Polarization Effects in Passage of Radiation Through a Resonant Medium

V. M. ARUTYUNYAN, É. G. KANETSYAN, AND V. O. CHALTYKYAN

Institute of Physics Research, Armenian Academy of Sciences

Submitted August 3, 1971

Zh. Eksp. Teor. Fiz. 62, 908-917 (March, 1972)

The polarization properties of an intense monochromatic wave traversing a resonance medium of two-level atoms without absorption are studied. The magnetic sublevel shifts of the atoms, refractive index and Stokes parameters are calculated. It is shown that the polarization ellipse rotates on traversal of the wave, and the rotation angle is calculated. Dispersion properties of the medium are considered. Expressions are obtained for the refractive index and absorption coefficients of a weak wave. Three-photon scattering, resonance absorption line splitting and four-photon parametric interaction effects for various cases of polarization of an intense wave are also considered.

1. INTRODUCTION

WHEN intense optical radiation acts on a resonant two-level medium, effects are produced that are missing in linear optics. In accordance with the character of their manifestation, these effects can be divided into two classes, self-action and multiphoton phenomena.

If the deviation from resonance is large and absorption can be neglected, then all the self-action effects are connected with the nonlinear part of the refractive index. Just as in transparent media, this nonlinearity causes self-focusing and defocusing, and also multiple broadening of the spectrum. The self-focusing phenomena in potassium vapor were first observed in and self-modulation spectrum broadening was observed in connected in transparent media. Among the self-action effects far from resonance is included also nonlinear delay of light (the dependence of the group velocity on the intensity). This effect, however, has not yet been observed experimentally.

The principal multiphoton phenomena in a two-level system are three-photon scattering and four-photon parametric interactions. In three-photon scattering, the atom absorbs two incident quanta of frequency ω , emits a quantum $2\omega-\omega_0$, and goes over into the excited states. In the four-photon effect, the atom absorbs two quanta of frequency ω and emits two others in accordance with the scheme $2\omega-\omega_1+\omega_2$.

Owing to the presence of coherence, four-photon interaction proceeds mainly in the direction of the incident radiation, thus leading to a broadening of the $\operatorname{spectrum}^{[2,3]}$. The angular features of four-photon scattering were investigated in[4,5]. Three-photon scattering in the direction of the incident radiation in rubidium vapor was first observed in[6]. However, as explained in[7], the three-photon process in the direction of the incident radiation is strongly suppressed by the competition of the four-photon scattering. This explains why no three-photon scattering was observed in [8]. In the direction opposite to the incident radiation, threephoton scattering always takes place and is appreciably enhanced[9]. An interesting feature of a resonant medium is also the high-frequency Stark effect, which has been investigated in detail for the absorption line

 $in^{[10,11]}$ and for the three-photon emission line $in^{[9]}$.

A theoretical analysis of these effects is based as a rule on the scalar equations of the resonant medium. Whereas in the linear theory this is justified, in the nonlinear case specific polarization phenomena occur. The equations of the resonant medium, with allowance for the polarization of the waves, have been formulated in terms of the density matrix, and in some cases (gas lasers, photon echo, self-induced transparency) they have been investigated in[12] (where a more detailed literature can be found). In the present paper we solve the Schrödinger equation, after which we calculate the polarizability of the medium, which enters in Maxwell's equations. Such a method is more lucid and makes it possible to calculate the energy shifts and predict immediately the possible absorption and emission processes and their cross sections[13].

The first two sections of this paper are devoted to the study of the polarization properties of an intense monochromatic wave passing through a resonant medium without absorption. We calculate the atomic level shifts, the refractive index, and the Stokes parameters. We show that the polarization ellipse is rotated in the course of passage, and calculate the angle of rotation. We note that the rotation of the polarization ellipse was obtained in [12] for the most general case of an absorbing medium without an explicit form of the dependence of the rotation angle on the intensity. In our concrete problem we are able to calculate exactly (in terms of the intensity) all the coefficients (see formula (3.5) for the rotation angle).

Sections 3 and 4 are devoted to the study of the dispersion properties of a resonant medium. Formulas are obtained for the refractive indices of a weak wave, for the line splitting in resonant absorption and in three-photon scattering, and also for four-photon parametric effects in different cases of polarization of an intense monochromatic wave.

Since relaxations are neglected throughout, i.e., it is assumed that the pulse duration is much shorter than the relaxation time, a monochromatic wave is understood in the sense of $T_{1,2} \gg T \gg \epsilon^{-1}$ (T is the pulse duration, ϵ the deviation from resonance, and $T_{1,2}$ the relaxation times). The question of turning on the interaction with the monochromatic field has been discussed in detail in $[1^{-3,7,13}]$.

