Influence of stationary electric and magnetic fields on resonant four-wave mixing processes

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(Submitted December 17, 1990)
Zh. Eksp. Teor. Fiz. **99**, 1684–1698 (June 1991)

The effect which stationary electric and magnetic fields coupling the excited states of the working transitions of an atomic subsystem can have on resonant four-wave mixing is investigated. The atom-field density matrix formalism developed for resonant four-wave mixing by Holm, Sargent *et al.* [Phys. Rev. A **31**, 3112 (1985); **31**, 3124 (1985)] is employed. The theory developed in this work is extended to the case of interaction of photons with different polarizations. It is shown that new combination tones and the corresponding spontaneous sources required for a quantum description of four-wave mixing then arise. The nonlinear Faraday effect for the pump wave is investigated in the case of a magnetic field acting on the atomic subsystem.

1. INTRODUCTION

Resonant four-wave mixing (FWM) has been intensely studied both theoretically and experimentally (see, e.g., Ref. 1). In the description of FWM processes it is possible to distinguish two main approaches: the semiclassical treatment, in which the electromagnetic field is described classically and the atomic subsystem is described quantum-mechanically, and a completely quantum-mechanical description. The simplest model of the atomic subsystem is a gas of two-level atoms. FWM was considered in Ref. 2 using this model for arbitrary intensities of the pump wave, in the linear approximation with respect to the field amplitudes of the modes symmetrically detuned from it and implementing the semiclassical description. A quantum theory in the formalism of the atom-photon density matrix developed by Scully and Lamb[3] was constructed by Holm, Sargent *et al.*[4] An analogous description was developed in Ref. 6 using Haken's method[5] and the apparatus of Keldysh's graphical technique for nonequilibrium Green's functions[7] in Ref. 8. The main difference between the quantum and semiclassical approaches has to do with the presence of spontaneous sources in the equations for the fields in the quantum treatment, which, naturally, are absent in the classical description. Note that the theory developed in Refs. 4 and 6 does not take account of reabsorption of radiation and is applicable mainly for optically thin media.[8]

The main parameters in resonant FWM are the intensity of the pump wave and the detuning of its frequency from resonance, which determine the kinetic coefficients in the equations for the conjugate modes.[2,4] The introduction of additional parameters would make it possible to more effectively influence the FWM process. This is important, for example, in problems of squeezed states of magnetic fields,[9] where FWM is a very efficient method.[6,10,11] One means of achieving such a parametric effect is to take advantage of the influence stationary electric and magnetic fields have on FWM processes, which is the subject of this paper. This problem is also of independent interest for questions associated, for example, with the propagation of intense laser radiation in a plasma, etc.

In the description of the atomic subsystem we will restrict ourselves to the three-level approximation, where the two upper levels are coupled by the stationary electric or magnetic field (in the latter case one considers transitions of the form $J=0\rightarrow J=1$, with the magnetic field directed parallel to the direction of propagation of the radiation). We will treat the monochromatic pump wave, which is allowed to be of arbitrary intensity, classically, and we will assume the side modes to be quantized.

2. BASIC EQUATIONS

In the present work we will make use of the description of the atom-photon density matrix[2] developed for FWM in Refs. 4. Some aspects of the problem will be analyzed by means of the Keldysh technique for nonequilibrium Green's functions.[7,8] We represent the Hamiltonian of the system under consideration in the form

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_r+\mathcal{H}_i+\mathcal{H}_c, \tag{1}$$

where the first two terms are the Hamiltonian of the unperturbed atomic subsystem and of the free radiation field, and $\mathcal{H}_i$ is the interaction Hamiltonian of the atoms with the pump wave and the stationary electric (or magnetic) field. The last term in Eq. (1) describes the interaction between the quantized electromagnetic field and the atomic subsystem. The equation of motion for the atom-photon density matrix has the form

$$i\dot{\rho}_{\rm ap}=[\mathcal{H},\rho_{\rm ap}]+i\Gamma(\rho_{\rm ap}), \tag{2}$$

where $\Gamma(\rho_{\rm ap})$ is an operator which describes relaxation processes. Carrying out the convolution in Eq. (2) over the photon variables, we obtain an equation for the atomic density matrix ρ, and, conversely, taking the trace over the atomic states, we have an equation for the photon component of the density matrix, P. Following Refs. 4, we represent the atom-photon density matrix in factored form, $\rho_{\rm ap}=P\rho$. Neglecting effects associated with radiation capture, and also with saturation of the atomic subsystem by the quantized fields (naturally assuming therefore that they are small), we have the following equation for the atomic density matrix:

$$i\dot{\rho}=[\mathcal{H}_0+\mathcal{H}_i,\rho]+i\Gamma(\rho). \tag{3}$$

Assuming that the amplitudes of all the fields vary only slightly during the characteristic lifetime of the atomic subsystem, we can find the stationary solution of Eq. (3) and

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thereupon obtain an equation for the slowly varying photon density operator P. This equation depends on the specific structure of the Hamiltonian $\mathscr{H}$. For example, for the single-mode model of the laser, the equation for the density operator of the radiation has the form[12]

$$\frac{d}{dt}P=-\frac{1}{2}A(a^+aP+Paa^+)+Aa^+Pa$$
$$-\frac{1}{2}C(a^+aP+Paa^+)+CaPa^+, \qquad (4)$$

where a^+ and a are the photon creation and annihilation operators. The coefficients A and C describe, respectively, the arrival (due to linear amplification) and departure of photons (due to radiation losses in the cavity). Expressions for the photon occupation numbers are given by the relation $n=\langle a^+aP\rangle$, and from Eq. (4) we find the equations of motion for the photon occupation numbers: $\dot{n}=\langle a^+a\dot{P}\rangle$.

3. FOUR-WAVE MIXING IN AN ELECTRIC FIELD

The atomic subsystem is depicted schematically in Fig. 1. The two upper close-lying levels (*2* and *3*) are coupled by the stationary electric field with field vector $\mathbf{E}_{st}$ (it is also possible to have collisional mixing between them with frequency ν_c). For definiteness we assume that the field vectors of the stationary electric field and the pump wave are parallel, although in the present problem the final results do not depend on the angle between them. The pump wave induces transitions between the first and second levels. In the case when the atomic subsystem is open ($\Gamma_j\neq 0$), the quantity q_j describes the rate of collisional pumping to the jth level. Employing the rotating-wave approximation, we represent the Hamiltonians $\mathscr{H}_i$ and $\mathscr{H}_c$ in the form

$$\mathscr{H}_i=U_L\mu_{12}\mathbf{E}_L(t)R_{21}^{+}+\mu_{32}\mathbf{E}_{st}R_{23}^{+}+\text{h.c.},$$
$$\mathscr{H}_c=i\hbar g\sum_{j=1,2}(R_{21}^{+}U_ja_j-R_{12}^{-}U_j^{+}a_j^{+}), \qquad (5)$$

where $\mathbf{E}_L(t)=\mathbf{E}_L\exp(i\omega_L t)$ is the field vector of the pump wave, a_j^{+} is the creation operator of the photons with wave vector $\mathbf{k}_j$ and frequency ω_j, μ_{jk} are the matrix elements of the dipole moment. U_j is the spatial mode factor, g is a coupling constant which we assume to be identical for both modes (we assume that the wave synchronism conditions $\mathbf{k}_1+\mathbf{k}_2=2\mathbf{k}_L$, $\omega_1+\omega_2=2\omega_L$ also apply for them), and R_{jk}^{+} are the projection operators onto the states j and k. These latter are a generalization of the Pauli matrices σ^{+} for two-level systems and in the present case have the form of 3×3 matrices. Using these operators, we write the components of the atomic density matrix as follows:

$$\rho_{jk}=\langle\rho R_{jk}^{+}\rangle,\quad \rho_{kj}=\langle\rho R_{kj}^{-}\rangle.$$

