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POLARIZATION EFFECTS IN NONLINEAR SPECTROSCOPY

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The passage of a weak monochromatic wave through a resonant gaseous medium located in a strong monochromatic field of a close frequency is considered. Level degeneracy and the tensor character of the collision term are taken into account. Amplification of the weak wave is determined as a function of the weak and strong wave polarization directions. A very distinct structure appearing on the gain line of the weak wave depends on relaxation processes in the medium and manifests itself differently for mutually parallel and mutually orthogonal wave polarizations. The possibility is discussed of employing the detected polarization effects to investigate collisions in a gas and to determine the level widths of the gas molecules.

MANY recent experimental and theoretical papers have reported investigations of interactions between monochromatic waves and quantum resonance systems.[1-4] In addition, the optical properties of atoms acted upon by several coherent waves have been investigated.[5,6] Such studies in nonlinear spectroscopy are very promising for investigations of laser radiation properties and for determining level widths and other properties of gas atoms and molecules.[1)]

In[4] an attempt was made to observe directly the Bennett "holes"[7,8] in the gain line of a He-Ne laser operating with the $3s_2 - 3p_4$ transition ($\lambda = 3.39\ \mu$) of atomic neon. The holes burnt by the strong field of the given laser were investigated by sending through the working resonator a weak wave from an identical laser and by scanning the frequency of the latter within the limits of the spectral line width. The recorded gain line was basically of Lorentzian shape on which there appeared a narrow dip, without mirror reflection, in the region where the frequencies of the weak and strong fields coincide. Therefore the observed narrow dip cannot be identified with one of the Bennett holes.

To account for the aforementioned effect we must calculate the amplification of a weak electromagnetic wave with a frequency ω_1 traveling in an active gaseous medium that is located in a strong resonant field with a frequency ω. In the absence of level degeneracy the gain of the weak signal was determined in[6] by calculating the probabilities that the weak field would be emitted or absorbed by an individual atom located in the strong field. This procedure enables one to determine the local value of the gain near the edge of the active medium. If the saturation parameter is sufficiently small, the amplification of the weak signal along the entire length of the laser can be characterized by the single value of the gain obtained in[6]. However, the strong laser field and its length are usually so large that the concept of a "gain" factor does not exist for the weak wave. This situation is associated with the fact that in the presence of a strong field an incoming weak wave of frequency ω_1 is divided into two waves of the same frequency ω_1 but with different dispersion laws and different gains. Therefore the amplification of the weak signal of frequency ω_1 is the resultant of the amplifications of the two waves.

Kuznetsov and Rautian[9,10] investigated the polarization of a medium acted upon by a strong and weak wave having parallel polarizations in the case of active atoms at rest and without degenerate levels; they observed a wave having the combination frequency $\omega_2 = 2\omega - \omega_1$. The appearance of combination frequencies (combination tones) from the interaction of two monochromatic waves in a resonant medium has also been reported in[11-14]. In the case of homogeneous broadening the formation of a narrow dip in the gain line of a weak signal can be accounted for with the aid of the dielectric susceptibility obtained in[9,10]. However, by following the same procedure we would lose other interesting effects, since the authors of[9,10] neglected atomic motion and level degeneracy, while taking only incomplete account of atomic collisions.

We shall determine the gain of a weak wave in the presence of a strong wave for both homogeneous and inhomogeneous broadening without neglecting either atomic motion or level degeneracy. The inclusion of degeneracy will permit a correct investigation of interacting resonance waves with different polarizations. It was found that the amplification of the weak wave under certain conditions depends largely on the mutual orientation of the weak- and strong-wave polarizations. At the center of the Bennett hole of an inhomogeneously broadened line a very narrow dip is formed, the width of which varies with the character of the wave polarizations. The depth of this narrow dip is sensitive to the nature of the atomic collisions and sometimes exceeds the depth of Bennett holes. When homogeneous broadening predominates, a narrow dip appears on the Lorentzian contour of the gain line because of the combining of the weak waves of frequency ω_1 and the combination frequency $2\omega - \omega_1$. Therein lies the difference from[14], where the interaction of two waves having an identical intensity and close frequencies ω and ω_1 was considered. In[14], because of mathematical difficulties,

[1)]The given references contain additional references to earlier investigations.

all waves with the combination frequencies $2\omega - \omega_1$, $2\omega_1 - \omega, \ldots$ were dropped and atomic collisions were completely ignored.

When elastic collisions induced by van der Waals and short-range interactions are unimportant, than in an inverted medium with $\gamma_1 \gg \gamma_2$ the profile of the narrow dip in the gain line of the weak signal does not depend on the polarizations of the weak and strong waves (γ_1 and γ_2 are the widths of the lower and upper levels). On the other hand, when the broadening of a degenerate level results mainly from elastic collisions the narrow dip is appreciably shortened in the case of parallel polarizations and spreads for orthogonal polarizations. The mentioned characteristics of the gain line were determined for an atomic resonance transition with $1 \to 0$ change of the total angular momentum; the zero angular momentum pertains to the lower level. We also note that the depth of Bennett holes in the gain line of a weak wave with orthogonal polarization depends strongly on the elastic collisions that induce transitions between sublevels of the degenerate level.

