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Multi-photon processes considering magnetic sublevels coherence

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The exact nonlinear theory of polarized radiation propagation is derived in adiabatic following approximation taking into account different populations and coherence of atom magnetic sublevels. The nonlinear refractive indices for circularly and linearly polarized waves in the resonant media with arbitrary angular moments j_1 and j_2 are obtained. The exact formulas for rotation angle of polarization ellipse axes (without deformation) are found on the sample of medium with $j_1 = 1/2$ and $j_2 = 3/2$. The influence of coherence of magnetic sublevels on multi-photon phenomena and their specific behavior is analyzed.

Keywords: multi-photon processes.

1. Introduction

The propagation of polarized intense laser radiation through a resonant medium is accompanied by specific nonlinear polarization effects, such as atomic sublevels splitting and shift, rotation of the polarization ellipse, *etc*. These phenomena are theoretically studied in different media in [1]–[4]. The general case of two-level system with arbitrary angular moments is considered in [3], [4]. Induced by an intense wave change of the weak probe signal polarization was first observed in alkali metal vapors in [5], [6]. This phenomenon, in particular, becomes the base of the high-resolution spectroscopy [7]. Interaction of quasi-monochromatic pulsed laser radiation with a two-level medium is considered in detail in monograph [8], nonlinear magnetic and optical coherence in coupled two-level systems is investigated in paper [9]. In paper [10], polarization dynamics of femtosecond pulses propagating in air is studied through computer simulation. A rich variety of dynamics that depends on the initial polarization state and power of the pulse is found. Taking into account magnetic sublevels coherence leads to the occurrence of new interference effects, particularly, to the population trapping [11]–[17]. For systems with degenerate levels, the consideration of saturation effects becomes essential since, in consequence of the optical pump of atoms, the saturation of absorption can occur at anomalously small radiation intensities. For studying nonlinear interference phenomena in the field of polarized radiation, the adiabatic following approximation is very promising.

In this paper, the influence of different populations and coherence of magnetic sublevels on the propagation of radiation through the two-level resonant medium is explored. Section 2 discusses the propagation of polarized radiation through the medium with arbitrary angular moments j_1 and j_2. In Sec. 3, the influence of coherence on multi-photon effects is investigated on the sample of medium with $j_1 = 1/2$ and $j_2 = 3/2$.

2. Propagation of polarized radiation through the resonant medium with arbitrary angular moments j_1 and j_2

Let us consider a two-level atom in the field of monochromatic wave specified by the vector potential

$$\mathbf{A} = \mathbf{A}_1(z)\exp\left[i(kz-\omega t)\right] + \mathbf{A}_1^*(z)\exp\left[-i(kz-\omega t)\right]. \tag{1}$$

Later on, it is convenient to turn to the circular components of waves $A_\pm = A_x \pm iA_y$. We assume that the atom has in the ground state an energy E_1 and an angular momentum j_1, while in the excited state – E_2 and j_2, respectively. For an isolated atom these states are degenerate with respect to the projection of the angular momentum.

The Hamiltonian operator $\hat{H}$ of the atom in the field of radiation in dipole approximation is

$$\hat{H} = \hat{H}_0 - \hat{\mathbf{d}}\mathbf{E} \tag{2}$$

where $\hat{H}_0$ denotes the Hamiltonian operator of the isolated atom, $\hat{\mathbf{d}}$ is the electric -dipole-moment operator, $\mathbf{E}$ is the electric-field vector, and

$$\mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}.$$

We find the solution of the Schrödinger equation

$$i\hbar\frac{\partial \Psi}{\partial t} = \hat{H}\Psi \tag{3}$$

in the form

$$\Psi = \sum_{m_1} a(m_1, t)\,\psi_m \exp\left(-\frac{i}{\hbar}E_1 t\right) + \sum_{m_2} b(m_2, t)\,\Phi_{m_2}\exp\left(-\frac{i}{\hbar}E_2 t + i\varepsilon t\right) \tag{4}$$

where ψ_m, Φ_μ are the wave functions of the isolated atom ($m = \pm j_1, \pm(j_1 - 1)$, $\mu = \pm j_2, \pm(j_2 - 1)$), $\varepsilon = (\omega_0 - \omega)$ is the resonance detuning.