Let $\boldsymbol{\rho}$ describe the column of slowly varying components of the density matrix of the atomic subsystem (i.e., we distinguish beforehand the rapidly oscillating factors proportional to ω_L):

$$\boldsymbol{\rho}^{T}=(\rho_{11},\rho_{22},\rho_{33},\rho_{12},\rho_{21},\rho_{32},\rho_{23},\rho_{13},\rho_{31}).$$

Then Eq. (3) has the form

$$i\dot{\boldsymbol{\rho}}=\mathbf{L}\boldsymbol{\rho}+\mathbf{q}, \qquad (6)$$

where $\mathbf{q}^{T}=(q_1,q_2,q_3,0,0,0,0,0,0)$, and the matrix $\mathbf{L}$ has the form

$$\mathbf{L}=\begin{vmatrix}
-i\Gamma_1 & i\gamma & 0 & -V^* & V & 0 & 0 & 0 & 0\\
0 & -i\gamma_2 & i\nu_c & V^* & -V & E^* & -E & 0 & 0\\
0 & i\nu_c & -i\gamma_3 & 0 & 0 & -E^* & E & 0 & 0\\
-V & V & 0 & \Delta_{12}^* & 0 & 0 & 0 & -E & 0\\
V^* & -V^* & 0 & 0 & -\Delta_{12} & 0 & 0 & 0 & E^*\\
0 & E & -E & 0 & 0 & \Delta_{32}^* & i\nu_c & 0 & -V\\
0 & -E^* & E^* & 0 & 0 & i\nu_c & -\Delta_{32} & V^* & 0\\
0 & 0 & 0 & -E^* & 0 & 0 & V & \Delta_{13}^* & 0\\
0 & 0 & 0 & 0 & E & -V^* & 0 & 0 & -\Delta_{13}
\end{vmatrix}, \qquad (7)$$

where

$$V=\mu_{12}\mathbf{E}_L/\hbar,\ E=\mu_{32}\mathbf{E}_{st}/\hbar,\ \gamma_2=\Gamma_2+\gamma+\nu_c,$$
$$\gamma_3=\Gamma_3+\nu_c,\ \Delta_{12}=\omega_L-\omega_{21}+i\gamma_{12},\ \Delta_{13}=\omega_L-\omega_{31}+i\gamma_{13},$$
$$\Delta_{32}=\omega_{32}+i\gamma_{32},\ \gamma_{jk}=(\gamma_j+\gamma_k)/2,\ \omega_{jk}=(\varepsilon_j-\varepsilon_k)/\hbar,$$

ε_j is the eigenenergy of the jth state of the atom, and γ is the rate of radiative relaxation of the transition $2\to 1$. In the case when the atomic subsystem is closed ($q_j=\Gamma_j=0$) Eq. (6) must be supplemented by the condition $\mathrm{Tr}\,\rho=1$.

For the photon density operator in second-order perturbation theory in the coupling constant g we have the following equation:[4]

$$\dot{P}=[A_1(a_1^+Pa_1-Pa_1a_1^+)+B_1(a_1Pa_1^+-a_1^+a_1P)$$
$$+C_1(a_1^+a_2^+P-a_2^+Pa_1^+)$$
$$+D_1(Pa_2^+a_1^+-a_1^+Pa_2^+)+\text{h.c.}]+(1\leftrightarrow 2). \qquad (8)$$

For the photon occupation numbers n_j and combination tone operators $\langle a_1a_2\rangle=\langle a_1a_2P\rangle$ we have equations which are identical in structure with the equations of Refs. 4:

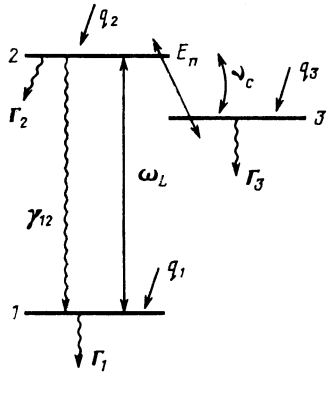

FIG. 1. Diagram of the levels of the atomic subsystem and transitions between them for FWM in a stationary electric field.

$$\frac{d}{dt}n_1=(A_1-B_1-k_p)n_1+(C_1^*-D_1^*)\langle a_1a_2\rangle+A_1+\text{h.c.}, \quad (9a)$$

$$\frac{d}{dt}\langle a_1a_2\rangle=(A_1+A_2-B_1-B_2-2k_c)\langle a_1a_2\rangle$$

$$+(C_1-D_1)n_2+(C_2-D_2)n_1+C_1+C_2. \quad (9b)$$

The coefficient k_c describes the intracavity losses (for the case of intracavity FWM). For the coefficients A_1, B_1, C_1, D_1, we have the following expressions:

$$\begin{aligned}A_1&=Ng^2i(\rho_{21}m_{41}-\rho_{22}m_{44}+\rho_{23}m_{47}),\\ B_1&=-Ng^2i(\rho_{21}m_{42}+\rho_{11}m_{44}+\rho_{32}m_{48}),\\ C_1&=-Ng^2i(\rho_{12}m_{41}+\rho_{22}m_{45}+\rho_{32}m_{49}),\\ D_1&=Ng^2i(\rho_{12}m_{24}-\rho_{11}m_{45}-\rho_{13}m_{47}),\end{aligned} \quad (10)$$

where ρ_{jk} is the stationary solution of system (6); $m_{jk}=\det M_{jk}/\det \mathbf{M}$, and M_{jk} is the minor of the matrix $\mathbf{M}=\mathbf{L}+\nu\mathbf{E}$ (here $\mathbf{E}$ is the identity matrix), $\nu=\omega_1-\omega_L$, and N is the number of atoms. We obtain the corresponding coefficients with index 2 from these expressions by making the substitution $\nu\to-\nu$ and taking the complex conjugate. The quantity $A_1+A_1^*$, which is the spontaneous source in the equations for the photon occupation number n_1, describes the resonant fluorescence spectrum, which was first obtained for an open two-level system by Rautian and Sobel'man,[13] and for a two-level atom by Mollow[14] (these results are contained in our expressions if one sets $\mathbf{E}_{st}$ and ν_c equal to zero). Analogously, the inhomogeneous term in Eq. (9b) C_1+C_2 is the "spontaneous" source for the quantum combination tone $\langle a_2a_1\rangle$. As was shown in Ref. 10, this quantity determines the squeezing of the fields in resonant FWM. The coefficients $B_1-A_1+\text{c.c.}$ and C_1-D_1 describe absorption (amplification) of the photons and four-wave mode coupling, respectively.