A very remarkable result is obtained when weak and strong waves simultaneously traverse an absorptive resonance medium with $\gamma_1 \ll \gamma_2$. For the $1 \to 0$ atomic transition in the absence of elastic collisions, near exact $\omega_1 = \omega$ resonance on the absorption curve of the weak wave there appears a narrow peak with width equal to γ_1 for parallel polarizations and γ_2 for orthogonal polarizations of the traveling waves. If elastic collisions are important here, then for parallel polarizations the narrow peak has width γ_1, as previously, but disappears for orthogonal polarizations.

The observed dependence of the weak-wave gain on the mutual orientations of the polarization vectors of the weak and strong waves is a new effect in nonlinear spectroscopy that can be utilized as an additional means of investigating the nature of atomic collisions and for determining the widths of gas atom levels. Similar nonlinear polarization effects have appeared in other problems[1,3,5] that involved strong and weak waves of different polarizations.

1. BASIC EQUATIONS

In order to understand physically why the gain depends on the wave polarizations, let us consider a weak wave of frequency ω_1 traveling through a resonator operating in a steady-state single mode at a close frequency ω. The results are easily extended to another case where the steady-state strong wave is replaced by a strong direct traveling wave that is either amplified or absorbed. Specifically, let us investigate the simplest atomic resonance transition with $1 \to 0$ change of the total angular momentum. The matrix structure of the collision term, with allowance for degenerate levels, has been determined for this transition.[15-18] We can therefore solve our problem for different polarization directions of the weak and strong waves.

The basic equations in the resonance approximation can be written as follows[2]:

$$\left(\frac{\partial^2}{\partial t^2} - c^2\frac{\partial^2}{\partial z^2}\right)\mathbf{A} = 4\pi c\int dv\,\mathbf{J}, \tag{1}$$

$$\left[i\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z} + \frac{\Gamma}{2}\right) - \omega_0\right]J_\alpha = \frac{3}{4}\gamma c\lambda\!\!\!^{-}(\rho_{\alpha\beta} - \rho_1\delta_{\alpha\beta})A_\beta, \tag{2}$$

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right)\rho_{\alpha\beta} = \frac{n}{3}W_2 f\delta_{\alpha\beta} - \gamma_2\rho_{\alpha\beta} - \delta S_{\alpha\beta} - \frac{i}{c}(J_\alpha A_\beta^* - A_\alpha J_\beta^*), \tag{3}$$

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right)\rho_1 = W_1 nf - \gamma_1\rho_1 + \gamma\rho_{\sigma\sigma} + \frac{i}{c}(\mathbf{J}\mathbf{A}^* - \mathbf{A}\mathbf{J}^*), \tag{4}$$

where

$$J_\alpha = -i\omega_0 d_{0\mu}{}^\alpha R_{\mu 0}, \quad \rho_{\alpha\beta} = 3d_{0\mu}{}^\alpha\rho_{\mu\mu'}d_{\mu'0}^\beta/|d_0{}^1|^2,$$

$$S_{\alpha\beta} = P\rho_{\sigma\sigma}\delta_{\alpha\beta} + Q\rho_{\alpha\beta} + G\rho_{\beta\alpha},$$

$$\gamma = 4|d_0{}^1|^2/9\lambda\!\!\!^{-}{}^3, \ \Gamma = \gamma_1 + \gamma_2 + \delta, \ \lambda\!\!\!^{-} = c/\omega_0.$$

Here $\rho_{\mu\mu'}$ and ρ_1 are the density matrix elements of the degenerate upper level and the lower level, respectively; $R_{\mu 0}$ is the density matrix describing transitions between upper and lower levels, with $d_{\mu 0}^\alpha$ and d_0' representing the dipole and reduced dipole moments of these transitions; W_1 and W_2 are excitation probabilities, per unit time, of the lower and upper levels as a result of pumping; γ is the probability that a quantum $\hbar\omega_0$ will be emitted spontaneously in unit time from an isolated atom; γ_1 and γ_2 are the widths of the lower and upper levels resulting from radiative processes and gas-kinetic inelastic collisions; δ is the width of the degenerate upper level as a result of van der Waals and short-range elastic collisions.

The ground-state density of the active atoms is n, and their velocity (v) distribution is described by the Maxwellian function

$$f = (1/u\sqrt{\pi})\exp(-v^2/u^2),$$

where $u = (2T/M)^{1/2}$ is the thermal velocity, T is the temperature, and M is the mass of an active atom. The quantities $\rho_{\alpha\beta}, \rho_1$, and J pertain to a group of atoms moving with velocity v, which is taken into account by $v\partial/\partial z$.

