omega)}{\gamma_a+\gamma_b-2in\Omega}+\sum_{p=1}^{2}\frac{|G_p|^2}{2}\left\{\frac{1}{\gamma_{ab}-i(\omega_p-\omega_0+n\Omega)}+\frac{1}{\gamma_{ab}+i(\omega_p-\omega_0-n\Omega)}\right\},$$
$$2B_n=\frac{1}{\gamma_{ab}+i(\omega_1-\omega_0-n\Omega)}+\frac{1}{\gamma_{ab}-i(\omega_2+\omega_0+n\Omega)}. \tag{7}$$

We note that $B_0=B_1^*$ and $F_0=F_0^*$.

To solve (6), we define the quantity

$$X_n=-\frac{F_n}{B_nG_1^*G_2}\frac{d_n}{d_{n-1}}. \tag{8}$$

From (6) we obtain

$$d_0=\frac{N_0\gamma}{F_0[1-2\mathrm{Re}(D_1X_1)]},\quad N_0=\frac{\lambda_a}{\gamma_a}-\frac{\lambda_b}{\gamma_b}, \tag{9}$$
$$X_n=(1-D_{n+1}X_{n+1})^{-1}. \tag{10}$$

The recurrence relations (10) are easily solved for the continuous fraction:

$$X_n=\cfrac{1}{1-\cfrac{D_{n+1}}{1-\cfrac{D_{n+2}}{1-\ldots}}},\quad D_m=\frac{|G_1|^2|G_2|^2B_m^2}{F_{m-1}F_m}, \tag{11}$$
$$m=1,2,3,\ldots.$$

According to Ref. 7, any continuous fraction can be converted into a series, which in our case takes the form

$$X_n=1-\frac{D_{n+1}}{Q_nQ_{n+1}}-\frac{D_{n+1}D_{n+2}}{Q_{n+1}Q_{n+2}}-\frac{D_{n+1}D_{n+2}D_{n+3}}{Q_{n+2}Q_{n+3}}-\ldots-\frac{D_{n+1}\ldots D_{n+m}}{Q_{n+m-1}Q_{n+m}}-\ldots, \tag{12}$$

where $n\geqslant 1$,

$$Q_{n-1}=Q_n=1,\quad Q_{n+1}=Q_n-D_{n+1}Q_{n-1}. \tag{13}$$

The convergence of the series (12) will be discussed in §3.

To complete the solution of the system of equations (3b), we determine σ_n and σ_{-n} from (5a), (5b), (8), (9) and (11), (12):

$$\sigma_n=-\frac{i}{2G_1}\frac{d_{n-1}(|G_1|^2-F_{n-1}D_nX_n/B_n)}{\gamma_{ab}+i(\omega_0-\omega_2-n\Omega)},\quad n\geqslant 1,$$
$$\sigma_{-n}=-\frac{i}{2G_2}\frac{d_n^*(|G_2|^2-F_n^*D_{n+1}^*X_{n+1}^*/B_{n+1}^*)}{\gamma_{ab}+i(\omega_0-\omega_2+n\Omega)},\quad n\geqslant 0. \tag{14}$$

Knowing σ_n, we can determine ρ_{ab}, and consequently also the polarization P of the medium:

$$P=d\sum_{n=-\infty}^{\infty}[\sigma_ne^{-i(\omega_2+n\Omega)t}+\sigma_n^*e^{i(\omega_2+n\Omega)t}]. \tag{15}$$

We have thus obtained the response of the medium not only at the frequencies of the perturbing and test wave $\omega_2(\sigma_0)$ and $\omega_1=\omega_2+\Omega(\sigma_1)$, but also at all frequencies of the combination tones $\omega_2+n\Omega$.

We note the following circumstances.