The resonant fluorescence spectrum of two-level systems in a strong electromagnetic field is a triplet of Lorentzians,[12,13] in which the side components are shifted relative to the central component by the generalized Rabi frequency $\Omega_R=[(\omega_L-\omega_0)^2+4V^2]^{1/2}$. The effect of a stationary electric field on the resonant fluorescence spectrum (more precisely, its unshifted component) in the three-level system which we are considering is shown in Fig. 2. For comparison with the spectra of a two-level atom, we make the lower level in the calculations for Figs. 2 and 4 the fundamental: $\nu_c=\Gamma_2=0$, $\Gamma_3=\gamma$, $q_2=q_3=0$, $q_1=i\gamma\rho_{33}$.

It is well known that the resonant fluorescence spectrum can be interpreted as resulting from transitions between the quasi-energy levels that are formed as a result of the action of the electromagnetic wave on the atomic subsystem (states of the "dressed" atom). The quasi-energy spectrum in the system under consideration here can be easily obtained from expressions for the poles of the retarded Green's functions. Implementing the approach developed in Ref. 8 using the Keldysh graphical technique, we obtain the following expressions for the retarded Green's functions:

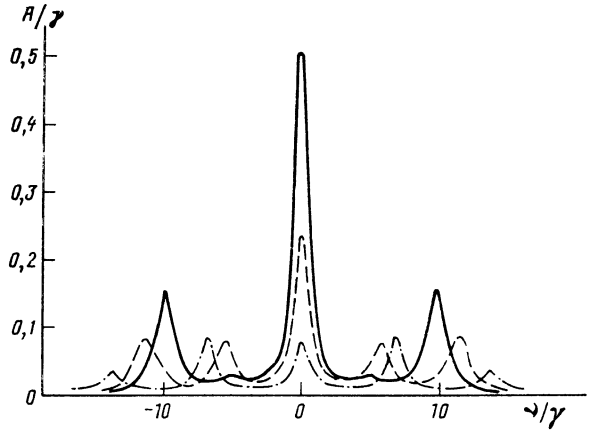

FIG. 2. Effect of a stationary electric field on the resonant fluorescence spectrum for $V/\gamma=5$, $\omega_L=\omega_{21}$, and $E/\gamma=1$ (solid curve), 2 (dashed curve), and 3 (dot-dashed curve).

$$\begin{aligned}G_{11}^r(\varepsilon)&=[\alpha_2(\varepsilon)\alpha_3(\varepsilon)-|E|^2]/S(\varepsilon),\\ G_{22}^r(\varepsilon+\omega_L)&=\alpha_1(\varepsilon)\alpha_3(\varepsilon)/S(\varepsilon),\\ G_{33}^r(\varepsilon+\omega_L)&=[\alpha_1(\varepsilon)\alpha_2(\varepsilon)-|V|^2]/S(\varepsilon),\end{aligned} \quad (11)$$

where

$$\alpha_1(\varepsilon)=\varepsilon,\ \alpha_2(\varepsilon)=\varepsilon+\Delta_{12},\ \alpha_3(\varepsilon)=\varepsilon+\Delta_{13},$$
$$S(\varepsilon)=\alpha_1(\varepsilon)\alpha_2(\varepsilon)\alpha_3(\varepsilon)-|V|^2\alpha_3(\varepsilon)-|E|^2\alpha_1(\varepsilon).$$

For $V, E\gg\Delta_{12}, \Delta_{13}$ it is possible to find an approximate solution of the cubic equation

$$S(\varepsilon)=(\varepsilon-\varepsilon_0)(\varepsilon-\varepsilon_-)(\varepsilon-\varepsilon_+)=0,$$

which describes the quasi-energy spectrum of the given system:

$$\varepsilon_0\approx-\Delta_{13}\frac{|V|^2}{W^2}, \quad \varepsilon_\pm\approx\pm W-\frac{\Delta_{12}+\Delta_{13}}{2}+\Delta_{13}\frac{|V|^2}{2W^2},$$
$$W^2=|V|^2+|E|^2. \quad (12)$$

Using the relations (12), it is possible to represent (11) in the form

$$G_{11}^r(\varepsilon)\approx\frac{1}{2W^2}\left(\frac{2|E|^2}{\varepsilon-\varepsilon_0}+\frac{|V|^2}{\varepsilon-\varepsilon_-}+\frac{|V|^2}{\varepsilon-\varepsilon_+}\right),$$
$$G_{22}^r(\varepsilon+\omega_L)\approx\frac{1}{2}\left(\frac{1}{\varepsilon-\varepsilon_-}+\frac{1}{\varepsilon-\varepsilon_+}\right), \quad (13)$$
$$G_{33}^r(\varepsilon+\omega_L)\approx\frac{1}{2W^2}\left(\frac{2|V|^2}{\varepsilon-\varepsilon_0}+\frac{|E|^2}{\varepsilon-\varepsilon_-}+\frac{|E|^2}{\varepsilon-\varepsilon_+}\right).$$

The quasi-energies of the atomic subsystem are represented schematically in Fig. 3. From expressions (13) and Fig. 3 it can be seen that as the strength of the stationary electric field grows the quasi-energy spectrum is redistributed and additional lines appear in the resonant fluorescence spectrum (see Fig. 2).

The photon absorption (gain) coefficient varies significantly. Along with the appearance of new lines in the absorp-

FIG. 3. Diagram of the quasi-energy levels in strong fields. The "intensity" of the middle sublevel of the second level is negligibly small.

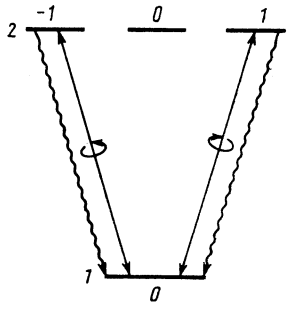

FIG. 5. Diagram of the levels of the atomic subsystem and transitions between them for FWM in a stationary magnetic field.

tion (gain) profile, the action of a stationary electric field results in the removal of saturation between the first and second levels. This causes the photon amplification region to disappear in strong fields (see Fig. 4). Other kinetic coefficients in Eqs. (9) also vary significantly.

Note that the situation of a three-level system in which the parametric equation is brought into being by an intense electromagnetic wave resonant with the transition between the second and third levels is similar to the parametric effect brought about by a stationary electric field, which we are considering here.