With regard to the collision term in (3) we must state the following.[16] Elastic collisions of the atoms produce mainly two effects: 1) the relaxation of atomic polarization, manifested in transitions between sublevels; 2) the relaxation of atomic velocities. The first of these effects is stronger, since it is proportional to the total collision cross section. The second effect involves, in addition, the mean scattering angle and is somewhat weaker. Accordingly, the collision term is divided, in the general case, into the sum of two components, $\delta S_{\alpha\beta}$ and an integral term. The relaxation of atomic polarization, described by $\delta S_{\alpha\beta}$, leads to several observed effects. For example, the relaxation of atomic polarization affects the power of the radiation at the central laser frequency, although the shape of the Lamb hole remains unchanged. The polarization of the traveling waves and of the laser radiation are affected most strongly by the aforementioned relaxation. Since we are here investigating polarization effects, the relaxation of sublevel transitions is taken into account fully in (3) by means of $\delta S_{\alpha\beta}$. The numerical coefficients of $\delta S_{\alpha\beta}$ for the special case of quasiresonant van der Waals collisions were calculated in[15] and in the general case satisfy the relations

$$3P + Q + G = 0,$$

$$\delta > 0, \quad P < 0, \quad |G| < Q.$$

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[2]Summations will be understood wherever vector or tensor indices are repeated. In our system of units $\hbar = 1$ and c is the velocity of light in a vacuum.

The integral portion of the collision term takes into account the spatial relaxation of velocities. This term strongly affects the shape of the Lamb hole, although its contribution to polarization effects is small. For simplicity, therefore, the integral portion of the collision term has been omitted in (3).

Equations (1)–(4) will be solved approximately, retaining only linear terms in the weak field. Therefore the vector potential **A** and the current **J** are written conveniently as

$$\mathbf{A} = [\mathbf{A}_+ + \mathbf{a}_1(z)e^{-i\Omega t} + \mathbf{a}_2(z)e^{i\Omega t}]e^{i(kz-\omega t)} + \mathbf{A}_- e^{-i(kz+\omega t)},$$
$$\mathbf{J} = [\mathbf{J}_+ + \mathbf{j}_1(z)e^{-i\Omega t} + \mathbf{j}_2(z)e^{i\Omega t}]e^{i(kz-\omega t)} + \mathbf{J}_- e^{-i(kz+\omega t)},$$
$$\Omega = \omega_1 - \omega = \omega - \omega_2,$$

where the plus and minus indices designate the amplitudes of the direct and reflected strong waves of frequency ω; the indices 1 and 2 of the vector potential and the current designate the amplitudes of weak waves with frequencies ω_1 and $\omega_2 = 2\omega - \omega_1$ traveling along the laser axis. We have, similarly,

$$\rho_{\alpha\beta} = \rho_{\alpha\beta}^0 + r_{\alpha\beta}e^{-i\Omega t} + r_{\beta\alpha}^* e^{i\Omega t},$$
$$\rho_1 = \rho_1^0 + re^{-i\Omega t} + r^* e^{i\Omega t}.$$

For the strong field we take the exact solution of (1)–(4) describing the steady-state single-mode laser regime in the absence of weak waves. In deriving the strong field from (1)–(4) we must add the boundary conditions of wave reflections at both ends of the resonator or introduce equivalent linear losses inside the resonator.

After the values of A_+, $\rho_{\alpha\beta}^0$, and ρ_1^0 for the strong field are obtained, the travel of the weak waves is described by the solution of the following equations:

$$(\omega_1 - \omega_k + icp)\mathbf{a}_1 = -2\pi\lambda\int dv\, \mathbf{j}_1 + ic\mathbf{a}_1(0), \quad (5)$$
$$(\omega_1 - 2\Omega - \omega_k - icp)\mathbf{a}_2^* = -2\pi\lambda\int dv\mathbf{j}_2^*, \quad (6)$$
$$\left(\omega_1 - \omega_0 - kv + \frac{i\Gamma}{2}\right)j_{1\alpha} = \frac{3}{4}\gamma c\lambda[(\rho_{\alpha\beta}^0 - \rho_1^0\delta_{\alpha\beta})a_{1\beta} + (r_{\alpha\beta} - r\delta_{\alpha\beta})A_{+\beta}], \quad (7)$$
$$\left(\omega_1 - 2\Omega - \omega_0 - kv - \frac{i\Gamma}{2}\right)j_{2\alpha}^* = \frac{3}{4}\gamma c\lambda[(\rho_{\beta\alpha}^0 - \rho_1^0\delta_{\alpha\beta})a_{2\beta}^* + (r_{\beta\alpha} - r\delta_{\alpha\beta})A_{+\beta}^*],$$
$$(\gamma_2 + \delta Q - i\Omega)r_{\beta\alpha} + \delta(Pr_{\sigma\sigma}\delta_{\alpha\beta} + Gr_{\alpha\beta}) = \quad (8)$$
$$= \frac{i}{c}(J_{+\alpha}^* a_{1\beta} + j_{2\alpha}^* A_{+\beta} - A_{+\alpha}^* j_{1\beta} - a_{2\alpha}^* J_{+\beta}), \quad (9)$$
$$(\gamma_1 - i\Omega)r = \gamma r_{\sigma\sigma} - \frac{i}{c}(\mathbf{J}_+^*\mathbf{a}_1 + \mathbf{j}_2^*\mathbf{A}_+ - \mathbf{A}_+^*\mathbf{j}_1 - \mathbf{a}_2^*\mathbf{J}_+), \quad (10)$$

where $\omega_k = kc$, $a_1(0)$ is the value of the amplitude $a_1(z)$ at the point $z = 0$ where the weak wave enters the medium, and we perform a Laplace transformation with respect to the variable z:

$$\partial a_1(z) / \partial z \to pa_1 - a_1(0).$$

In (7)–(10) we neglected pv as compared with γ_1, γ_2, and Γ; these quantities usually exceed the effective value of pv by several orders of magnitude.