The field of radiation (1) takes off the degeneracy of atomic levels and thereby, leads to the occurrence of $m = 2j_1 + 1$ new quasi-stationary wave functions of the system "atom + field". For a circularly polarized wave ($A_1^+ = 0, A_1^- = A$) these wave functions are

$$\Psi_m = \alpha_m \Bigg\{ \psi_m \exp\left(-\frac{i}{\hbar}E_1 t\right) + \frac{i\omega_0 d^* \exp(ikz)}{\varepsilon\lambda c(1 + \sqrt{1 + \xi_m})} A \times \left[\sqrt{\frac{(j_1 - m)(j_1 + m + 1)}{j_1(j_1 + 1)(2j_1 + 1)}} \delta_{j_1, j_2} - \sqrt{\frac{(j_2 + m)(j_2 + m + 1)}{j_2(2j_2 - 1)(2j_2 + 1)}} \delta_{j_2, j_1 + 1} + \sqrt{\frac{(j_1 - m)(j_1 - m - 1)}{j_1(2j_1 - 1)(2j_1 + 1)}} \delta_{j_1, j_2 + 1} \right] \Phi_{m+1} \exp\left(-\frac{i}{\hbar}E_2 t + i\varepsilon t\right) \Bigg\} \exp\left(-\frac{i}{\hbar}\lambda_{1m} t\right) \tag{5}$$

where d is the reduced dipole matrix element, and the following notations are introduced:

$$|\alpha_m|^2 = \frac{1 + \sqrt{1 + \xi_m}}{2\sqrt{1 + \xi_m}},$$

$$\lambda_{1m} = \frac{\hbar\varepsilon}{2}\left(1 - \sqrt{1 + \xi_m}\right),$$

$$\xi_m = \frac{\xi_-}{2j_1 + 1}\left[\frac{(j_1 - m)(j_1 + m + 1)}{j_1(j_1 + 1)} \delta_{j_1, j_2} + \frac{(j_2 + m)(j_2 + m + 1)}{j_2(2j_2 + 1)} \delta_{j_2, j_1 + 1} + \frac{(j_1 - m)(j_1 - m - 1)}{j_1(2j_1 - 1)} \delta_{j_1, j_2 + 1}\right],$$

$$\xi_- = \frac{|d|^2 \left|E_{1-}^2\right|}{\varepsilon^2 \hbar^2}.$$

Assuming that the states Ψ_m are coherent prior to the switching on of the interaction, the wave function of the entire system "atom + field" will be determined in the form

$$\tilde{\Psi} = \sum_m \gamma_m \Psi_m. \tag{6}$$

The coefficients γ_m are found from the initial data.

Substituting the average dipole momentum of the atom $\mathbf{d} = \langle \tilde{\Psi}|\mathbf{d}|\tilde{\Psi}\rangle$ into the Maxwell equation for the slowly varying amplitudes A_{1-}, we find the refractive index of the medium for circularly polarized wave

$$n^{(-)} = 1 + q\frac{c}{\omega_0}\sum_{m=n=-j_1}^{j_1}\rho_{mm}\left[\frac{(j_1+m+1)(j_1-m)}{\sqrt{1+\xi_m}\, j_1(j_1+1)(2j_1+1)}\delta_{j_1,j_2}\right. \tag{7}$$
$$+\frac{(j_2+m)(j_2+m+1)}{\sqrt{1+\xi_m}\, j_2(2j_2-1)(2j_2+1)}\delta_{j_2,j_1+1}$$
$$\left.+\frac{(j_1-m)(j_1-m-1)}{\sqrt{1+\xi_m}\, j_1(2j_1+1)(2j_1-1)}\delta_{j_1,j_2+1}\right]$$

where $q = (\pi|d|^2\omega_0 n)/\varepsilon\hat{}c$ (n is the density of the atoms). We have introduced the density matrix $\rho_{mn} = \gamma_m\gamma_n^*$ of coherent states Ψ_m and Ψ_n^*.