1. A spatial structure is significant for the optical fields. In the expression for the Hamiltonian $V(t)$, the

form of the field was not specified. This makes it possible to use Eqs. (14) and (15) to solve a number of problems involving interaction between fields and a medium with homogeneously broadened absorption line. If $|E_i|$ does not depend on the spatial coordinate z, and $\varphi_i = k_i z + \psi_i$ $(i=1,2)$, we are dealing with the field of two waves, traveling in the same direction in the case $|k_1 - k_2| \ll |k_1|$ and in opposite directions at $|k_1 + k_2| \ll |k_1|$. Another possibility is to assume that the phases φ_i are independent of z, and that the field amplitude are periodic functions of z: $|E_i| = E_{0i}|\sin k_i z|$; in this case we are dealing with a field of two standing waves.

2. The combination tones that occur at the frequencies $\omega_n = \omega_2 + n\Omega$ have wave vectors $\mathbf{k}_n = \mathbf{k}_2 + n(\mathbf{k}_1 - \mathbf{k}_2)$, where n is an integer, positive or negative [see (15)]. The amplitudes of these waves in a strong field are comparable in order of magnitude with the amplitudes E_1 and E_2 if the dispersion relation $|\mathbf{k}_n| = \omega_n \varepsilon_0^{1/2}/c$ is satisfied, where ε_0 is the dielectric constant of the medium without the contribution of the resonant transition. This equality is satisfied in the case of waves traveling in the same direction. Therefore when two such waves propagate in an extended nonlinear medium, the combination tones must be taken into account. Another situation arises when the waves propagate in opposite directions. The dispersion relation does not hold for them. An approximate estimate (see Ref. 8, p. 160) an approximate estimate yields the following definition of the ratio of the amplitude of the combination tone E_m to the amplitude of one of the fundamental waves E_j $(j=1,2)$:

$$\left|\frac{E_m}{E_j}\right| \approx \left|\frac{P_m}{4\pi P_j}\right| \frac{\alpha_j \lambda}{m(m+1)}, \tag{16}$$

where P_m and P_j are the polarizations of the medium for the combination and fundamental waves, α_j is the absorption coefficient of the j-th wave, and λ is the wavelength. Since usually $\alpha_j\lambda \ll 1$ we find even at $|P_m/P_j| \approx 1$ that $|E_m/E_j| \ll 1$. Consequently, in interaction of opposing waves the combination tones can be disregarded. Thus, the problem of propagation of two strong opposing waves in an extended nonlinear medium is closed and must be solved with the aid of the response, obtained in the present section, of the medium at the frequencies ω_1 and ω_2.

§3. MULTIPHOTON RESONANCES IN THE ABSORPTION COEFFICIENTS OF THE TEST AND PERTURBING WAVES

The absorption (amplification) coefficients of traveling waves are defined in terms of the imaginary part of the polarizability of the medium:

$$K_1 = -\frac{8\pi\omega d^2}{\varepsilon_0^{1/2} c\hbar} \operatorname{Im}\left(\frac{\sigma_1}{G_1^*}\right); \quad K_2 = -\frac{8\pi\omega d^2}{\varepsilon_0^{1/2} c\hbar} \operatorname{Im}\left(\frac{\sigma_0}{G_2^*}\right). \tag{17}$$

Substituting (7), (9) and (11) in (14), and substituting (14) in turn in (17), we obtain the following expressions for the absorption coefficients K_j:

$$K_j = \frac{K_0}{1-2\operatorname{Re}(D_1X_1)} \left\{ \frac{1}{1+f_j^2} \left[1 + \frac{I_1}{1+f_1^2} + \frac{I_2}{1+f_2^2} \right]^{-1} - \frac{1}{I_j} \left[\operatorname{Re}\left(\frac{D_1X_1}{1-if/2}\right) + (-1)^j f_s \operatorname{Im}\left(\frac{D_1X_1}{1-if/2}\right) \right] \right\} \quad (s,j=1,2;\ s\neq j), \tag{18}$$

$$I_j = d^2E_j/\hbar^2\gamma_{ab}\gamma, \quad f_j = (\omega_j - \omega_0)/\gamma_{ab}, \quad f = (\omega_1 - \omega_2)/\gamma_{ab}.$$