The solution of (6)–(10) depends on the mutual orientation of the weak and strong wave polarizations. Therefore we shall consider the cases of parallel and orthogonal polarization separately, and shall begin by determining the strong field inside the laser.

2. THE STRONG FIELD INSIDE THE LASER

Since the specific forms of A_+ and A_- are unimportant in connection with polarization effects, we shall determine the strong field of a steady-state single-mode laser in the approximation where the rapid spatial modulation of the population inversion, $\exp(\pm i2kz)$, can be neglected.[6,19] From (1)–(4) we obtain

$$\rho_{\alpha\beta}^0 - \rho_1\delta_{\alpha\beta} = N\left[\delta_{\alpha\beta} + \frac{3F(\gamma_2\delta_{\alpha\beta} - \gamma l_\alpha l_\beta)}{\gamma_2 + \delta(Q+G)}\right], \quad (11)$$
$$F = \frac{\lambda A_0^2}{4h}\left(\frac{\Gamma}{(\Delta - kv)^2 + \Gamma^2/4} + \frac{\Gamma}{(\Delta + kv)^2 + \Gamma^2/4}\right), \quad (12)$$
$$N = N_0 / (1 + \Lambda_\parallel F), \quad (13)$$
$$\Lambda_\parallel = \frac{3\gamma}{\gamma_1}\left(1 - \frac{\gamma}{\gamma_2}\right) + \frac{\gamma}{\gamma_2}\left(1 + \frac{2\gamma_2}{\gamma_2 + \delta(Q+G)}\right), \quad (14)$$

where $A_0 = |A_+| = |A_-|$, **l** is the unit polarization vector of the strong field in the laser, $\Delta = \omega - \omega_0$ is the detuning of the strong field, and N_0 is the steady-state density of the population inversion:

$$N_0 = \left[\left(\frac{1}{3\gamma} - \frac{1}{\gamma_1}\right)\frac{\gamma W_2}{\gamma_2} - \frac{W_1}{\gamma_1}\right]n.$$

The relations (11)–(14) enable us to obtain the dielectric susceptibility $\varkappa$ for the direct wave:

$$\varkappa = \frac{3}{4}\gamma\lambda^3\int dv\frac{fN}{\Delta - kv + i\Gamma/2}. \quad (15)$$

Equating the imaginary part of (15) to the given losses, we obtain a transcendental equation for A_0.

3. THE CASE OF PARALLEL POLARIZATIONS

From (9) and (10) we obtain

$$(r_{\alpha\beta} - r\delta_{\alpha\beta})l_\alpha l_\beta = \frac{R}{3\gamma c}[A_0(\mathbf{j}_1 - \mathbf{j}_2^*) + \mathbf{a}_2^*\mathbf{J}_+ - \mathbf{J}_+^*\mathbf{a}_1], \quad (16)$$
$$R = \left(1 - \frac{3\gamma}{\gamma_1 - \gamma_2}\right)\frac{\gamma}{\Omega + i\gamma_2} + \left(1 + \frac{\gamma}{\gamma_1 - \gamma_2}\right)\frac{3\gamma}{\Omega + i\gamma_1} + \frac{2\gamma}{\Omega + i[\gamma_2 + \delta(Q+G)]}, \quad (17)$$

where **l** is the polarization vector of the weak and strong waves.

The insertion of (11) and (16) into (7) and (8) leads to the following expression for the Laplace transform a_1 of the weak wave (ω_1) amplitude:

$$a_1 = ia_1(0)\left[ip + \frac{\omega_1 - \omega_k}{c} - I_1 - \frac{I_3}{ip - (\omega_2 - \omega_k)/c - I_2}\right]^{-1}, \quad (18)$$
$$I_1 = \frac{3\pi}{2}\gamma\lambda^2\left[\int\frac{-Nf\,dv}{\Omega + \Delta - kv + i\Gamma/2} + \frac{\lambda A_0^2}{4}\int dv\frac{fNR}{D}\cdot \times\left(\frac{2\Omega + i\Gamma}{\Omega + \Delta - kv + i\Gamma/2} - \frac{\Omega}{\Delta - kv - i\Gamma/2}\right)\right],$$
$$I_3 = -\frac{3\pi}{8}\gamma\lambda^3 A_0^2 I_0\int dv\frac{fNR}{D}\frac{\Omega + i\Gamma}{\Delta - kv + i\Gamma/2},$$
$$I_0 = \frac{3\pi}{8}\gamma\lambda^3 A_0^2\int dv\frac{fNR}{D}\frac{\Omega + i\Gamma}{\Delta - kv - i\Gamma/2},$$
$$D = (\Delta - kv)^2 - (\Omega + i\Gamma/2)(\Omega + i\Gamma/2 - \lambda A_0^2 R/2),$$

where N is defined in (13), and I_2 is obtained from I_1 by means of the substitutions $\Delta \to -\Delta$ and $kv \to -kv$.