2. BOUND STATES OF AN ATOM IN THE FIELD OF AN INTENSE WAVE

We consider the interaction of a two-level atom, which has in the ground state an energy E_1 and an angular momentum $j_1=1/2$, and in the excited state respectively E_2 and $j_2=3/2$ (for an isolated atom these states are degenerate with respect to the projection of the angular momentum), with an intense wave propagating along the z axis and specified by a vector potential

$$A = Re^{i(kz-\omega t)} + c.c., \tag{2.1}$$

where $\mathbf{R}(\mathbf{z},\,\mathbf{t})$ varies slowly compared with the exponential. The wave functions of the atom in the absence of the field will be designated by $\psi(\mathbf{m})$ and $\Phi(\mu)$ ($\mathbf{m}=\pm\frac{1}{2}$, $\mu=\pm\frac{1}{2}$, $\pm\frac{3}{2}$), and the solution of the Schrödinger equation

$$i\hbar \frac{\partial \Psi}{\partial t} = (H_0 - d\mathbf{E}) \Psi, \quad H_0 \begin{pmatrix} \psi(m) \\ \Phi(\mu) \end{pmatrix} = E_{1,2} \begin{pmatrix} \psi(m) \\ \Phi(\mu) \end{pmatrix}$$
 (2.2)

will be sought in the form

$$\Psi = a(m)\psi(m)\exp\left(-\frac{i}{\hbar}E_1t\right) + b(\mu)\Phi(\mu)\exp\left(-\frac{i}{\hbar}E_2t + i\varepsilon t\right),$$
(2.3)

where d is the operator of the dipole moment of the atom, $\mathbf{E} = -\partial \mathbf{A}/c\partial t$, $\epsilon = \omega_0 - \omega$, $\omega_0 = (\mathbf{E}_2 - \mathbf{E}_1)/\hbar$. Then, in the case of a monochromatic wave $(\mathbf{R} = \mathbf{R}(z))$ we obtain the following values for the split energy levels of the ground and excited states $(\mathbf{E}_1')_{1,2}$ and $(\mathbf{E}_2')_{1,2,3}$:

$$(E_{1}')_{1,2} = E_{1} + \frac{1}{2}\hbar\epsilon(1 - \Xi_{1,2}),$$

$$(E_{2}')_{1,2} = E_{2} - \frac{1}{2}\hbar\epsilon(1 - \Xi_{1,2}),$$

$$(E_{2}')_{3} = E_{2}.$$
(2.4)

Here

$$\Xi_{i,2} \equiv \sqrt{1 + \xi_{i,2}};$$

 $\xi_{1,2}$ are dimensionless intensity parameters:

$$\xi_{1,2} = \frac{4\pi |d|^2}{3c\hbar^2c^2} P\left(1 \mp \frac{\eta_2}{2}\right); \tag{2.5}$$

d is the reduced matrix element of the transition, $P=\omega^2|R|^2/2\pi c$ is the energy flux in the wave (2.1), and η_2 is the Stokes parameter corresponding to circular polarization. The polarization tensor is specified in the form

$$J_{\alpha\beta} = \frac{\omega^2}{2\pi c} \overline{R_{\alpha} R_{\beta}}, \quad \alpha, \beta = x, y, \quad \operatorname{Sp} \hat{J} = P,$$

$$\hat{J} = \frac{P}{2} \begin{pmatrix} 1 + \eta_3 & \eta_1 - i\eta_2 \\ \eta_1 + i\eta_2 & 1 - \eta_3 \end{pmatrix},$$
(2.6)

where $\eta_{3,1}$ determine the linear polarization along the x axis and at an angle $\pi/4$ to this axis. The wave functions of the states $(E_1')_{1,2}$ (the atom was in the ground state prior to the turning on of the interaction) are then equal to

$$\Psi_{1,2} = C_{1,2} \left\{ \exp\left(-\frac{i}{\hbar} E_1 t\right) \psi\left(\mp \frac{1}{2}\right) + \frac{i\omega_0 d^*}{c\hbar \epsilon \, \gamma 6} \frac{e^{i\hbar \epsilon}}{1 + \Xi_{1,2}} \left[R^{(\mp)} \Phi\left(\pm \frac{1}{2}\right) - \sqrt{3} \, R^{(\pm)} \Phi\left(\mp \frac{3}{2}\right) \right] \right. \\ \left. \times \exp\left(-\frac{i}{\hbar} E_2 t + i\epsilon t\right) \right\} \exp\left[\frac{i\epsilon t}{2} (\Xi_{1,2} - 1)\right],$$

$$\left| C_{1,2} \right|^2 = (\Xi_{1,2} + 1)/2\Xi_{1,2},$$
(2.7)

where we have introduced the notation $R^{(\pm)} = R_X \pm iR_y$.