Analysis of Eq. (18) shows that it generalizes a large number of physical situations. At $E_1 = E_2 = 0$ we obtain the linear absorption coefficients, at $E_s = 0, E_j \neq 0$ $(s \neq j)$ we obtain the nonlinear absorption coefficient of the j-th wave, determined by the first term in the curly brackets of (18). Substituting $I_j = 0$ in (18) (in this case $X_1 = 1$), we obtain the coefficient of absorption of a weak wave in the presence of a strong one.[1,7]

The symmetry of the problem of wave interaction in a nonlinear medium is determined by the relations that follow from (18) between the absorption coefficients K_1 and K_2 $[K_j = K_j(f_1, f_2, I_1, I_2), j=1,2]$:

$$K_2(f_1, f_2, I_1, I_2) = K_1(-f_2, -f_1, I_2, I_1),$$
$$K_2(f_1, f_1, I_1, I_2) = K_1(f_1, f_1, I_2, I_1). \tag{19}$$

As expected, the absorption coefficients become equal at equal intensities $I_1 = I_2$ and at equal distances of the wave frequencies from the line center $|f_1| = |f_2|$. In experiment, the frequency of the perturbing wave ω_2 coincided with the absorption-line center $(f_2 = 0, f_1 = f)$, and the frequency of the test wave was scanned between the principal maxima. In this case the spectra of K_1 and K_2 are symmetrical about the line center, and can be represented after a number of transformations in the form

$$K_1 = \frac{K_0}{4+f^2} \left\{ \frac{2}{I_1} + \left[\frac{4+f^2}{I_1 + (1+f^2)(1+I_2)} + \frac{2}{I_1}(f \operatorname{Im}(D_1X_1) - 1) \right] (1 - 2\operatorname{Re}(D_1X_1))^{-1} \right\}, \tag{20}$$

$$K_2 = \frac{K_0}{4+f^2} \left\{ \frac{2+f^2}{I_2} + \left[\frac{4+f^2}{1+I_2+I_1/(1+f^2)} - \frac{2}{I_2}\left(f \operatorname{Im}(D_1X_1) + 1 + \frac{f^2}{2} \right) \right] (1 - 2\operatorname{Re}(D_1X_1))^{-1} \right\}.$$

The quantities D_1 and X_1 are determined by Eqs. (11) and (12). The extrema of the absorption coefficients K_1 and K_2 are determined by the extrema of the term $f\operatorname{Im}(D_1X_1)$. The maxima of D_1X_1 are determined, according to (13), by the maxima of D_n $(n=1,2,3,\ldots)$. It is easiest to obtain the maxima of D_n by determining the poles of D_n. According to (11) and (7), the poles of D_n are the roots of the equations $F_{n-1} = 0$ and $F_n = 0$, which take in the case $f_2 = 0, \gamma_a = \gamma_b = \gamma$ the form

$$[(\varkappa - inf)(1-inf) + I_2][1 - i(n+1)f][1 - i(n-1)f] + I_1(1-inf)^2 = 0$$
$$(n = 0, 1, 2, \ldots), \quad \varkappa = \gamma/\gamma_{ab}. \tag{21}$$

We denote the roots of Eq. (21) for a certain arbitrary n by $f_n^{(i)}$. The real part $\operatorname{Re} f_n^{(i)}$ of the root determines the position of the n-th extremum corresponding to the $2n-1$ quantum resonance.[4] The poles of D_1 are determined by the relations

$$n=0, \quad f_0^{(1,2)} = \pm i[1 + I_1/(\varkappa + I_2)], \tag{22}$$

$$n=1, \quad f^3 + if^2[(3+I_1)/2 + \varkappa] - f[(1+3\varkappa)/2 + I_1 + I_2] - i[\varkappa + I_1 + I_2]/2 = 0. \tag{23}$$