According to the selection rules of angular momentum projections, $d_{z,j_2,m_2}^{j_1,m_1}$ is proportional to δ_{m_1,m_2}, therefore for linearly polarized wave it is convenient to select the x axis along the wave propagation, while the z axis is along the polarization. The similar evaluations lead to the refractive index

$$n_z = 1 + \frac{2qc}{\omega_0}\sum_{m=n=-j_1}^{j_1}\rho_{mn}\left[\frac{m^2}{\sqrt{1+\xi_m}\, j_1(j_1+1)(2j_1+1)}\delta_{j_1,j_2}\right. \tag{8}$$
$$+\frac{j_2^2-m^2}{\sqrt{1+\xi_m}\, j_2(2j_2-1)(2j_2+1)}\delta_{j_2,j_1+1}$$
$$\left.+\frac{j_1^2-m^2}{\sqrt{1+\xi_m}\, j_1(2j_1+1)(2j_1-1)}\delta_{j_1,j_2+1}\right].$$

Comparing expressions (7) and (8) with corresponding formulae in papers [3], [4], one can see that the consideration of non-uniform population of magnetic sublevels modifies the expressions of refractive indices. In particular case, when states Ψ_m are incoherent and uniformly populated, *i.e.*, $\rho_{mn} = \delta_{mn}/(2j_1+1)$, the results obtained coincide with the results of the above-mentioned papers [3], [4].

We will study the influence of non-uniform population of magnetic sublevels coherence on the resonant rotation of polarization ellipse by the example of the medium with angular moments $j_1 = 1/2$ and $j_2 = 3/2$. For this case, it is convenient to represent the density matrix ρ_{mn} through Stock's parameters η_1, η_2, η_3

$$\rho = \frac{1}{2}\begin{bmatrix} 1+\eta_3 & \eta_1 - i\eta_2 \\ \eta_1 + i\eta_2 & 1-\eta_3 \end{bmatrix} \tag{9}$$

where the diagonal element η_3 defines the different populations of magnetic sublevels in the field of the wave, whereas the non-diagonal elements η_1, η_2 characterize the coherence of magnetic sublevels.

The refractive indices for circularly components of elliptically polarized wave $A_\pm$ are:

$$n^+ = 1 + \frac{q_1 c}{\omega_0}\left[\frac{3(1+\eta_3)}{\sqrt{1+\mu_1}} + \frac{1-\eta_3}{\sqrt{1+\mu_2}}\right], \tag{10}$$

$$n^- = 1 + \frac{q_1 c}{\omega_0}\left[\frac{1+\eta_3}{\sqrt{1+\mu_1}} + \frac{3(1-\eta_3)}{\sqrt{1+\mu_2}}\right],$$

$$q_1 = \frac{\pi |d|^2 \omega_0 n}{12 c \hat{\ } \varepsilon},$$

$$\mu_{1,2} = \frac{\omega_0^2 |d|^2}{6 c^2 \hat{\ }^2 \varepsilon^2}\left(3|A^\pm|^2 + |A^\mp|^2\right).$$

Formula (10) describes rotation of the axes of the polarization ellipse (without deformation) through an angle γ, where

$$\gamma = q_1 z\left(\frac{1+\eta_3}{\sqrt{1+\mu_1}} - \frac{1-\eta_3}{\sqrt{1+\mu_2}}\right). \tag{11}$$

For simplicity, let us assume that the atom prior to the interaction was in the ground state 1. If states $\psi_{\pm 1/2}$ are incoherent prior to the switching on of the interaction ($\eta_1 = \eta_2 = \eta_3 = 0$) the rotation angle is

$$\gamma = q_1 z\left(\frac{1}{\sqrt{1+\mu_1}} - \frac{1}{\sqrt{1+\mu_2}}\right). \tag{12}$$

For atoms with initial state $\psi_{+1/2}$ ($\eta_3 = 1$)

$$\gamma = \frac{2q_1 z}{\sqrt{1+\mu_1}}. \tag{13}$$

It follows from (11) that in the general case of coherent states, the linearly polarized light ($\mu_1 = \mu_2 = \mu$) is exposed to polarization plane rotation. Whereas for incoherent states, as is obvious from (12), the linear polarization is not changed through the propagation.