Note also that Eq. (9a) can be used in plasma media to describe the transfer of radiation resonant with the lines schematically depicted in Fig. 1.[15] In this case the photons with wave vector $\mathbf{k}_L$ (different from $\mathbf{k}_1$) describe the reabsorption of radiation, and we assume that $|\mathbf{E}_L|^2 \sim n_L \ll 1$. [The term in Eq. (9a) corresponding to FWM here, of course, is absent.] Since the transfer equation should be linear in the occupation numbers (by virtue of their smallness), it is necessary to set V equal to zero in the expressions for the absorption coefficients ($B_1 - A_1$ + c.c.), and it is also necessary to limit the expression for the spontaneous source to linear accuracy in $|V|^2$ with the further substitution $|V|^2 \to \int n_L d\mathbf{k}_L$. It is then necessary in these kinetic coefficients to average over the field strength $\mathbf{E}_{st}$, which plays the role of the instantaneous value of the Stark microfields.[15] The system of equations (9a) and (7) thus obtained is closed, taking account, in particular, of reabsorption of radiation. The expressions for the spontaneous emission in the rescattering function coincide in this case with those obtained in Ref. 15.

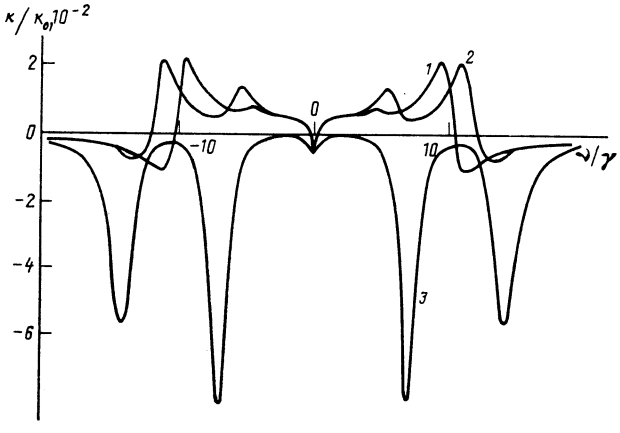

FIG. 4. Coefficient of absorption of the quantized radiation in the presence of a strong laser field and a strong stationary electric field for $V/\gamma = 5, \omega_L = \omega_{21}$, and $E/\gamma = 1$ (curve 1), 3 (curve 2), 5 (curve 3); k_0 is the unsaturated absorption coefficient in the line center.

4. FOUR-WAVE MIXING IN A STATIONARY MAGNETIC FIELD

As the atomic subsystem in this case we use the model of a two-level atom with $J = 0$ in the ground state and $J = 1$ for the upper level. The quantization axis is taken to be aligned with the magnetic field vector $\mathbf{H}$. An electromagnetic field propagates in this direction, so transitions in the atomic subsystem take place only for $\Delta m_J = 1$ (see Fig. 5) and the system is in fact three-level. It is assumed that relaxation in the atoms is of a radiative nature.

An important difference between the FWM processes in a stationary magnetic field and FWM processes in a stationary electric field considered here has to do with the more complicated polarization structure of the radiation in the former case: in this case there are two types of polarization (left and right) for all types of quanta, in contrast with one type in the latter case. This means that FWM can now take place via two channels

$$\mathbf{k}_{1\sigma}+\mathbf{k}_{2\sigma}=2\mathbf{k}_{L\sigma}, \quad \mathbf{k}_{1\sigma}+\mathbf{k}_{2\sigma'}=\mathbf{k}_{L\sigma}+\mathbf{k}_{L\sigma'}$$

(here $\sigma \neq \sigma'$) where σ denotes the type of polarization: left ($-$) or right ($+$); it is natural to assume that $\omega_1 + \omega_2 = \omega_L$. Counterpropagating FWM in an analogous system was studied in Ref. 16. There the semiclassical description was used, and FWM was considered only in one channel, for photons with identical polarization.

The interaction Hamiltonians of the considered system in the rotating wave approximation have the form

$$\mathcal{H}_i = \sum_{\sigma} (U_{L\sigma}\boldsymbol{\mu}_{12\sigma}\mathbf{E}_{L\sigma}R^+_{12\sigma} + \text{h.c.}) - \boldsymbol{\mu}_m \mathbf{H} R_z,$$
$$\mathcal{H}_c = i\hbar g \sum_{\sigma,j} (R^+_{12\sigma}U_{j\sigma}a_{j\sigma} - R^-_{21\sigma}U_{j\sigma}{}^* a^+_{j\sigma}), \quad j = 1, 2. \tag{14}$$

The notation used here is similar to that used in Eqs. (5). In particular, $\mathbf{E}_{L\sigma}(t) = \mathbf{E}_{L\sigma} \cdot \exp(i\omega_L t)$ is the electric field vector of the pump wave with polarization σ, $a_{j\sigma}$ is the creation

operator of a photon with wave vector $\mathbf{k}_{j\sigma}$, and $\boldsymbol{\mu}_m$ is the magnetic moment.

In what follows to simplify the description of the atomic states we will use the following notational conventions: $|2\sigma\rangle = |\sigma\rangle$, $|1\rangle = |0\rangle$. For the projection operators the following commutation relations are fulfilled:

$$[R_3, R_{+0}^+]=2R_{+0}^+, \quad [R_3, R_{0+}^-]=-2R_{0+}^-,$$
$$[R_z, R_{+0}^+]=-R_{+0}^+, \quad [R_z, R_{0+}^-]=R_{0+}^-.$$

The components of the atomic density matrix are written as

$$\rho_{+0}=\langle R_{+0}^+\rho\rangle, \quad \rho_{++}=\langle R_{+0}^+R_{0+}^-\rho\rangle,$$
$$\rho_{+-}=\langle R_{+0}^+R_{0-}^-\rho\rangle, \quad \rho_{00}=\langle R_{0\sigma}^-R_{\sigma 0}^+\rho\rangle.$$

Separating out the rapidly oscillating factors $\sim\exp(\pm i\omega_L t)$, for the components of the atomic density matrix we have

$$i\dot{\boldsymbol{\rho}}=\mathbf{Y}\boldsymbol{\rho}, \tag{15}$$

where

$$\boldsymbol{\rho}^T=(\rho_{++}, \rho_{--}, \rho_{00}, \rho_{0+}, \rho_{+0}, \rho_{0-}, \rho_{-0}, \rho_{+-}, \rho_{-+}),$$

and the matrix $\mathbf{Y}$ has the form

$$\mathbf{Y}=\begin{vmatrix} -i\gamma & 0 & 0 & V & -V^* & 0 & 0 & 0 & 0 \\ 0 & -i\gamma & 0 & 0 & 0 & \overline{V}^* & -\overline{V} & 0 & 0 \\ i\gamma & i\gamma & 0 & -V^* & V & -\overline{V} & \overline{V}^* & 0 & 0 \\ V & 0 & -V & \Delta_+^* & 0 & 0 & 0 & 0 & \overline{V} \\ -V^* & 0 & V^* & 0 & -\Delta_+ & 0 & 0 & -\overline{V}^* & 0 \\ 0 & \overline{V} & -\overline{V}^* & 0 & 0 & \Delta_-^* & 0 & V & 0 \\ 0 & -\overline{V}^* & \overline{V}^* & 0 & 0 & 0 & -\Delta_- & 0 & -V^* \\ 0 & 0 & 0 & 0 & -\overline{V} & V^* & 0 & \delta^* & 0 \\ 0 & 0 & 0 & \overline{V}^* & 0 & 0 & -V & 0 & -\delta \end{vmatrix}, \tag{16}$$

where

$$\Delta_\pm=\omega_L-\omega_0\mp\Omega+i\gamma/2, \quad \delta=2\Omega+i\gamma, \quad V=-\boldsymbol{\mu}_{0+}\mathbf{E}_{L+}/\hbar,$$
$$\overline{V}=-\boldsymbol{\mu}_{0-}\mathbf{E}_{L-}/\hbar, \quad \Omega=\boldsymbol{\mu}_m\mathbf{H}/\hbar,$$

and ω_0 is the frequency of the transition $1\to 2$. Since the atomic subsystem is closed, the total probability is conserved, and the system (15), which is linearly independent, must be supplemented by the condition $\mathrm{Tr}\,\rho = 1$.