The poles of (18) determine the character of the weak-signal gain. The existence of two poles indicates that two weak waves of identical frequency ω_1 but with different dispersion laws are traveling in the medium; this was not noted in[6 9-14]. Similarly, two waves occur having the same combination frequency $\omega_2 = 2\omega - \omega_1$ but different dispersion laws; at the limiting point z = 0 they cancel each other. Thus a single incoming weak wave interacting with a strong wave will generate in the resonant medium three additional weak waves differing either in frequency or with regard to their dispersion laws. This fact is important in connection with the stability of the steady-state regime, since four frequencies of a weak fluctuating field will correspond to a given wavelength.

In accordance with (18), for $\gamma_2 \ll \gamma_1$ and moderate values of A_0 the weak-wave intensity as a function of ω_1 far from the strong resonance $\omega_1 = \omega$ represents the ordinary gain line with Bennett holes. This follows from the fact that as Ω increases the integral for I_1 retains only its first member, while I_3 and I_0 vanish. Therefore in the region $|\Omega| \gg \gamma_2$ the weak-signal gain is essentially of the usual type. However in the narrow region $|\Omega| \lesssim \gamma_2$ at the center of a Bennett hole a dip appears on the gain line.

The profile of the dip is especially distinct under the conditions

$$\lambdabar\gamma A_0^2 / \hbar\gamma_1\Gamma \ll 1, \quad \lambdabar\gamma A_0^2 / \hbar\gamma_2\Gamma \ll 1, \tag{19}$$

when an expansion with respect to the strong field A_0 is valid. In this region the intensity $I_{\omega_1}(z)$ of the weak field with frequency ω_1 for arbitrary detuning Δ of the strong field and $|\Omega| \ll \Gamma$ is given by

$$\begin{aligned}\frac{I_{\omega_1}(z)}{I_{\omega_1}(0)} = e^{Kz}\Big\{1 - \frac{3\pi}{2}\int dv \frac{fN_0\lambdabar^2 z\gamma\Gamma\Lambda_{\parallel}F}{(\Omega+\Delta-kv)^2+\Gamma^2/4} - \\ -\frac{3\pi}{2}\int dv \frac{fN_0\lambdabar^2 z\gamma\Gamma F_+}{(\Delta-kv)^2+\Gamma^2/4}\Big[\Big(1-\frac{3\gamma}{\gamma_1-\gamma_2}\Big)\Big(1-\frac{2\Omega(\Delta-kv)}{\gamma_2\Gamma}\Big)\frac{\gamma\gamma_2}{\Omega^2+\gamma_2^2} \\ +\Big(1+\frac{\gamma}{\gamma_1-\gamma_2}\Big)\Big(1-\frac{2\Omega(\Delta-kv)}{\gamma_1\Gamma}\Big)\frac{3\gamma\gamma_1}{\Omega^2+\gamma_1^2} \\ +\Big(1-\frac{2\Omega(\Delta-kv)}{\Gamma[\gamma_2+\delta(Q+G)]}\Big)\frac{2\gamma[\gamma_2+\delta(Q+G)]}{\Omega^2+[\gamma_2+\delta(Q+G)]^2}\Big]\Big\} \\ K = \frac{3\pi}{2}\int dv \frac{fN_0\lambdabar^2\gamma\Gamma}{(\Omega+\Delta-kv)^2+\Gamma^2/4} \qquad F = \frac{\lambdabar A_0^2}{4}\frac{\Gamma}{(\Delta-kv)^2+\Gamma^2/4}.\end{aligned} \tag{20}$$

For $ku \gg \Gamma$ the second term in the curly brackets of (20) describes Bennett holes in the ordinary gain line. The other terms represent a narrow dip at the center of the Bennett hole, with a width that is determined mainly by the smaller of the quantities γ_1 and γ_2. When the weak and strong direct waves traverse an absorptive or amplifying medium, then in (20) the substitution $F \to F_+$ is necessary for the case $A_0 \approx$ const.

As Δ changes, the center of the narrow dip is shifted together with the frequency of the strong field. Equation (20) shows that the narrow dip exhibits some asymmetry, which vanishes only for $\Delta = 0$. When the Doppler width greatly exceeds the collisional width ($ku \gg \Gamma$), the asymmetric term is smaller than the main term by the factor $\Gamma\Delta/(ku)^2$. In the opposite limiting case, $ku \ll \Gamma$, the corresponding ratio is $\Omega\Delta/\gamma_{1,2}\Gamma$ and can exceed unity.

To simplify (18) without loss of the given relationship, we investigate the weak-wave gain for zero detuning of the strong field ($\omega = \omega_0$). The weak-field amplitudes become

$$a_1(z) = {}^1/_2 a_1(0)\,(e^{iI_0z} + e^{-iI_0z})\,e^{i(\Omega/c - I_1)z}, \tag{21}$$

$$a_2^*(z) = {}^1/_2 a_1(0)\,(e^{iI_0z} - e^{-iI_0z})\,e^{i(\Omega/c - I_1)z}, \tag{22}$$

where in I_1 and I_0 we have inserted $\Delta = 0$ and A_0 is arbitrary.