The roots of (23) at $I_1 \ll 3$ are given by

$$f_1^{(1,2)} = \pm[I_1 + I_2 - (1-\varkappa)^2/4]^{1/2} - i(1+\varkappa)/2, \quad f_1^{(3)} = -i/2. \tag{24}$$

At $\varkappa = 1$ and $I_1 \to 0$ we have $\mathrm{Re} f_1^{(1,2)} = \pm I_2^{1/2}$, and the position of the principal maxima is determined by the Rabi frequency $\Omega_{1,2} = \pm G_2$. Thus, the roots $f_1^{(1,2)}$ determine the evolution of the position and the widths of the symmetrical single-photon resonances with increasing intensity of the test field, and in the case $\varkappa = 1$ Eq. (24) (as follows from calculations by Eq. (20) and from the experiments (see Figs. 1–3) determines fairly well the position of the single-photon resonances also at $I_1 \gtrsim 3$.

For $n \geqslant 2$, we solve Eq. (21) approximately. At $n \gg 1$, Eq. (21) takes the form

$$[(\varkappa - inf)(1-inf)+I_1+I_2](1-inf)^2=0. \qquad (25)$$

The roots of this equation are

$$f_n^{(1,2)} = \pm\frac{1}{n}\left[I_1+I_2-\left(\frac{1-\varkappa}{2}\right)^2\right]^{1/2} - i\frac{1+\varkappa}{2n}, \quad f_n^{(3,4)} = -\frac{i}{n}. \qquad (26)$$

The roots $f_n^{(1,2)}$ determine the positions and widths of the $2n-1$ photon resonances. The values of $\mathrm{Re} f_n^{(1)}$ at $\varkappa = 1$ are compared with the experimental values and with those calculated from Eq. (20) in Table I.[5] From the table and from the comparison of $f_n^{(1,2)}$ with the curves of Fig. 1 and 2 it is seen that the positions of all the resonances are described, with an error not larger than 10%, by the simple formula Ω_R/n, where Ω_R is given by Eq. (2) in the case $\gamma = \gamma_{ab}$.

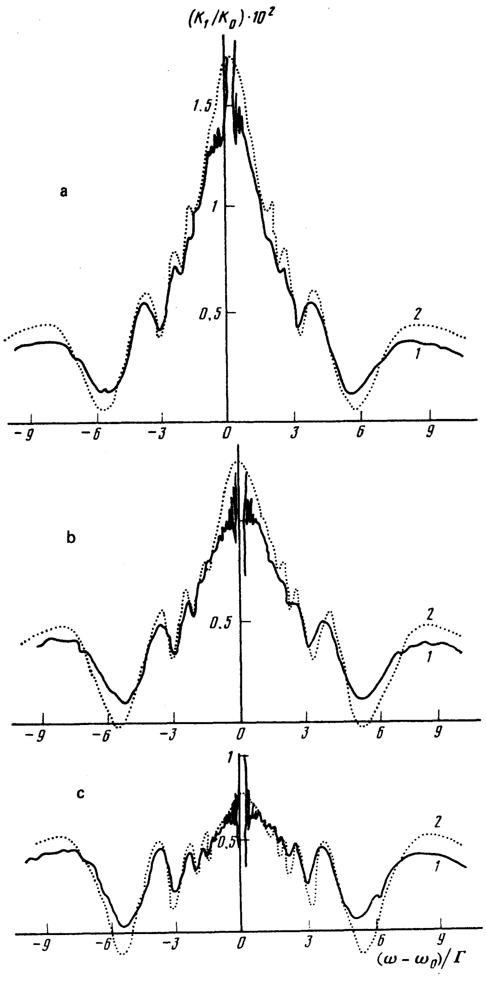

FIG. 1. Nonlinear absorption coefficient K_1/K_0 at $I_2 = 34.81$. a) $\rho = 1.1$; b) $\rho = 1$; c) $\rho = 0.9$; 1) experimental curve, 2) theoretical curve calculated from Eq. (20).