The equation of motion for the photon density matrix in the given case has the form

$$\dot{P}=\sum_{j,\sigma,\sigma'}[A_{j\sigma,j\sigma'}(a_{j\sigma}^+Pa_{j\sigma'}-Pa_{j\sigma'}a_{j\sigma}^+)$$
$$+B_{j\sigma,j\sigma'}(a_{j\sigma'}Pa_{j\sigma}^+-a_{j\sigma}^+a_{j\sigma'}P)]$$
$$+\sum_{j\neq j,\sigma,\sigma'}[D_{j\sigma,j'\sigma'}(Pa_{j'\sigma'}^+a_{j\sigma}^+-a_{j'\sigma'}^+Pa_{j\sigma}^+)$$
$$+C_{j\sigma,j'\sigma'}(a_{j\sigma}^+a_{j'\sigma'}^+P-a_{j'\sigma'}^+Pa_{j\sigma}^+)]+\text{h.c.} \tag{17}$$

This equation has a significantly more complicated structure than Eq. (8). This is because FWM, as was noted above, can occur between photons of different polarization. Therefore now along with averages of the form $\langle a_{j\sigma}^+a_{j\sigma}\rangle = n_{j\sigma}$ (the occupation number of photons with wave vector $\mathbf{k}_{j\sigma}$) and $\langle a_{j\sigma}a_{j'\sigma}\rangle$ $(j\neq j')$, which determined the photon dynamics in the previous case, there appear new correlators $\langle a_{j\sigma'}^+a_{j\sigma}\rangle$, $\langle a_{j\sigma}a_{j\sigma'}\rangle$ $(\sigma\neq\sigma')$. The equations of motion for the occupation numbers and the combination tone operators in the present case have the form

$$\frac{dn_{j\sigma}}{dt}=\alpha_{j\sigma}n_{j\sigma}+B_{j\sigma}\langle a_{j\sigma'}^+a_{j\sigma}\rangle+\chi_{j\sigma}\langle a_{j\sigma}a_{j'\sigma}\rangle$$
$$+\xi_{j\sigma}\langle a_{j\sigma}a_{j'\sigma'}\rangle+A_{j\sigma j\sigma}+\text{h.c.},$$

$$\frac{d\langle a_{j\sigma}^\mp a_{j\sigma}\rangle}{dt}=(\alpha_{j\sigma'}+\alpha_{j\sigma}^*)\langle a_{j\sigma'}^+a_{j\sigma}\rangle+\beta_{j\sigma'}n_{j\sigma}+\beta_{j\sigma}^*n_{j\sigma'}+\chi_{j\sigma'}\langle a_{j'\sigma'}a_{j\sigma}\rangle$$
$$+\chi_{j\sigma}^*\langle a_{j\sigma'}^+a_{j'\sigma}^+\rangle+\xi_{j\sigma'}\langle a_{j'\sigma}a_{j\sigma}\rangle+\xi_{j\sigma}^*\langle a_{j\sigma'}^+a_{j'\sigma'}^+\rangle+A_{j\sigma'j\sigma}+A_{j\sigma j\sigma'}, \tag{18}$$

$$\frac{d\langle a_{j\sigma}^+a_{j'\sigma}^+\rangle}{dt}=(\alpha_{j\sigma}+\alpha_{j'\sigma})\langle a_{j\sigma}^+a_{j'\sigma}^+\rangle$$
$$+\beta_{j\sigma}\langle a_{j'\sigma}^+a_{j'\sigma'}^+\rangle+\beta_{j'\sigma}\langle a_{j\sigma}^+a_{j'\sigma'}^+\rangle+\chi_{j\sigma}n_{j'\sigma}$$
$$-\chi_{j'\sigma}n_{j\sigma}+\xi_{j\sigma}\langle a_{j'\sigma'}a_{j'\sigma}\rangle+\xi_{j'\sigma}\langle a_{j\sigma}^+a_{j\sigma'}\rangle+C_{j\sigma j'\sigma}+C_{j'\sigma j\sigma},$$

$$\frac{d\langle a_{j\sigma}^+a_{j'\sigma'}^+\rangle}{dt}=(\alpha_{j\sigma}+\alpha_{j'\sigma'})\langle a_{j\sigma}^+a_{j'\sigma'}^+\rangle$$
$$+\beta_{j\sigma}\langle a_{j\sigma'}^+a_{j'\sigma'}^+\rangle+\beta_{j'\sigma'}\langle a_{j\sigma}^+a_{j'\sigma}^+\rangle$$
$$+\chi_{j\sigma}\langle a_{j'\sigma}a_{j'\sigma'}\rangle+\chi_{j'\sigma'}\langle a_{j\sigma}^+a_{j\sigma'}\rangle+\xi_{j\sigma}n_{j'\sigma'}+\xi_{j'\sigma'}n_{j\sigma}+C_{j\sigma j'\sigma}+C_{j'\sigma' j\sigma},$$

where

$$\alpha_{j\sigma}=A_{j\sigma j\sigma}-B_{j\sigma j\sigma}-k_p, \quad \beta_{j\sigma}=A_{j\sigma j\sigma'}-B_{j\sigma j\sigma'},$$
$$\chi_{j\sigma}=C_{j\sigma j'\sigma}-D_{j\sigma j'\sigma}, \quad \xi_{j\sigma}=C_{j\sigma j'\sigma'}-D_{j\sigma j'\sigma'},$$

and it is assumed that $j \neq j'$ and $\sigma \neq \sigma'$. The coefficients A, B, C, D are determined as follows:

$$
\begin{aligned}
A_{1+1+} &= Ng^2 i(z_{43}\rho_{0+} - z_{44}\rho_{++} - z_{46}\rho_{+-}),\\
B_{1+1+} &= Ng^2 i(-z_{44}\rho_{00} + z_{41}\rho_{+-} + z_{49}\rho_{-0}),\\
A_{1+1-} &= Ng^2 i(z_{43}\rho_{-0} - z_{44}\rho_{-+} - z_{46}\rho_{--}),\\
B_{1+1-} &= -Ng^2 i(z_{42}\rho_{-0} + z_{46}\rho_{00} + z_{48}\rho_{+0}),\\
C_{1+2+} &= -Ng^2 i(z_{43}\rho_{0+} + z_{45}\rho_{++} + z_{47}\rho_{-+}),\\
D_{1+2+} &= -Ng^2 i(z_{41}\rho_{0+} + z_{45}\rho_{00} - z_{48}\rho_{0-}),\\
C_{1+2-} &= -Ng^2 i(z_{43}\rho_{0-} + z_{45}\rho_{+-} + z_{47}\rho_{--}),\\
D_{1+2-} &= -Ng^2 i(z_{42}\rho_{0-} + z_{47}\rho_{00} - z_{49}\rho_{0+}),
\end{aligned}
\tag{19}
$$