3. Propagation of two waves through the system with $j_1 = 1/2$ and $j_2 = 3/2$

To study multi-photon effects such as Rayleigh scattering, three-photon scattering, parametric four-photon interaction, let us assume that along with the intense monochromatic wave (1) a weak quasi-monochromatic wave $\mathbf{A}_2(z, t)$ propagates in the medium. We have for its potential:

$$\begin{gathered} \mathbf{A} = \mathbf{A}_2(z,t)\exp(-i\omega t) + \mathbf{A}_2^*(z,t)\exp(i\omega t), \\ |\mathbf{A}_2(z,t)| \ \mathrm{V}\ |\mathbf{A}_1(z)|. \end{gathered} \tag{14}$$

It is convenient for further consideration to expand the field $\mathbf{A}_2(z, t)$ to the Fourier integral

$$\mathbf{A}_2(z,t) = \int_{-\infty}^{+\infty} \mathbf{F}(z,\omega')\exp\left[i(\omega-\omega')t\right]\mathrm{d}\omega' \tag{15}$$

where ω' is the carrier frequency of weak quasi-monochromatic wave.

In order to exclude four-wave parametric interaction that takes place when the intense and weak waves propagate in the same direction, let us first assume that they propagate in opposite directions. The equations of propagation for slowly varying amplitudes of $\mathbf{F}(z, \omega')$ in the field of strong linearly polarized along the x axes ($\mu_1 = \mu_2 = \mu$), wave (1) are:

$$\begin{aligned} -\frac{\partial F_x(\omega')}{\partial z} + i\frac{\omega-\omega'}{c}F_x(\omega') &= iD_1\left[F_x(\omega') - \frac{\eta_1}{2i}F_y(\omega')\right], \\ -\frac{\partial F_y(\omega')}{\partial z} + i\frac{\omega-\omega'}{c}F_y(\omega') &= i\frac{D_1}{4}\left[F_y(\omega') - \frac{2\eta_1}{i}F_x(\omega')\right], \end{aligned} \tag{16}$$

where

$$D_1 = \frac{q_1\varepsilon\left(\sqrt{1+\mu}+1\right)^2}{1+\mu}\frac{1}{\omega_\alpha-\omega'} - \frac{q_1\varepsilon\left(\sqrt{1+\mu}-1\right)^2}{1+\mu}\frac{1}{\omega_\sigma-\omega'}, \tag{17}$$

$\omega_\alpha = \omega + \varepsilon\sqrt{1+\mu}$ is the pole of Stark shifted one-photon absorption, $\omega_\sigma = \omega - \varepsilon\sqrt{1+\mu}$ is the pole of three-photon scattering process. D_1 and $D_1/4$ characterize the refractive indices of x and y polarization components of weak wave in Eq. (16), the terms $\pm D_1\eta_1/2$ define the energy transfer from one weak wave component to the other one.

By solving the set of Eq. (16), the following solutions are obtained:

$$F_{x,y}(\omega') = \exp\left[\left(-i\frac{5}{8}D_1 + i\frac{\omega-\omega'}{c}z\right)\right]$$
$$\times\left[F_{x,y}^{z=0}(\omega')\cos(rz) \mp i\frac{3}{8}\frac{D_1}{r}F_{x,y}^{z=0}(\omega')\sin(rz) \pm \eta_1\frac{D_1}{2r}F_{y,x}^{z=0}(\omega')\sin(rz)\right], \tag{18}$$
$$r = \frac{D_1}{8}\sqrt{9+16\eta_1^2}\,.$$

It is obvious from expressions obtained that under the action of intense field the energy is transferred periodically from one weak wave component of polarization to the other. This is caused by magnetic levels coherence and disappears when $\eta_1 = 0$.