Equations (21) and (22) show that the weak waves with frequencies ω_1 and ω_2 have identical intensities when $|\mathrm{Im}(I_0 z)| \equiv |I_0''|\, z > 1$. The combined intensity exhibits beats as a result of interference effects. After averaging over the beats the combined intensity $I(z)$ of the weak fields (21) and (22) becomes

$$I(z) = I(0)\ \mathrm{ch}\,(2I_0''z)\,e^{2I_1''z}, \tag{23}$$

where $I(0)$ is the intensity of the weak wave entering the medium.

The physical picture is further simplified for a homogeneously broadened line with $ku \ll \Gamma$:

$$I_1 = -2\pi\varkappa / \lambdabar - I_0.$$

Here κ is the dielectric susceptibility (15) with the replacement $\omega \to \omega_1$ in the denominator. In this case the vector potential $a_1(z, t)$ at the frequency ω_1 becomes

$$a_1(z,t) = \frac{1}{2}a_1(0)\,[1+\exp(i\cdot 2I_0z)]\exp\Big\{i\Big[\Big(k_1+\frac{2\pi\varkappa}{\lambdabar}\Big)z - \omega_1 t\Big]\Big\}. \tag{24}$$

The term unity in the square brackets corresponds to a weak wave at $\omega_1 = k_1 c$ traveling through a medium with the given dielectric susceptibility (15). The gain of this wave depends mainly on the level populations. The term $\exp(i\cdot 2I_0 z)$ corresponds to a wave with a different gain that depends on both the level populations and the polarization of the medium (a nonlinear interference effect). This second wave bears information about the nonlinear interaction of the waves and causes fine structure on the weak-signal gain line; sharp changes of the gain near the resonance $\omega_1 = \omega$ are very sensitive to relaxation processes in the medium. During generation with $\gamma_2 \ll \gamma_1$ a narrow dip of width γ_2 appears in the vicinity of $\omega_1 = \omega$ on the gain line. At the center of the dip we have $I_0'' > 0$, which means that in the vicinity of the narrow dip the second wave is always weaker than the first. The situation is similar for the combination frequency ω_2. In accordance with (24) the narrow dip has maximal depth when $I_0''z \gg 1$. Then the intensity (23) of the weak waves at the center of this dip is about one-half less than at its edges.

When $2|I_0|z \ll 1$ the exponential in the curly brackets of (24) can be expanded in a series. Then in a linear approximation with respect to $I_0 z$ the expression for (24) coincides with the corresponding expansion of a single wave:

$$a_1(z,t) = a_1(0)\exp\{i[(k_1 + I_0 + 2\pi\varkappa/\lambdabar)z - \omega_1 t]\}. \tag{25}$$

The gain of the wave represented by (25) is exactly equal to that given in[6] if in (25) we insert $\delta = 0$ and neglect the contribution from the term $\gamma\rho_{\sigma\sigma}$ in (4).

We now give the final result for the important case of a homogeneously broadened line ($ku \ll \Gamma$) subject to the inequalities

$$\delta \ll \gamma_2 \ll \gamma_1, \quad g\gamma_2/\gamma_1 \ll 1,$$

where $g = 3_\gamma \lambdabar A_0^2/\hbar\gamma_1\gamma_2$ is the saturation parameter. In the vicinity of a narrow dip with $|\Omega| \ll \gamma_1$ the intensity and shape of the weak signal are given by (23) and (24), in which $I_1'' = q - I_0''$,

$$q = \frac{3\pi N_0\lambdabar^2\gamma}{\gamma_1(1+2g)}, \quad I_0' = -\frac{I_0''\Omega}{\gamma_2(1+g)}, \tag{26}$$
$$I_0'' = \frac{qg(1+g)\gamma_2^2}{\Omega^2+\gamma_2^2(1+g)^2},$$

where the single-primed term is the real part, and the double-primed term is the imaginary part, of the given quantity.

For typical values of the experimental parameters:

$$z \sim 10^2 \text{ cm.} \quad qz \sim 1, \quad g \sim 1$$

the expansion in terms of $I_0 z$ is not permissible. In this case the intensity (23) of the weak signal has a convenient form since the characteristic fine structure of the gain line is caused by the argument of the exponential:

$$I(z) = I(0) e^{2qz}(1 + e^{-4I_0'' z}) / 2.$$

When determining the width γ_2 of the upper level from the experimental gain line it must be remembered that the strong field broadens the narrow dip in accordance with (26). With further enhancement of the strong field ($g\gamma_2/\gamma_1 \gg 1$) the width of the dip reaches the order Γ and the resonance levels split into two doublets.[6]

In the approximation (19) the intensity $I_{\omega_2}(z)$ of the combination frequency $\omega_2 = 2\omega - \omega_1$ is appreciably smaller than the value already represented in (20):

$$I_{\omega_2}(z) = I_{\omega_2}(0) e^{Kz} \left| \frac{3\pi}{2} R \int dv \frac{fN_0 \lambda\!\!\!^{-2} z\gamma F_+}{\Delta - kv + i\Gamma/2} \right|^2. \tag{27}$$

The intensity of the combination frequency ω_2 differs from zero only in the vicinity of the $\omega_2 = \omega$ resonance; the width of this spectral interval is determined by the smaller of the widths γ_1 and γ_2. Despite the smallness of (27) as compared with (20), beats of the intensity of the combined field of frequencies ω_1 and ω_2 will always be observed for those values of the experimental parameters that permit the observation of a narrow dip. This result is associated with the fact that the combined field includes an interference term containing the product of the amplitudes a_1 and a_2^* and equaling the third term in (20) in order of magnitude.