where ρ_{jk} is the stationary solution of system (16), $z_{jk} = \det Z_{jk}/\det \mathbf{Z}$, and Z_{jk} is the minor of the matrix $\mathbf{Z} = \mathbf{Y} + \nu\mathbf{E}$ (here $\mathbf{E}$ is the identity matrix), and $\nu = \omega_1 - \omega_L$. We obtain the corresponding coefficients for $j = 1$ and $\sigma = (-)$ by making the substitution $\Omega \to -\Omega$, $V \leftrightarrow \bar{V}$, and for $j = 2$ and $\sigma = (+)$, by making the substitution $\nu \to -\nu$ followed by taking the complex conjugate. To obtain the coefficients for $j = 2$ and $\sigma = (-)$, it is necessary to make both of these substitutions.

The coefficients A and C are free terms in Eqs. (18); they have the meaning of spontaneous sources for the photon occupation numbers and quantum combination tones. Since the lower level is the ground state in the atomic subsystem under consideration here, these coefficients have two components in their own spectrum: an unshifted (or elastic) component, which is proportional to $\delta(\nu)$, and a shifted (inelastic) component. The unshifted component is especially important in weak electromagnetic fields ($V \ll \gamma$). Its contribution here is decisive, and in this case FWM has for the most part a degenerate character ($\nu = 0$). We separate out the unshifted component in the spectrum of the spontaneous sources:

$$
\begin{aligned}
A^{\mathrm{el}}_{\sigma\sigma'} &= 2\pi N g^2 \rho_{0\sigma}\rho_{\sigma' 0}\delta(\nu),\\
C^{\mathrm{el}}_{\sigma\sigma'} &= 2\pi N g^2 \rho_{0\sigma}\rho_{0\sigma'}\delta(\nu),
\end{aligned}
\tag{20}
$$

where

$$
A_{\sigma\sigma'} = (A_{j\sigma j\sigma'} + A^{*}_{j\sigma' j\sigma}), \qquad C_{\sigma\sigma'} = (C_{j\sigma j'\sigma} + C_{j'\sigma' j\sigma}),
$$

and A^{el} and A^{inel} are the elastic and inelastic components of the quantity $A = A^{\mathrm{el}} + A^{\mathrm{inel}}$ (here σ and σ' may coincide).

In strong electromagnetic fields ($V \gg \gamma$) the unshifted component, as a result of saturation, turns out to be small and the main contribution to the spectra of the spontaneous sources comes from the inelastic component.

The magnetic field has a substantial effect on the kinetic coefficients [see Eq. (19)] which determine the equations of motion of the photons (18). Figure 6 shows the dependence on the magnitude of the magnetic field of the inelastic components of the resonant fluorescence spectrum for the photons with left polarization (A_{++}) and the "spontaneous" source (A_{+-}) of the combination tone $\langle a_+^+ a_- \rangle$. For simplicity (and in order to compare with the spectra of the two-level atom), we set $V = \bar{V}$ and $\omega_L = \omega_0$ in the calculations.

We calculate, as in the previous case, the quasi-energy spectrum of the atomic subsystem. For the retarded Green's functions $G_{jj}{}^r$ in the energy representation we have the following expressions:

$$
\begin{aligned}
G_{00}{}^r(\varepsilon) &= \alpha_+\alpha_-/s, \quad G_{++}{}^r(\varepsilon+\omega_L) = (\alpha_0\alpha_- - |\bar{V}|^2)/s,\\
G_{--}{}^r(\varepsilon+\omega_L) &= (\alpha_0\alpha_+ - |V|^2)/s,
\end{aligned}
\tag{21}
$$

where

$$
\alpha_0 = \varepsilon, \quad \alpha_\pm = \varepsilon + \Delta_\pm, \quad s(\varepsilon) = \alpha_0\alpha_+\alpha_- - |V|^2\alpha_- - |\bar{V}|^2\alpha_+.
$$

For $V, \bar{V}, \Omega \gg \Delta_\pm$ we find an approximate solution of the cubic equation:[1)]

$$
\begin{aligned}
s(\varepsilon) &= (\varepsilon-\varepsilon_0)(\varepsilon-\varepsilon_+)(\varepsilon-\varepsilon_-) = 0,\\
\varepsilon_0 &= -\Delta\left(1 + \frac{\Omega^2}{3W^2}\right), \quad \varepsilon_\pm = \pm W - \frac{\Delta}{2}\left(1 - \frac{\Omega^2}{3W^2}\right),
\end{aligned}
\tag{22}
$$

where

$$
W = (|V|^2 + |\bar{V}|^2 + \Omega^2)^{1/2}, \quad \Delta = \omega_L - \omega_0 + i\gamma/2.
$$

Utilizing expressions (21), we represent expressions (20) in the form

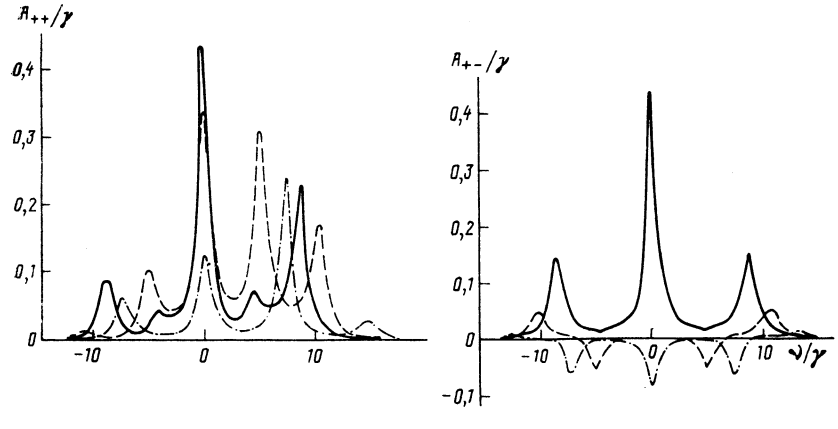

FIG. 6. Resonant fluorescence spectra for photons with left polarization (a) and for the "spontaneous" source (A_{+-}) of the combination tone $\langle a_+^+ a_- \rangle$ (b) for $V/\gamma = 3$ and $\Omega/\gamma = 1$ (solid curve), 3 (dashed curve), and 6 (dot-dashed curve).