Let us compare the atomic level shifts determined by formulas (2.4) and (2.5) for different cases of polarized

light. In the case of linear polarization $\eta_2 = 0$, $\xi_1 = \xi_2 = \xi = 4\pi |\mathbf{d}|^2 P/3 \mathrm{ch}^2 \epsilon^2$, and in the case of circular polarization $\eta_2 = 1$ and $\xi_1 = \xi/2 = \xi_2/3$, so that at the same wave power the maximum shift is obtained in the case of circular polarization. In the case of completely unpolarized light we also have $\xi_1 = \xi_2 = \xi$, and in the sense of the level shift such light behaves as if it were linearly polarized.

It is interesting to note that in the general case population inversion takes place between the magnetic sublevels (see (2.7)).

3. PASSAGE OF INTENSE MONOCHROMATIC WAVE THROUGH A RESONANT MEDIUM

Let us consider the passage of an intense wave (2.1) through a resonant medium of the atoms described in Sec. 2. Assuming that the atom was in the ground state prior to the turning on of the interaction, writing down the equation

$$\left(\Delta - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) A = -\frac{4\pi n}{c} \frac{\partial}{\partial t} \langle \mathbf{d} \rangle \tag{3.1}$$

(n is the density of the atoms) for the potential in the approximation of electric-dipole resonant transitions, and using the functions (2.7) to calculate the quantum-mechanical mean value $\langle \mathbf{d} \rangle$ of the dipole moment of the atom, we obtain the following transmission equations for the components $\mathbf{R}^{(\pm)}$ of the wave vector potential amplitude,

$$\frac{d}{dz}R^{(\pm)} = iq \left(\frac{3}{\Xi_{1,2}} + \frac{1}{\Xi_{2,1}}\right) R^{(\pm)},$$

$$q = \pi\omega_0 |d|^2 n/12c\hbar\varepsilon.$$
(3.2)

It follows from these equations that the parameters $\xi_{1,2}$ are not altered by the passage of the wave. Thus, we can introduce for the wave components $R^{(t)}$ the refractive indices

$$n^{(\pm)} = 1 + \frac{qc}{\omega_0} \left(\frac{3}{\Xi_{1,2}} + \frac{1}{\Xi_{2,1}} \right),$$
 (3.3)

as follows from the solution of (3.2). Unlike the components $\mathbf{R}^{(\pm)}$, in the general case there is no wave vector for the Cartesian components $\mathbf{R}_{\mathbf{X},\mathbf{y}}$. Equations (3.2) also make it possible to explain the polarization properties of the transmitted radiation. If the incident wave is circularly polarized (we assume that $\mathbf{R}^{(+)}(0)=0$), then $\mathbf{R}^{(+)}(\mathbf{z})=0$ and the polarization remains unchanged. A linearly polarized wave likewise does not change polarization. For such a wave, the refractive index of the medium is given by

$$n = 1 + 4qc / \omega_0 \Xi. \tag{3.4}$$

In the general case of elliptic polarization, Eqs. (3.2) lead to rotation of the axes of the polarization ellipse (without deformation) through an angle γz , where

$$\gamma = 2q(1/\Xi_1 - 1/\Xi_2). \tag{3.5}$$

We can consider the passage of a quasimonochromatic wave by regarding $R^{(\pm)}(0,t)$ as slowly varying in time and replacing in (3.2) d/dz by d/dz - d/cdt. Then the solutions of these equations will depend on R(0, t-z/c) and $\xi(0, t-z/t)$. We write down these solutions for the Stokes parameters, since the polarization properties of quasimonochromatic radiation are determined by the tensor (2.6) (even though in the gen-

eral case the polarization tensors are of higher order, we assume here that all reduce to a second-order tensor):

$$P(z) = P, \quad \eta_2(z) = \eta_2,$$

$$\eta_{1,3}(z) = \eta_{1,3} \cos \gamma z \pm \eta_{3,1} \sin \gamma z.$$
(3.6)

These relations show, in particular, that the degree of polarization $\eta = (\eta_1^2 + \eta_2^2 + \eta_3^2)^{1/2}$ of the radiation does not change on passage through the medium. Nor do the power P and the probability η_2 of circular polarization change. On the other hand, the parameters $\eta_{1,3}$ oscillate with frequency γz . It is interesting to note that by determining experimentally the length of the oscillations $z_0 = 2\pi/\gamma$ we can determine the degree η_2 of the circular polarization (if we also know the power of the wave), as follows from (3.5) and (2.5) (in the case of small nonlinearities $\xi_{1,2} \ll 1$ we have $z_0 \approx 2\pi/q\xi\eta_2$). The same oscillations cause the parameters η_1 and η_3 to be modulated over the cross section of the beam with a characteristic modulation dimension equal to ${f r_0}({
m qz}\,\xi_{\,0}\eta_{\,2})^{-1}\ll{f r_0}$ when $\xi \ll 1$ (r₀ is the beam radius, and ξ_0 is the value of ξ at the center).