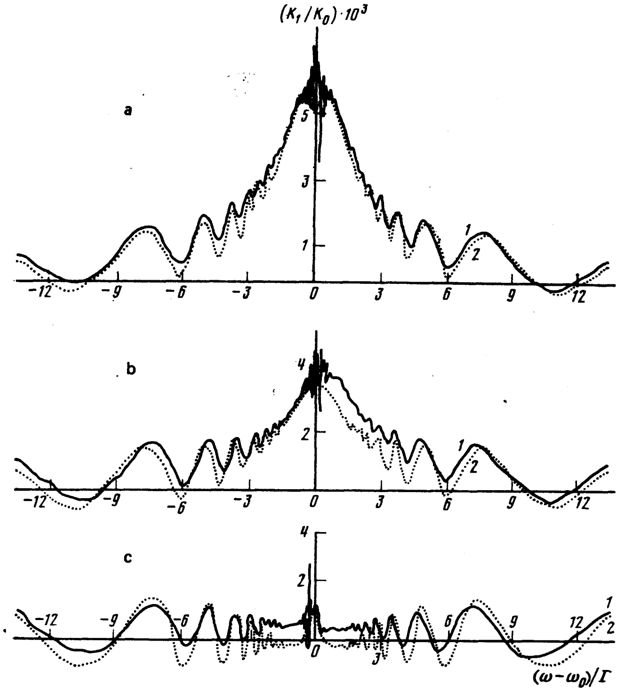

FIG. 2. The same as Fig. 1 for $I_2 = 141.61$.

An interesting question is that of the number of steps of the continued fraction F_1 [see (11)] or, equivalently, the number of terms (12) for X_1 which is sufficient to calculate the absorption coefficient (20) with the required accuracy. It is necessary that all n^* allowed resonances appear on the calculated absorption curves. Since each n-th resonance corresponds to a pole of D_n, which appears for the first time in the n-th term of the series (12), the minimum number of terms in the series is n^*, where n^* is determined by equation (27). If we take, with a certain margin, n^*+1 terms of the series (12), then it appears that this number

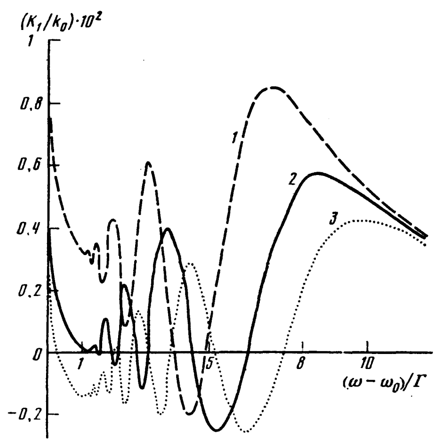

FIG. 3. Nonlinear absorption coefficient K_1/K_0 at $\rho = 0.8$, calculated from Eq. (20). 1) $I_2 = 23.6$; 2) $I_2 = 34.81$; 3) $I_2 = 47.6$.

TABLE I. Position of multiphoton maxima of the absorption coefficient for the case $I_2 = 141.61$, $\rho = 0.8$, $\varkappa = 1$, n is the number of the maximum, f_T is the position of the maximum of K_1 calculated with a computer using Eq. (20), f_F—value obtained from (26), f_E—value obtained in experiment.[9]

f_n	n					
	2	3	4	5	6	7
f_T	7.5	4.9	3.6	2.9	2.4	2.0
f_F	7.6	5.0	3.8	3.0	2.5	2.2
f_E	7.7	5.0	3.8	3.0	2.6	2.3