$$G_{00}^{r}(\varepsilon)=\frac{1}{2W^2}\left(\frac{2\Omega^2}{\varepsilon-\varepsilon_0}+\frac{|V|^2+|\bar{V}|^2}{\varepsilon-\varepsilon_+}+\frac{|V|^2+|\bar{V}|^2}{\varepsilon-\varepsilon_-}\right),$$

$$G_{++}^{r}(\varepsilon+\omega_L)=\frac{1}{2W^2}\left(\frac{2|\bar{V}|^2}{\varepsilon-\varepsilon_0}+\frac{|V|^2+\Omega^2+\Omega W}{\varepsilon-\varepsilon_+}\right.$$

$$\left.+\frac{|V|^2+\Omega^2-\Omega W}{\varepsilon-\varepsilon_-}\right),$$

$$G_{--}^{r}(\varepsilon+\omega_L)=\frac{1}{2W^2}\left(\frac{2|V|^2}{\varepsilon-\varepsilon_0}+\frac{|\bar{V}|^2+\Omega^2-\Omega W}{\varepsilon-\varepsilon_+}\right.$$

$$\left.+\frac{|\bar{V}|^2+\Omega^2+\Omega W}{\varepsilon-\varepsilon_-}\right). \qquad (23)$$

It is clear from these results that the presence of a magnetic field leads, first of all, to the appearance of five lines in the resonant fluorescence spectrum (in general, seven), and, second, to an asymmetric shape of the radiation spectrum (for prescribed polarization) even for $V=\bar{V}$ (see Fig. 6a).

5. NONLINEAR FARADAY EFFECT

In our consideration of FWM in a magnetic field, we have not allowed for the dispersion properties of the medium, which can have a substantial effect on the propagation of the radiation, for example, in the case of waves with linear polarization. This phenomenon in weak electromagnetic fields ($V\ll\gamma$) has been well studied.[19] In recent years the nonlinear Faraday effect—the rotation of the polarization plane of intensely resonant radiation ($V\gg\gamma$) has been studied both theoretically and experimentally.[20,22]

Let us consider the influence of the dispersion properties of the medium on the propagation of the pump wave and of electromagnetic probe signals with linear polarization. The rotation angle of the polarization plane (RAPP) of the pump wave ($V=\bar{V}$) is given by the expression

$$\varphi=(n_--n_+)l\omega_L/2c, \qquad (24)$$

where l is the linear dimension of the medium, $n_\sigma=1+2\pi N\,\mathrm{Re}(\mu_{\sigma 0}\rho_{0\sigma}/E_{L\sigma})$ is the index of refraction for a wave with polarization σ. Using the stationary solution of Eq. (15), we obtain an expression for φ, valid for arbitrary values of the parameter V/γ:

$$\varphi=\frac{2a|\Omega|}{Q}[(F+2DV^4)(4\Delta_L^2-|\Delta_-|^2-|\Delta_+|^2+2V^2)$$

$$+2V^2(V^4-|\Delta_-|^2|\Delta_+|^2)], \qquad (25)$$

where

$$F=|\Delta_-^*\Delta_++V^2|^2,\quad D=(2V^2+|\Delta_-|^2+|\Delta_+|^2-8\Omega^2)/|\delta|^2,$$
$$\Delta_L=\omega_L-\omega_0,\quad Q=(3V^4+F)[2F+2(2D+1)V^4+V^2(|\Delta_-|^2$$
$$+|\Delta_+|^2)]+3V^2\{(|\Delta_-|^2+|\Delta_+|^2)(F+(2D+1)V^4)$$
$$+2V^2|\Delta_-|^2|\Delta_+|^2\},\quad a=\pi\omega_L lN|\mu|^2/\hbar c.$$

Here $V^2=|V|^2$. A similar result (albeit in still more cumbersome form) was obtained in Ref. 21.

In Fig. 7 it is clear that the shape of the RAPP line strongly depends on the parameter V/γ ($\Omega\gg\gamma$). For $V\gg\Omega$ the RAPP becomes a sign-invariant quantity, while for $V\geqslant 2\Omega$ it acquires a quasi-Lorentzian shape (as a function of the magnitude of the detuning of the radiation from resonance). Figure 8 shows the dependence of the RAPP of intense electromagnetic radiation on the parameter Ω/V at exact resonance ($\omega_L=\omega_0$). In these results the shape of the RAPP line is seen to be in qualitative agreement with the experimental results of Ref. 20.

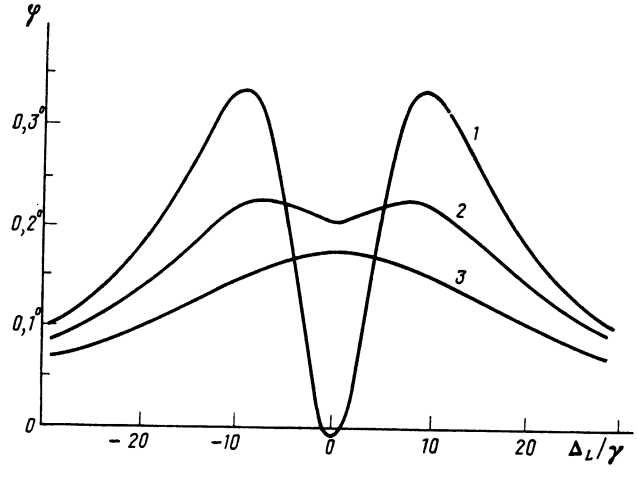

FIG. 7. Dependence of the rotation angle of the polarization plane of a strong electromagnetic wave on its resonance detuning, calculated according to formula (25) for $a=10$, $\Omega/\gamma=5$, and $V/\gamma=5$ (curve *1*), 7 (curve *2*), and 10 (curve *3*).

However, there is a substantial difference in the magnitude of the angle φ shown in Fig. 7 from the results of Ref. 20, consisting of the opposite dependence of φ on V: whereas the RAPP decreases in Fig. 7 as the intensity of the radiation grows, in the results of Ref. 20, on the contrary, it increases. The dependence indicated in Fig. 7 is due to saturation of the atomic subsystem. In the experiment of Buevich *et al.*[20] the thermal motion of the atoms is a significant factor: the Doppler linewidth satisfies $\Delta\omega_D\gg\gamma$ and $\Delta\omega_D\sim\Omega$. Therefore an increase of the intensity of the radiation leads to two effects (besides the nonlinear interference effect, which al-

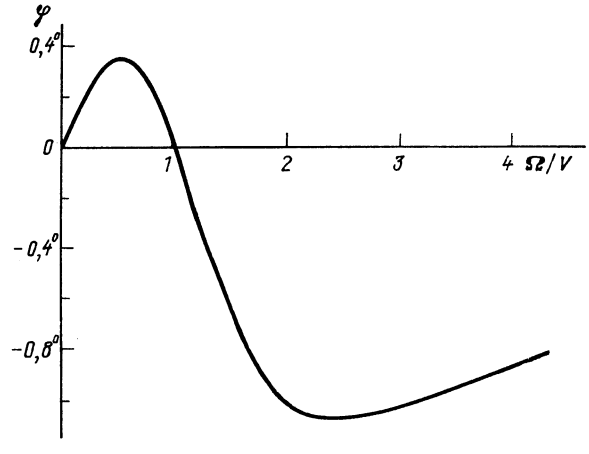

FIG. 8. Rotation angle of the polarization plane of an electromagnetic wave in exact resonance as a function of the parameter Ω/V for $V/\gamma=5$, $a=10$.

ters the shape of the RAPP line of the radiation): saturation of the atomic subsystem and increase in the number of resonant particles (effectively interacting with the radiation) due to field broadening. Since the decrease of φ as a consequence of saturation has a power-law dependence on V, whereas the V-dependence of its increase due to growth of the number of resonant particles is exponential, naturally the overall effect is an increase of the absolute value of φ.