Formulas (3.6) have one more interesting feature. Let us assume that the incident beam is almost completely polarized along the x axis, i.e., $\eta_3 \approx 1$, $\eta_1 = 0$, $\eta_2 \ll 1$. Then at low intensities it follows from (3.6) that $\eta_3(z) \approx \cos{(qz\xi\eta_2)}$. We then have for the probabilities $w_{x,y}$ of the polarizations along the axes x and y respectively, starting from the definition (2.6).

$$w_{x,y} = \frac{1}{2} [1 \pm \cos(qz\xi\eta_2)].$$
 (3.7)

Therefore, even when $\eta_2 \ll 1$, provided only $qz \xi \eta_2 \gtrsim 1$, the appearance of a y component of the field has a considerable probability.

We note that all the foregoing pertains to the integral (with respect to time) characteristics of the beam. If we are interested in the waveform of the pulse, then it is not necessary to carry out the averaging indicated in (2.6). In this case we can use the solutions of (3.2). In each section of the beam the quantities $|R_{x,y}|^2$ will be modulated in time, and the time of such modulation will be of the order of $T(qz\xi)^{-1} \ll T$ (T is the pulse duration).

4. DISPERSION CHARACTERISTICS OF RESONANT MEDIUM

To study the dispersion properties of the medium, let us consider the passage of a weak wave through the medium in the presence of an intense wave propagating the opposite direction ($\sec^{[7]}$), i.e., we consider in place of (2.1) a potential in the form

$$A = R(z)e^{i(kz-\omega t)} + A_{z}(z, t)e^{-i(kz+\omega t)} + \text{c.c.},$$

$$|A_{z}(z, t)| \le |R(z)|.$$
(4.1)

The atomic amplitudes a(m) and $b(\mu)$ in (2.3) will also be represented in the form of sums

$$a(m) = a_1(m) + a_2(m), \quad b(\mu) = b_1(\mu) + b_2(\mu),$$

where $|a_2(m)| \ll |a_1(m)|$, $|b_2(\mu)| \ll |b_1(\mu)|$. Further, calculating the weak component of the Doppler moment of the atom in an approximation linear in $A_2(z, t)$ and lowering the order of Eq. (3.1) for $A_2(z, t)$, we obtain the following equations for the Fourier components $F(z, \omega')$

of the weak field:

$$\frac{\partial}{\partial z}F^{(\pm)}(z,\omega') = \alpha^{(\pm)}(\omega')F^{(\pm)}(z,\omega') + i\beta^{(\pm)}(\omega')e^{i\gamma z}F^{(\mp)}(z,\omega').$$

$$F^{(\pm)}(z,\omega') = F_{z}(z,\omega') + iF_{z}(z,\omega').$$
(4.2)

where the coefficients $\alpha^{(\pm)}(\omega')$ and $\beta^{(\pm)}(\omega')$, which are functions of the running frequency ω' , are given by

$$\alpha^{(+)} = i \frac{\omega - \omega'}{c} - 3iq\varepsilon \left\{ \frac{\Xi_{1} + 1}{2\Xi_{1}} \left(1 - \frac{\omega_{0}^{2} |d|^{2} |R^{(+)}|^{2}}{2\hbar^{2}c^{2}\varepsilon^{2}\xi_{1}} \right) \right.$$

$$\times \frac{1}{\omega' - \omega_{1a'} + i\delta/2} + \frac{\omega_{0}^{2} |d|^{2} |R^{(+)}|^{2}}{2\hbar^{2}c^{2}\varepsilon^{2}\xi_{1}}$$

$$\times \left[\frac{(\Xi_{1} + 1)^{2}}{4\Xi_{1}^{2}} - \frac{1}{\omega' - \omega_{1a} + i\delta/2} - \frac{(\Xi_{1} - 1)^{2}}{4\Xi_{1}^{2}} - \frac{1}{\omega' - \omega_{1s} + i\delta/2} \right] \right\}$$

$$- iq\varepsilon \left\{ \frac{\Xi_{2} + 1}{2\Xi_{2}} \left(1 - \frac{\omega_{0}^{2} |d|^{2} |R^{(+)}|^{2}}{6\hbar^{2}c^{2}\varepsilon^{2}\xi_{2}} \right) - \frac{1}{\omega' - \omega_{2a'} + i\delta/2} + \frac{\omega_{0}^{2} |d|^{2} |R^{(+)}|^{2}}{6\hbar^{2}c^{2}\varepsilon^{2}\xi_{2}} \right]$$