suffices to describe the behavior of the coefficients K_1 and K_2 in the entire region in which they are defined. Indeed, all the $D(0)$ are equal at $f=0$:

$$D=\frac{I_1I_2}{(1+I_1+I_2)^2}<\left(\frac{1}{2}\right)^2,$$

and accordingly the n-th term of the series D_1X_1 at $f=0$ is of the order of $D^n < 4^{-n}$. At $f^2 \gg (I_1+I_2)/n$ we have

$$|D_n| \approx \gamma^2 I_1 I_2/[n(\omega-\omega_0)]^2$$

and the n-th term of the series D_1X_1 is at $f^2 \gg I_1+I_2$ of the order of

$$(\gamma^2 I_1 I_2)^n/(n!(\omega-\omega_0)^n)^2.$$

Thus, the series D_1X_1 decreases rapidly at small $f^2 \ll 1$, just as at large detunings $f^2 \gg I_1+I_2$, and if the series has n^*+1 terms it describes all the multiphoton resonances.

The absorption-line wings $f^2 \gtrsim I_1+I_2$ can be described by Eqs. (20) with $X_1=1$. The imaginary and real parts of the expression for D_1 take the form

$$\mathrm{Re}\,D_1=AC/B, \quad \mathrm{Im}\,D_1=fAF/B, \tag{28}$$

where

$$A=I_1I_2/[I_1+(1+f^2)(1+I_2)],$$
$$B=[1-f^2(2+3/\varkappa)+I_1(1-f^2)+I_2]^2+f^2[3+1/\varkappa-(2/\varkappa)f^2+2(I_1+I_2)]^2,$$
$$C=1+(^{19}/_4-1/\varkappa)f^2+(^{13}/_4-9/2\varkappa)f^4+(1-2/\varkappa)f^6$$
$$+I_1[1+^{15}/_4f^2+^{9}/_4f^4+^{1}/_2f^6]+I_2[1+^{19}/_4f^2+3f^4],$$
$$F=1+1/\varkappa+(^{9}/_2+19/4\varkappa)f^2+(2+13/4\varkappa)f^4+f^6/\varkappa$$
$$+[(7+3f^2)I_1-(1+4f^2)I_2]f^2/4.$$

At intensities $I_1+I_2 \leqslant 1$ [in this case, according to the estimate (27), there is only one-photon resonance], calculations by means of Eqs. (20), using (28) at $X_1=1$, determine the absorption coefficients at any detuning away from the line center.

§4. COMPARISON OF THEORY WITH EXPERIMENT

To compare the theory with experiment, computer calculations were made, using Eq. (20), of the absorption coefficient of the test wave K_1 for the following cases: ratio of the amplitudes of the test and perturbing waves $\rho=0.9$, 1.0, and 1.1 at perturbing-wave intensities $I_2=34.81$ and 141.61 (Figs. 1 and 2). These figures show also the experimental curves in addition to the caculated ones.[9] A comparison of these curves shows that the theory and experiment are in good agreement. The best agreement between theory and experiment takes place where the absorption coefficient $|K_1|$ is relatively large ($\rho=1.1$), and the worst agreement when $|K_1|$ is small ($\rho=0.9$). The apparent reason is that the absolute experimental errors are equal in both cases. This suggests that the discrepancies between theory and experiment lie within the limits of the experimental error.

In addition to multiphoton maxima, calculations show that at the line center ($f=0$) (see Figs. 2 and 3) there appears a narrow extremum, namely a maximum at $\rho<1$ and a minimum at $\rho>1$ [Fig. 2(c)], with a shape that is sharply peaked and different from a Lorentz curve. It follows from (24) and (26) that each of the D_n has a pole at $f=0$. Thus, near $f=0$ an interference of sorts takes place between the resonances. The width of produced extremum depends on the ratio ρ of the wave amplitudes. At $\rho=0.8$ the half-width of the maximum is $\Delta f \approx 1/2-1/4$ (see Fig. 3).