4. THE CASE OF ORTHOGONAL POLARIZATIONS

From the foregoing discussion it is clear that a specific interaction of monochromatic waves takes place when their frequencies lie within the limits of the natural width of the resonance levels. Far from resonance ($|\Omega| \gg \Gamma$) no combination frequency appears and the weak-wave gain is of the usual form. We shall therefore begin by solving (5)—(10) in the vicinity of the narrow dip, assuming $\omega_1 = \omega$ and $\omega_2 = \omega$ in (7) and (8). This means that we are neglecting Ω as compared with Γ. We shall then consider the case of (19) when level broadening by the strong field can be neglected. The resulting equations are simplified by expanding all expressions in terms of the strong field. With the foregoing assumptions, from (9) and (10) we have

$$(r_{\alpha\beta} - r\delta_{\alpha\beta}) l_\beta = -\frac{3N\gamma f F_+}{2A_0}\left[\left(\frac{2iR}{3\gamma} - R_1\right)(\mathbf{l}\mathbf{a}_1) l_\alpha + \left(\frac{2iR}{3\gamma} - R_2\right)(\mathbf{l}\mathbf{a}_2^*) l_\alpha + R_1 a_{1\alpha} + R_2 a_{2\alpha}^*\right], \tag{28}$$

$$R_1 = \frac{1}{\gamma_2 - i\Omega + \delta(Q+G)} + \frac{1}{\gamma_2 - i\Omega + \delta(Q-G)},$$

where $\mathbf{l}$ is the polarization vector of the strong wave and R_2 is the difference between the fractions in (28).

Equations (5)—(8) now reduce to two equations for the amplitudes of weak waves with polarizations that are perpendicular to $\mathbf{l}$:

$$(icp + \omega_1 - \omega_k + R_{11}) a_1 + R_{12} a_2^* = ica_1(0), \tag{29}$$

$$R_{21} a_1 - (icp - \omega_1 + \omega_k - R_{22}) a_2^* = 0, \tag{30}$$

where we have introduced the notation

$$R_{11} = 2\pi\omega_0\varkappa_\perp - \frac{9\pi}{4} R_1 \int dv \frac{fN_0 \lambda\!\!\!^{-2} c\gamma^2 F_+}{\Delta - kv + i\Gamma/2}, \tag{31}$$

$$R_{22} = 2\pi\omega_0\varkappa_\perp^* - 2\Omega - \frac{9\pi}{4} R_1 \int dv \frac{fN_0 \lambda\!\!\!^{-2} c\gamma^2 F_+}{\Delta - kv - i\Gamma/2}. \tag{32}$$

R_{12} and R_{21} equal the last terms of (31) and (32), respectively, when we substitute $R_1 \to R_2$, and $\kappa_\perp$ is given by (15) with the substitution

$$N \to N_0(1 - \Lambda_\perp F),$$

$$\Lambda_\perp = \frac{3\gamma}{\gamma_1}\left(1 - \frac{\gamma}{\gamma_2}\right) + \frac{\gamma}{\gamma_2}\left(1 - \frac{\gamma_2}{\gamma_2 + \delta(Q+G)}\right).$$

Equations (29) and (30) are solved easily. Inverting from the Laplace transform to coordinate dependence, we obtain

$$a_1(z) = a_1(0)\left[1 + \frac{iz}{c}\left(\omega_1 - \omega_k + \frac{R_{11} + R_{22}}{2}\right)\right] \exp\left(\frac{i(R_{11} - R_{22})z}{2c}\right).$$

The exponential here must also be expanded and will retain the term with A_0^2. Finally, the intensity $I_\perp(z)$ of a weak wave having the frequency ω_1 and orthogonal polarization in the vicinity of a narrow dip can be written as

$$\frac{I_\perp(z)}{I_\perp(0)} = e^{Kz}\left\{1 - \frac{3\pi}{2}\int dv \frac{fN_0 \lambda\!\!\!^{-2} z\gamma\Gamma}{(\Delta - kv)^2 + \Gamma^2/4} \times \left[\Lambda_\perp F + \frac{3F_+}{2}\frac{\gamma[\gamma_2 + \delta(Q+G)]}{\Omega^2 + [\gamma_2 + \delta(Q+G)]^2}\left(1 - \frac{2\Omega(\Delta - kv)}{\Gamma[\gamma_2 + \delta(Q+G)]}\right) + \frac{3F_+}{2}\frac{\gamma[\gamma_2 + \delta(Q-G)]}{\Omega^2 + [\gamma_2 + \delta(Q-G)]^2}\left(1 - \frac{2\Omega(\Delta - kv)}{\Gamma[\gamma_2 + \delta(Q-G)]}\right)\right]\right\}. \tag{33}$$

Equation (33) is the part of the Doppler contour where a dip exists at the center of a Bennett hole at the point where the strong and weak field frequencies coincide. This dip characteristically contains the width γ_2 of the degenerate level. This result is associated with the fact that atoms having zero projection of the total angular momentum can only absorb a weak wave with orthogonal polarization. Only atoms with ± 1 projection of the angular momentum participate in amplifying the wave with orthogonal polarization. Therefore the narrow dip (33) contains the width γ_2 of the upper level.