$$\times \left[\frac{(\Xi_{2} + 1)^{2}}{4\Xi_{2}^{2}} - \frac{1}{\omega' - \omega_{2a} + i\delta/2} - \frac{(\Xi_{2} - 1)^{2}}{4\Xi_{2}^{2}} - \frac{1}{\omega' - \omega_{2s} + i\delta/2} \right] \right\};$$

$$\beta^{(+)} = q\varepsilon \frac{\omega_{0}^{2} |d|^{2}}{8\hbar^{2}c^{2}} R^{(-)*}(0) R^{(+)}(0) [f(\omega', \xi_{1}) + f(\omega', \xi_{2})],$$

$$\beta^{(-)}(\omega') = \beta^{(+)*}(\omega'),$$

$$4 - \Gamma \Xi_{1} + 1 - 1$$

$$f(\omega', \xi_1) = \frac{4}{\varepsilon^2 \xi_1} \left[\frac{\Xi_1 + 1}{2\Xi_1} \frac{1}{\omega' - \omega_{1a}' + i\delta/2} \right]$$

$$\times \frac{(\Xi_1 + 1)^2}{4\Xi_1^2} \frac{1}{\omega' - \omega_{1a} + i\delta/2} - \frac{(\Xi_1 - 1)^2}{4\Xi_1^2} \frac{1}{\omega' - \omega_{1s} + i\delta/2} \right]$$

and the coefficient $\alpha^{(-)}(\omega')$ is obtained from (4.3) by means of the substitutions $\xi_1 \to \xi_2$, $\mathbf{R}^{(+)} \to \mathbf{R}^{(-)}$. The frequencies $\omega_{1\mathbf{a},\,2\mathbf{a},\,1\mathbf{S},\,2\mathbf{S}}$ and $\omega'_{1\mathbf{a},\,2\mathbf{a}}$ which enter in (4.3) and (4.4) and determine the physical processes that occur when an atom having energy levels (2.4) interacts with an intense monochromatic wave $\mathbf{R}(\mathbf{z})$, are given by the formulas

$$\omega_{1a, 2a} = \omega + \varepsilon \Xi_{1,2}, \quad \omega_{1s, 2s} = \omega - \varepsilon \Xi_{1,2},
\omega'_{1a, 2a} = \omega + \frac{1}{2} \varepsilon (\Xi_{1,2} + 1).$$
(4.5)

The intense-wave amplitude R(z) in (4.3) and (4.4) is the solution of Eq. (3.2), and the infinitesimal imaginary increment $i\,\delta/2$ in the denominators was introduced to ensure correct circuiting around the poles.

In the general case, when the intense wave is elliptically polarized, the components $F^{(+)}$ and $F^{(-)}$ of the weak radiations become coupled, as seen from (4.2), with a coupling coefficient $\beta^{(+)}(\omega')$, and the general solution (4.2) takes the form

$$F^{(\pm)}(\omega',z) = C_1^{(\pm)}(\omega')e^{\tau_1(\omega')z} + C_2^{(\pm)}(\omega')e^{\tau_2(\omega')z}$$
(4.6)

where the roots $\mathbf{r}_{1,2}(\omega')$ of the characteristic equation are equal to

$$r_{i,2}(\omega') = \frac{1}{2}(\alpha^{(+)} + \alpha^{(-)} + i\gamma)$$

$$\pm \left\{ \frac{1}{4}(\alpha^{(+)} - \alpha^{(-)} - i\gamma)^2 - |\beta^{(+)}(\omega')|^2 \right\} \frac{1}{2}.$$
(4.7)

Since $r_{1,2}$ are pure imaginary, as seen from (4.3) and (4.7), the parametric coupling of the components $F^{(+)}$ and $F^{(-)}$ does not lead to the occurrence of regions of exponential amplification, and can be attributed, as follows from (4.4), to a four-photon process of the type $(R^{(+)} + (F^{(-)}) \rightarrow (R^{(-)}) + (F^{(+)})$, which proceeds without a change of frequency.

Since the coupling coefficient is proportional to $\beta^{(+)} \sim (4\pi \, \mathrm{cP}/\omega^2) \cdot (\eta_3 + \mathrm{i}\, \eta_1)$, i.e., it does not contain the probability of the circular polarization, the components