§5. ANOMALOUS CHANGE OF THE ABSORPTION COEFFICIENT WITH CHANGE OF THE WAVE-AMPLITUDE RATIO

Experiment[5] has shown that when the ratio of the wave amplitudes near unit changes by ±10%, $K_1(f)$ changes by several times near $f=0$. K_1 changes by a factor of two at $I_2=34.81$, and by 5–6 times at $I_2=141.6$. It can be shown that the anomalously large change of $K_{1,2}(0)$ follows from Eqs. (20). As $f\to 0$, the values of D_n determined by Eq. (11) become equal to each other and are given by

$$D_n \to D = I_1I_2/(1+I_1+I_2)^2.$$

The quantity D_1X_1 takes the form of a continued fraction

$$D_1X_1=\cfrac{D}{1-\cfrac{D}{1-\cfrac{D}{1-\dots}}}$$

which contracts into the expression $[1-(1-4D)^{1/2}]/2$. Accordingly, the expressions for $K_1(0)$ and $K_2(0)$ take the form[6]

$$K_{1,2}=\frac{K_0}{2I_{1,2}}\left\{1-\frac{1\pm(I_2-I_1)}{[1+2(I_1+I_2)+(I_1-I_2)^2]^{1/2}}\right\}. \tag{29}$$

Calculations in accordance with Eq. (29), in full agreement with experiment,[5] account for the rapid character of the variation of the absorption coefficients $K_1(0)$ and $K_2(0)$ when the ratio ρ of the amplitude varies near $\rho=1$. These changes of K_1 and K_2 have opposite tendencies. In accord with (19) we have $K_2(0,0,I_1,I_2)=K_1(0,0,I_2,I_1)$. When ρ decreases from unity, K_1 decreases and K_2 increases, and vice versa when ρ increases from unity. Thus, for $I_2^{1/2}=5.9$ we have the ratio

$$\frac{K_1(0,\rho=1.1)}{K_1(0,\rho=0.9)}=\frac{K_2(0,\rho=0.9)}{K_2(0,\rho=1.1)}=2.3,$$

in experiment[5,6] this value amounted to approximately 2. The anologous ratio for $I_2^{1/2}=11.9$ is 5.6; the experimental value was 5–6. A refinement of the experimental value encounters difficulties involved with the accurate determination of the absorption coefficients of one field at a frequency that coincides with or is close to the frequency of another field.

An approximate estimate of the relative change of the absorption coefficients at zero when ρ are close to unity ($|1-\rho^2| \ll 1$) yields

$$\frac{K_{1,2}(0,\rho)-K_{1,2}(0,1)}{K_{1,2}(0,1)}=\pm\frac{\rho^2-1}{\rho^2+1}\left\{\frac{I_2(1+\rho^2)}{[1+2I_2(1+\rho^2)]^{1/2}-1}-1\right\}. \quad (30)$$

As seen from (30), the value of this change depends strongly on the value of the total intensity of the waves $I_2(1+\rho^2)$, the coefficient in (30) increases rapidly with increasing intensity of the perturbing wave at a specified amplitude ratio ρ.

We note certain features of the absorption coefficient K_1 of the test field at $\Omega=0$. When the conditions $I_1=I_2 \ll 1$ are satisfied we have

$$K_1(0)=K_0(1-I_1-2I_2). \quad (31)$$

Equation (31) demonstrates the possibility of measuring the saturation coefficient by determining the slope of the straight line in the plot of $k_1(0)/K_0$ against $|E_1|^2$ or $|E_2|^2$. At $I_1, I_2 \gg 1$ the absorption coefficient $K_1(0)$, regarded as a function of I_1 at a specified I_2, has a maximum if the following relation holds:

$$I_{1max}=I_2+2(I_2)^{2/3}+{}^2/_3(2I_2)^{1/3}+O(1),$$
$$K_{1max}(0, I_{1max}, I_2)=(K_0/I_2)[1-{}^5/_2(2I_2)^{-1/3}]. \quad (32)$$