When broadening of the degenerate level is caused predominantly by elastic collisions, then in the case of orthogonal polarizations the observed dip has the width δ ($ku \gg \delta$) like the Bennett holes. For parallel polarization under the same conditions the width of the dip is determined by the smaller of the widths γ_1 and γ_2. In the opposite limiting case, $\delta \ll \gamma_2$ and $\delta \ll \gamma_1$, the width of the narrow dip for parallel polarizations does not change, but for orthogonal polarizations it assumes the value γ_2.

The elastic-collision dependence of Bennett holes in the weak-wave gain line for orthogonal polarization is accounted for as follows. Pumping excites atoms with a Maxwellian velocity distribution identically on all three sublevels. We orient the quantization axis along the polarization vector of the strong field. Then the strong field will induce transitions only between atomic states with zero projections of the total angular momentum. The excited atoms with projections ± 1 will have their velocities distributed according to a Maxwellian function that is not perturbed by the strong field. A weak field with orthogonal polarization interacts only with these atoms. Therefore, far from the narrow dip the gain line of this weak field is Dopplerian with small Bennett holes that are caused by corresponding prominences on the velocity distribution of atoms on the lower level ($\gamma_2 \ll \gamma_1$). Intense elastic collisions mix the atoms on the sublevels and make the Bennett holes identical for atoms on all three sublevels. Consequently the gain line of a weak wave with orthogonal polarization becomes similar to the gain line for parallel polarizations if we neglect the aforementioned narrow dip.

The gain $K_\perp$ of a weak wave with orthogonal polarization far from a narrow dip is

$$K_\perp = \frac{3\pi}{2}\int dv \frac{fN_0\lambda^2\gamma\Gamma}{(\omega_1-\omega_0-kv)^2+\Gamma^2/4}\frac{1+(\Lambda_\parallel-\Lambda_\perp)F}{1+\Lambda_\parallel F}. \quad (34)$$

However, the gain $K_\parallel$ of a weak wave with parallel polarization under identical conditions is given by (34) without the second term in the numerator. Thus the downward shifting of the entire gain-line contour as A_0 increases will depend on the character of the wave polarization; this is manifested in the relative depth of the narrow dip. For strong elastic collisions $K_\parallel$ and $K_\perp$ become identical outside the narrow dip.

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[1] I. M. Beterov and V. P. Chebotaev, ZhETF Pis. Red. 9, 216 (1969) [JETP Lett. 9, 127 (1969)].
[2] C. V. Shank and S. E. Schwarz, J. Quant. Elect. QE-4, 1018 (1968).
[3] T. Ya. Popova, A. K. Popov, S. G. Rautian, and R. I. Sokolovskiĭ, Zh. Eksp. Teor. Fiz. 57, 850 (1969) [Sov. Phys.-JETP 30, 466 (1970)].
[4] M. A. Gubin, A. I. Popov, and E. D. Protsenko, Zh. Eksp. Teor. Fiz. 54, 701 (1968) [Sov. Phys.-JETP 27, 374 (1968)].
[5] T. Ya. Popova, A. K. Popov, S. G. Rautian, and A. A. Feoktistov, Zh. Eksp. Teor. Fiz. 57, 444 (1969) [Sov. Phys.-JETP 30, 243 (1970)].
[6] S. G. Rautian, Trudy FIAN 43, 3 (1968).
[7] W. R. Bennett, Phys. Rev. 126, 580 (1962).
[8] W. R. Bennett, Appl. Opt. Suppl. No. 1 on Optical Masers, 1962, p. 24.
[9] T. I. Kuznetsova and S. G. Rautian, Zh. Eksp. Teor. Fiz. 49, 1605 (1965) [Sov. Phys.-JETP 22, 1098 (1966)].
[10] T. I. Kuznetsova, Trudy FIAN 43, 116 (1968).
[11] J. R. Fontana, R. H. Pantell, and R. G. Smith, J. Appl. Phys. 33, 2085 (1962).
[12] B. Senitzky, G. Gould and S. Cutler, Phys. Rev. 130, 1460 (1963).
[13] W. E. Lamb, Phys. Rev. 134, A1429 (1964).
[14] A. Dienes, Phys. Rev. 174, 400 and 414 (1968).
[15] A. I. Alekseev and V. M. Galitskiĭ, Zh. Eksp. Teor. Fiz. 57, 1002 (1969) [Sov. Phys.-JETP 30, 548 (1970)].
[16] A. I. Alekseev and V. M. Galitskiĭ, report at All-Union Symposium on the Physics of Gas Lasers, Novosibirsk, July, 1969.
[17] C. H. Wang and W. J. Tomlinson, Phys. Rev. 181, 115 (1969).
[18] C. H. Wang, W. J. Tomlinson, and R. T. George, Phys. Rev. 181, 125 (1969).
[19] A. I. Alekseev, in Prokhozhdenie izlucheniya cherez veshchestvo (The Passage of Radiation through Matter), Atomizdat, 1968, p. 17.

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