 $F^{(+)}$ and $F^{(-)}$ do not become coupled for a circularly polarized ($\eta_3 = \eta_1 = 0$, $\eta_2 = 1$) intense monochromatic light, and for each of them we can introduce a refractive index and an absorption coefficient. For the case when $R^{(+)} = 0$ and $R^{(-)} \neq 0$, the latter are given by

$$n_{2}^{(+)}(\omega') = 1 + \frac{q\varepsilon c}{\omega} \left(3 \frac{\Xi_{1} + 1}{2\Xi_{1}} \frac{1}{\omega - \omega_{1a}} + \frac{\Xi_{2} + 1}{2\Xi_{2}} \frac{1}{\omega - \omega_{2a}} \right),$$

$$\kappa_{2}^{(+)}(\omega') = 2\pi q\varepsilon \left[3 \frac{\Xi_{1} + 1}{2\Xi_{1}} \delta(\omega' - \omega_{1a}') + \frac{\Xi_{2} + 1}{2\Xi_{2}} \delta(\omega' - \omega_{2a}') \right]; (4.8)$$

$$n_{2}^{(-)}(\omega') = 1 + \frac{q\varepsilon c}{\omega} \left\{ 3 \frac{(\Xi_{2} + 1)^{2}}{4\Xi_{2}^{2}} \frac{1}{\omega' - \omega_{2a}} - 3 \frac{(\Xi_{2} - 1)^{2}}{4\Xi_{2}^{2}} \frac{1}{\omega' - \omega_{2a}} + \frac{(\Xi_{1} + 1)^{2}}{4\Xi_{1}^{2}} \frac{1}{\omega' - \omega_{1a}} - \frac{(\Xi_{1} - 1)^{2}}{4\Xi_{1}^{2}} \frac{1}{\omega' - \omega_{1a}} \right\}, \quad (4.9)$$

$$\begin{split} \kappa_{2}^{(-)}(\omega') &= 2\pi q\epsilon \left[3 \frac{(\Xi_{2} + 1)^{2}}{4\Xi_{2}^{2}} \, \delta(\omega' - \omega_{2a}) - \frac{3(\Xi_{2} - 1)^{2}}{4\Xi_{2}^{2}} - \delta(\omega' - \omega_{2a}) \right. \\ &\left. + \frac{(\Xi_{1} + 1)^{2}}{4\Xi_{1}^{2}} \, \delta(\omega' - \omega_{1a}) - \frac{(\Xi_{1} - 1)^{2}}{4\Xi_{1}^{2}} \, \delta(\omega' - \omega_{1a}) \, \right], \end{split}$$

where the indices (±) pertain to the waves $F^{(\pm)}$ and $\xi_2 = 3\xi_1$.

Formulas (4.9) show that the component $F^{(-)}$ of the weak wave polarized along the same circle as the intense wave experiences a split resonant absorption and amplification at the split three-photon frequencies

$$\omega_{1a, 2a} \approx \omega_0 + \frac{1}{2} \epsilon \xi_{1,2}, \quad \omega_{1a, 2s} \approx 2\omega - \omega_0 - \frac{1}{2} \epsilon \xi_{1,2}$$

$$(\xi_{1,2} \leqslant 1).$$
(4.10)

On the other hand, the component $F^{(+)}$, which is circularly polarized in the opposite direction, is only absorbed at the frequencies

$$\omega'_{1a,2a} \approx \omega_0 + \frac{1}{4} \epsilon \xi_{1,2} \quad (\xi_{1,2} \ll 1).$$
 (4.11)

Formulas (4.10) and (4.11) give the Stark shifts of the absorption-line components in the case of small non-linearities.

In the case when the intense wave is linearly polarized, the weak-wave components polarized parallel and perpendicular to the intense wave are likewise not coupled. Thus, if the wave R is polarized along the x axis, then $\eta_1=0$, $\eta_3\neq 0$, and $\beta^{(-)}=\beta^{(+)}$, as a result of which Eqs. (4.2) enable us to introduce refractive indices and absorption coefficients for the components $F_{X,Y}$ of the weak field $(\Xi\equiv\sqrt{1+\xi})$:

$$n_{x} = 1 + \frac{q \varepsilon c}{4 \omega \Xi^{2}} \left[\frac{(\Xi + 1)^{2}}{\omega' - \omega_{a}} - \frac{(\Xi - 1)^{2}}{\omega' - \omega_{s}} \right],$$

$$\kappa_{x} = \frac{\pi q \varepsilon}{2 \Xi^{2}} \left[(\Xi + 1)^{2} \delta(\omega' - \omega_{a}) - (\Xi - 1)^{2} \delta(\omega' - \omega_{s}) \right]; \quad (4.12)$$

$$n_{y} = 1 + 4(n_{x} - 1) + 3 \frac{q \varepsilon c}{\omega} \frac{\Xi + 1}{2\Xi} \frac{1}{\omega' - \omega_{a}'}$$

$$\kappa_{y} = 4\kappa_{x} + 3\pi q \varepsilon \frac{\Xi + 1}{\Xi} \delta(\omega' - \omega_{a}'). \quad (4.13)$$

Inasmuch as the shifts for a linearly polarized wave coincide ($\xi_1 = \xi_2 = \xi$, $\omega_{1a, 1S} = \omega_{2a, 2S} = \omega_{a, S}$, $\omega_{1a}' = \omega_{2a}' = \omega_a'$), the absorption line has two components, ω_a and ω_a' , and absorption at the second frequency is experienced by the component F_y polarized perpendicular to the intense wave. A gain at the three-photon frequency ω_S occurs in both components F_x and F_y . We note that in the case when the wave R is polarized at an angle $\pi/4$ to the x axis ($\eta_1 \neq 0$, $\eta_3 = 0$), there exist refractive indices and absorption coefficients for the weak-wave

components $F_x \pm F_y$ that are polarized at the angles $\pm \pi/4$ to the x axis.