In conclusion, we point out the analogy between the density-matrix equations (3b) and the Bloch equations in the theory of magnetic resonance. The system (3b) for "slow" matrix elements under the condition that the relaxation constants are equal $\gamma_a=\gamma_b=\gamma_{ab}=\gamma$ goes over into the Bloch vector equation with account taken of the relaxation and of the constant magnetic field:

$$\left(\frac{d}{dt}+\gamma\right)\mathbf{M}=\gamma_0[\mathbf{M}\times(\mathbf{H}_0+\mathbf{H}_1)]+\gamma\mathbf{M}_0. \quad (33)$$

The following relations hold between the variables and the parameters of the system (3b), on the one hand, and the components of the magnetic moment $\mathbf{M}$:

$$M_x=\sigma+\sigma^*, \quad M_y=i(\sigma-\sigma^*), \quad M_z=\rho_{aa}-\rho_{bb}, \quad (34a)$$

between the components of the effective constant magnetic field $\mathbf{H}_0$:

$$\gamma_0 H_{0x}=G_2, \quad \gamma_0 H_{0y}=0, \quad \gamma_0 H_{0z}=\omega_2-\omega_0, \quad (34b)$$

between the components of the effective circularly polarized alternating magnetic field $\mathbf{H}_1$ at the frequency $\Omega=\omega_1-\omega_2$:

$$\gamma_0 H_{1x}=G_1\cos(\Omega t+\varphi), \quad \gamma_0 H_{1y}=G_1\sin(\Omega t+\varphi), \quad H_{1z}=0, \quad (34)$$

and also between the projections of the constant magnetic moment $\mathbf{M}_0$:

$$M_{0x}=M_{0y}=0, \quad M_{0z}=(\lambda_a-\lambda_b)/\gamma.$$

It is important that in our case the fields $\mathbf{H}_0$ and $\mathbf{H}_1$ are not perpendicular, and to our knowledge there are no published solutions of the Bloch equation (33) for such a case. The expressions obtained in the present paper can be used to describe magnetic resonance with fields (34b) and (34c).

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1) In particular, in a certain range of the detunings Ω the absorption gives way to amplification. This effect was observed in Refs. 2 and 3.

2) The experiments were performed in the radio-frequency band on the Zeeman transition of the ground state 5^1S_0 of Cd^{113} atoms.

3) In the experiment of Ref. 4, which was carried out in the radio band, there is only one relaxation constant γ, but it would be very interesting to carry out a similar experiment in the optical band, where $\gamma_a \neq \gamma_b \neq \gamma_{ab}$. Having in mind the performance of such an experiment, we shall solve Eqs. (3) retaining the difference between the relaxation constants γ_a, γ_b, and γ_{ab}, all the more since this complicates the solution only insignificantly.

4) The recurrence relations (6) and (7) were obtained earlier[6] for the case of equal level widths $\gamma_a=\gamma_b$.

5) Multiphoton resonances condense as $f\to 0$. The number of resolved maxima can be obtained from the resolution condition: the distance between the centers of neighboring maxima is larger than the sum of their half-widths:

$$(I_1+I_2)^{1/2}/(n-1)-(I_1+I_2)^{1/2}/n \geq 1/n+1/(n-1).$$

From this we get for the number of resolved maxima n^*

$$n^*=[(I_1+I_2)^{1/2}+1]/2. \quad (27)$$

The dependence of n^* on the summary intensity of the waves describes the experimental situation[4,9] with sufficient accuracy.

6) Expression (29) follows also from the equations of Ref. 6, which were derived by another method. We note that the fields retain their individuality also when the frequencies are equal. Accordingly, K_1(29) coincides with the nonlinear absorption coefficient $K_0/(1+I_1)$ of a monochromatic field only if $I_2=0$.

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