The equations of propagation for Fourier components $\mathbf{F}(z, \omega')$ of wave (15) in the case when the strong and the weak waves propagate in the same direction (the intense wave is linearly polarized along the axes x, $\mu_1 = \mu_2 = \mu$) are given by

$$\frac{\partial F_x(\omega')}{\partial z} + i\frac{\omega-\omega'}{c}F_x(\omega') = iD_1\left[F_x(\omega') - \frac{\eta_1}{2i}F_y(\omega')\right]$$
$$+ iD_2\exp\left(\frac{8iq_1z}{\sqrt{1+\mu}}\right)\left[F_x^*(2\omega-\omega') + \frac{\eta_1}{2i}F_y^*(2\omega-\omega')\right],$$
$$-\frac{\partial F_y(\omega')}{\partial z} + i\frac{\omega-\omega'}{c}F_y(\omega') = i\frac{D_1}{4}\left[F_y(\omega') - \frac{2\eta_1}{i}F_x(\omega')\right] \tag{19}$$
$$+ i\frac{D_2}{4}\exp\left(\frac{8iq_1z}{\sqrt{1+\mu}}\right)\left[-F_y^*(2\omega-\omega') + \frac{2\eta_1}{i}F_x^*(2\omega-\omega')\right] + iD_3F_y(\omega')$$

where:

$$D_2 = \frac{2q_1\varepsilon^2\mu}{\sqrt{1+\mu}}\frac{1}{(\omega-\omega')^2-\varepsilon^2(1+\mu)},$$

$$D_3 = 3q_1\varepsilon\frac{1+\sqrt{1+\mu}}{2\sqrt{1+\mu}}\frac{1}{\omega'_\alpha-\omega'}, \tag{20}$$

$\omega'_\alpha = \omega + \frac{\varepsilon}{2}\left(\sqrt{1+\mu}+1\right)$ is the pole of Stark shifted Rayleigh elastic scattering.

As is seen from (19), x and y components of weak radiation polarizations are connected in the medium at the frequency ω' as well as $2\omega-\omega'$. This process is due to the non-degenerate four-photon parametric interaction of the waves and is characterized by the nonlinear coefficient D_2. Atom absorbs two photons of x polarization of the intense wave and emits two photons with different polarizations of weak wave transferring from one coherent state to the other.

The coefficient D_3 defines the Rayleigh elastic scattering when an atom absorbs a photon of the strong field polarized along the x axis and emits a photon of the weak field polarized along the y axis. It must be noted that in the case of incoherent magnetic sublevels [1] when $\eta_1 = 0$, this process does not take place in the adiabatic following approximation.

4. Summary

The complicated interferential polarization effects in resonant medium associated with atom magnetic sublevels coherence are found.

From the nonlinear refractive indices for circularly and linearly polarized waves in the resonant media with arbitrary angular moments j_1 and j_2 obtained, it follows that the linearly polarized light undergoes a rotation of the plane of polarization, whereas in the case of uniformly populated and incoherent states, the linear polarization remains unchanged in the process of propagation.

The allowance for different populations of the magnetic sublevels and their coherence as well changes the behavior of multi-photon processes. So, for the counterpropagating waves the common three-photon scattering is accomplished by energy transfer from one polarization component of the weak wave to the other.

For the propagation of intense and weak waves in the same direction, the picture of nondegenerate four photon parametric interaction changes simultaneously with the occurrence of the other process. As distinct from incoherent case, the elastic Rayleigh scattering occurs. The atom absorbs a photon of strong wave with the x polarization and emits a photon of the weak wave with the y polarization transferring from one coherent state to the other.

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