5. PARAMETRIC FOUR-PHOTON INTERACTIONS WITH CHANGE OF FREQUENCY

We now consider the passage of a weak wave in the same direction as the intense wave, i.e., we introduce in place of (4.1) the potential

$$\mathbf{A} = [\mathbf{R}(z) + \mathbf{A}_{2}(z, t)] e^{i(hz - \omega t)} + \mathbf{c.c.},$$
$$|\mathbf{A}_{2}(z, t)| \ll |\mathbf{R}(z)|. \tag{5.1}$$

We solve the problem in the same manner as in Sec. 4. The polarization of the intense wave R will be assumed circular ($\xi_2 = 3\xi_1$) or linear ($\xi_1 = \xi_2 = \xi$), since the general case is too cumbersome.

In the case of circular polarization ($\mathbf{R}^{(+)}=0$) we obtain for the weak-wave Fourier components $\mathbf{F}^{(-)}(\mathbf{z},\,\omega')$ the equation

$$\left(\frac{\partial}{\partial z} + i \frac{\omega - \omega'}{c}\right) F^{(-)}(\omega', z) = 2iq\varepsilon \left[\Lambda_1 \cdot (\omega') F^{(-)}(\omega', z) + \Lambda_2 \cdot (\omega') \exp\left[2iqz\left(\frac{1}{\Xi_1} + \frac{3}{\Xi_2}\right)\right] F^{(-)} \cdot (2\omega - \omega', z)\right],$$
(5.2)

where the coefficients $\Lambda_{1,2}(\omega')$ are given by

$$\Lambda_{1}(\omega') = \frac{(\Xi_{1} + 1)^{2}}{4\Xi_{1}^{2}} \frac{1}{\omega_{1a} - \omega' + i\delta/2} - \frac{(\Xi_{1} - 1)^{2}}{4\Xi_{1}^{2}} \frac{1}{\omega_{1s} - \omega' + i\delta/2} + \frac{3}{4} \left[\frac{(\Xi_{2} + 1)^{2}}{\Xi_{2}^{2}} \frac{1}{\omega_{2s} - \omega' + i\delta/2} - \frac{(\Xi_{2} - 1)^{2}}{\Xi_{2}^{2}} \frac{1}{\omega_{2s} - \omega' + i\delta/2} \right],$$
(5.3)

$$\Lambda_{z}(\omega') = \frac{\omega_{o}^{2} |d|^{2} R^{(-) \cdot z}(0)}{24 \hbar^{2} c^{2} e^{2}} \left\{ \frac{1}{\Xi_{1}^{2}} \left(\frac{1}{\omega_{1s} - \omega' + i\delta/2} - \frac{1}{\omega_{1a} - \omega' + i\delta/2} \right) + \frac{9}{\Xi_{z}^{2}} \left(\frac{1}{\omega_{2s} - \omega' + i\delta/2} - \frac{1}{\omega_{2a} - \omega' + i\delta/2} \right) \right\}.$$
 (5.4)

An investigation of the roots of the characteristic equation (5.2) shows that $F^{(-)}$ interacts parametrically with the intense wave; this gives rise to a region of exponential amplification, which at small nonlinearities ($\xi_{1,2} \ll 1$) is determined by the relation

$$(\omega - \omega')^2 < \frac{5}{2} \varepsilon^2 \xi_1. \tag{5.5}$$

There is no parametric behavior for the component $F^{(+)}$. The refractive index and the absorption coefficient for this component are

$$n^{(+)} = 1 + \frac{q\varepsilon c}{\omega} \left(\frac{3}{2} \frac{\Xi_1 + 1}{\Xi_1} \frac{1}{\omega_{1a}' - \omega'} + \frac{\Xi_2 + 1}{2\Xi_2} \frac{1}{\omega_{2a}' - \omega'} \right),$$

$$\kappa^{(+)} = 2\pi q\varepsilon \left[\frac{3}{2} \frac{\Xi_1 + 1}{\Xi_1} \delta(\omega' - \omega_{1a}') + \frac{\Xi_2 + 1}{2\Xi_2} \delta(\omega' - \omega_{2a}') \right],$$
(5.6)

i.e., the passages of the wave $F^{(+)}$ parallel and antiparallel to the intense wave proceed in the same manner (compare with (4.8)).