Let us now consider in this same statement of the problem the influence of dispersion on the propagation of the linearly polarized electromagnetic probe signal. In order to avoid FWM with quanta of a different frequency, we assume that the probe signal propagates counter to the strong field. Here the symmetry of the system and the quantization axis is not changed, and to describe the polarizability of the medium at the frequency of the probe field we may use the first four of Eqs. (19). Treating the probe signal as a combination of waves of both left and right polarization, we define the field strength, following Ref. 4, as $\langle a_{j\sigma}\rangle = E_{j\sigma} = \langle a_{j\sigma}P\rangle$. Dropping the index j and making use of Eq. (17), we have the following Maxwell's equations:

$$\left(c^2\frac{\partial^2}{\partial z^2}-\frac{\partial^2}{\partial t^2}\right)\mathbf{E}_+=-4\pi\omega^2(\alpha_+\mathbf{E}_++\beta_+\mathbf{E}_-),$$
$$\left(c^2\frac{\partial^2}{\partial z^2}-\frac{\partial^2}{\partial t^2}\right)\mathbf{E}_-=-4\pi\omega^2(\alpha_-\mathbf{E}_-+\beta_-\mathbf{E}_+). \tag{26}$$

The dispersion equations in this case have the form

$$(\omega^2\varepsilon_+-c^2\mathbf{k}_+^2)(\omega^2\varepsilon_--c^2\mathbf{k}_-^2)-16\pi^2\omega^4\beta_+\beta_-=0, \tag{27}$$

where $\varepsilon_\pm = 1+4\pi\alpha_\pm$. For the wave vectors it is necessary, in order to satisfy the condition of wave synchronism, that we have $\mathbf{k}_+ - \mathbf{k}_- = \mathbf{k}_{L+} - \mathbf{k}_{L-}$. We seek the solution of Eq. (27) in the form

$$\mathbf{k}_+=\omega/c+\varkappa,\quad \mathbf{k}_-=\mathbf{k}_+-\Delta\mathbf{k},\quad \Delta\mathbf{k}=\mathbf{k}_{L+}-\mathbf{k}_{L-}.$$

An approximate solution of Eq. (27) linear in α and β has the form

$$\varkappa_{1,2}=(\bar\alpha_++\bar\alpha_-+\Delta k\pm\zeta)/2,$$
$$\zeta=((\bar\alpha_+-\bar\alpha_--\Delta k)^2+\bar\beta_+\bar\beta_-)^{1/2}, \tag{28}$$

where $\bar\alpha = -2\pi i\omega\alpha/c$. The solution of Eq. (27) with the boundary conditions $\mathbf{E}_\pm(0)=\mathbf{E}_{\pm 0}$ and $|\mathbf{E}_{+0}|=|\mathbf{E}_{-0}| = 2^{-1/2}E_0$ has the form

$$\mathbf{E}_+(z)=\frac{\mathbf{E}_{+0}}{2}\left[e^{i\varkappa_1 z}+e^{i\varkappa_2 z}+\frac{\bar\alpha_+-\bar\alpha_--\Delta k+\bar\beta_+}{\zeta}(e^{i\varkappa_1 z}-e^{i\varkappa_2 z})\right], \tag{29}$$

$$\mathbf{E}_-(z)=\frac{\mathbf{E}_{-0}}{2}\left[e^{i\varkappa_1 z}+e^{i\varkappa_2 z}+\frac{\bar\alpha_--\bar\alpha_++\Delta k+\bar\beta_-}{\zeta}(e^{i\varkappa_1 z}-e^{i\varkappa_2 z})\right].$$

Combining these expressions, we find the field vector of the total field:

$$\mathbf{E}(z)=\frac{E_0}{2}\left[\mathbf{e}_x\left(\cos\frac{\Delta\varkappa z}{2}+i\frac{\bar\beta_++\bar\beta_-}{\zeta}\sin\frac{\Delta\varkappa z}{2}\right)+\mathbf{e}_y\frac{\bar\alpha_--\bar\alpha_+-\Delta k+\bar\beta_--\bar\beta_+}{\zeta}\sin\frac{\Delta\varkappa z}{2}\right]\exp\left[i\left(\frac{\omega}{c}+\frac{\varkappa_1+\varkappa_2}{2}\right)z\right], \tag{30}$$

where $\Delta\varkappa = \varkappa_1 - \varkappa_2$ and $\mathbf{e}_x$ and $\mathbf{e}_y$ are the unit polarization vectors.

It is clear from expression (30) that the polarization structure of the probe signal is changed: it is now elliptically polarized. This is because the FWM coupling constant is large, $\beta\sim\alpha$, and also because the kinetic coefficients are asymmetric, an effect which is caused by the magnetic field even for $V=\bar V$.

In conclusion we note that for FWM in a stationary magnetic field we did not take collisions of the atoms with each other into account, nor with the buffer gas (if there is one in the system). Formally, as in the previous case for FWM in a stationary electric field (i.e., phenomenologically speaking), taking interactions into account is not a source of difficulty. However, by virtue of the idealized nature of the given problem, the value of this procedure is not great. As should be clear, it would be necessary to do this in conjunction with another problem—the consideration of real multilevel atomic subsystems, which is a nontrivial task and requires separate attention. Besides, in the present work we have also not taken account of the thermal motion of the atoms, which, for example, as was noted above, can have a considerable effect on the magnitude of the rotation angle of the polarization plane of the pump wave. In Ref. 8 it was shown that the thermal motion of the atoms can strongly modify not only the absolute values of the kinetic coefficients, but also the line shapes. Therefore this simple (from a physical point of view) kinematic effect is of significant interest and requires additional attention.

In conclusion the author expresses his deep appreciation to A. N. Starostin for his support in this work and helpful discussions.

[1)] The three-level system in a stationary magnetic field and excited by an intense resonant wave considered in the present work is similar to the system which was examined in connection with the Hanle effect[17] in Ref. 18. Equation (22) is practically equivalent to the characteristic equation of Ref. 18, from which it follows that there are seven lines in the spontaneous emission spectrum which can be interpreted as transitions between the quasi-energy levels in a state of the dressed atom. This agrees with the results of the present work. However, the resonant fluorescent spectrum in Ref. 18 was not investigated explicitly, since the main object of study there was the total intensity of the scattered radiation and not the spectral intensity, which is the object of study of the present work.

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Translated by P. F. Schippnick