In the case of linear polarization $(R_x \neq 0, R_y = 0)$ we have for the components $F_{x,y}$ of the weak radiation

$$\frac{\partial}{\partial z}F_{x,y}(\omega',z) = i\Lambda_{1x,y}(\omega')F_{x,y}(\omega',z) + i\Lambda_{2x,y}(\omega')e^{8iqz/2}F_{x,y}^{\bullet}(2\omega - \omega',z),$$
(5.7)

where the coefficients $\Lambda_{1,2X,V}(\omega')$ are given by

$$\begin{split} \Lambda_{ix}(\omega') &= -\frac{\omega - \omega'}{c} + q\epsilon \frac{(\Xi + 1)^2}{\Xi^2(\omega_s - \omega')} - q\epsilon \frac{(\Xi - 1)^2}{\Xi^2(\omega_s - \omega')}, \\ \Lambda_{iy}(\omega') &= -\frac{\omega - \omega'}{c} + \frac{3}{2} q\epsilon \frac{\Xi + 1}{\Xi(\omega_a' - \omega')} - \frac{1}{4} \left[\Lambda_{tx}(\omega') + \frac{\omega - \omega'}{c} \right], \end{split}$$

$$\Lambda_{2x}(\omega') = -4\Lambda_{2y}(\omega') = \frac{4q\omega_0^2|d|^2R_x^2(0)}{3\hbar^2c^2\Xi} \frac{1}{(\omega - \omega')^2 - \epsilon^2\Xi^2}$$
(5.8)

The roots of the characteristic equations (5.7), calculated with the aid of (5.8), show that the components $F_{x,y}$ have regions of exponential gain. For F_x this region, as in the scalar theory, is determined by the rela-

$$(\omega - \omega')^2 < \varepsilon^2 \xi$$

and for the y component of the weak radiation the frequency regions where gain occurs is determined at $\xi \ll 1$ by the inequality

$$(\omega - \omega')^2 < \varepsilon^2 \xi / 4. \tag{5.9}$$

¹D. Grischkowsky, Phys. Rev. Lett. 24, 866 (1970).

³V. M. Arutyunyan, N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, Dokl. Akad. Nauk SSSR 49, 28 (1969).

⁴Yu. M. Kirin, S. G. Rautian, A. E. Semenov, and B. M. Chernoborod,

Zh. Eksp. Teor. Fiz. Pis'ma Red. 11, 340 (1970) [JETP Lett. 11, 226

⁵A. M. Bonch-Bruevich, V. A. Khodovoĭ, and V. V. Khromov, Zh. Eksp. Teor. Fiz. Pis'ma Red. 11, 431 (1970) [JETP Lett. 11, 290 (1970)].

⁶N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, Zh. Eksp. Teor. Fiz. Pis'ma Red. 8, 518 (1968) [JETP Lett. 8, 316 (1968)].

⁷V. M. Arutyunyan, É. G. Kanetsyan, and V. O. Chaltykyan, Zh. Eksp. Teor. Fiz. 59, 195 (1970) [Sov. Phys.-JETP 32, 108 (1971)].

⁸P. P. Sorokin, N. S. Shiren, J. R. Lankard, E. C. Hammond, and T.

G. Kazyaka, Appl. Phys. Lett. 10, 44 (1967). ⁹V. M. Arutyunyan, N. N. Badalyan, V. A. Iradyan, and M. E. Movse-

syan, Zh. Eksp. Teor. Fiz. 60, 62 (1971) [Sov. Phys.-JETP 33, 34 (1971)].

¹⁰A. M. Bonch-Bruevich and V. A. Khodovoĭ, Usp. Fiz. Nauk 93, 71 (1967) [Sov. Phys.-Usp. 10, 637 (1968)].

¹¹A. M. Bonch-Bruevich, N. P. Kostin, V. A. Khodovoĭ, and V. V. Khromov, Zh. Eksp. Teor. Fiz. 56, 144 (1969) [Sov. Phys.-JETP 29, 82

¹²A. I. Alekseev, Doctoral Dissertation, Moscow, 1970; A. I. Alekseev and V. M. Galitskii, Zh. Eksp. Teor. Fiz. 57, 1002 (1969) [Sov. Phys.-JETP 30, 548 (1970)].

¹³M. L. Ter-Mikaelyan and A. O. Melikyan, Zh. Eksp. Teor. Fiz. 58, 285 (1970).

Translated by J. G. Adashko

²V. M. Arutyunyan, N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, Zh. Eksp. Teor. Fiz. 58, 37 (1970) [Sov. Phys.-JETP 31, 22 (1970)].