

**NONLINEAR
AND QUANTUM OPTICS**

The Polarization of Two-Level Atom in the Polychromatic Field¹

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Received June 28, 2014

Abstract—The numerical solution for polarization for two-level atom in polyharmonic field has been made. The analytical solution for partial case of symmetrical case relative to the frequency of the homogeneously broadened transition confirmed numerically. The results can be used in the nonlinear comb spectroscopy.

DOI: 10.1134/S0030400X15060028

INTRODUCTION

The goal of the present work is to study the polarization spectrum of two-level system with homogeneously broadened line in a weak polyharmonic light field.

Nowadays comb spectroscopy is a fast developing area of spectroscopy allowing to detect with high sensitivity atomic and molecular lines in the wide spectral range with resolution limited by Doppler broadening for one-photon transitions. The radiation spectrum consists of narrow equidistant peaks (comb spectrum). The main advantage of comb spectroscopy method is the possibility to detect simultaneously all spectral lines. The nonlinear effects are not taken into account in usual comb spectroscopy. We will show that the nonlinear effects play an important role when the Rabi-frequency of each comb component is comparable (or less) with width the value of the relaxation constants levels (lines). Recently we showed [1] that in counter-propagating combs the homogeneously broadened peaks arise on the wide Doppler counter due to multi-photon processes in the absorption spectrum.

ANALYTICAL SOLUTION FOR POLARIZATION OF TWO-LEVEL SYSTEM INTERACTING WITH POLYHARMONIC FIELD

Let's consider two-level atomic system driven by polyharmonic field with $(2K + 1)$ -monochromatic components:

$$E(t) = \frac{1}{2} \left(\left[E_{s0} + \sum_{m=1}^K E_{sm} (e^{im\Delta_s t} + e^{-im\Delta_s t}) \right] e^{i\omega_{s0} t} + \text{c. c.} \right),$$

where $\omega_{s0} = (\omega_{2K+1} + \omega_1)/2$ is the middle (carrier) frequency of polyharmonic field, $\Delta_s = \omega_{j+1} - \omega_j$ —frequency distance between field component, E_{sm} —the amplitude of m -component of field.

The system of differential equations for density matrix elements in the rotation wave approximation for non-moving atom has the form:

$$\begin{aligned} d\rho_{12}/dt &= -(\Gamma + i\delta)\rho_{12} + iV_{21}^* N_{12}, \\ dN_{12}/dt &= \lambda_{12} - \gamma N_{12} - 4 \text{Im}(V_{21}\rho_{12}), \end{aligned} \quad (1)$$

where $\lambda_{12} = \lambda_1 - \lambda_2$ is the difference of pumping to the levels, $V_{21} = -d_{21}E/\hbar$ is the matrix element of interaction energy in dipole approximation; d_{21} —dipole transition momentum, $N_{12} = \rho_{11} - \rho_{22}$ —the detuning of transition frequency from the middle frequency; $\delta = \omega_{s0} - \omega_{21}$ —population difference; $\omega_{21}, \gamma = \gamma_1 = \gamma_2$,

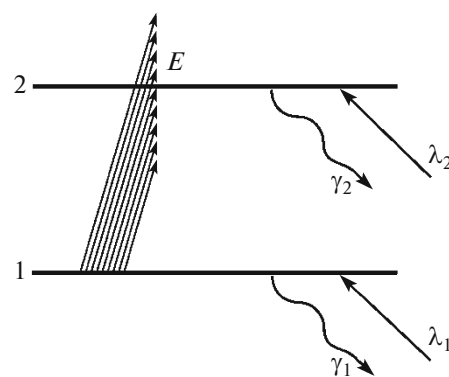


Fig. 1. The scheme of two-level atom interacting field E .

¹ The article was translated by the authors.

Γ —the constants of longitude and transverse relaxations, the widths of level and line (Fig. 1).

The system of differential equations for density matrix has a solution for the special case when the radiation components arranged symmetrically relative to the transition frequency $\delta = 0$ and the relaxation constants equal to each other $\gamma = \Gamma$ [2, 3].

Substituting a new variable $y = N_{12} + i2 \text{Im} \rho_{12}$, the system of equations (1) reduces to a single equation:

$$dy/dt = -(\gamma - i2V_{21})y + \lambda_{12}, \quad (2)$$

where

$$V_{21} = \frac{1}{2} \Omega_{s0} + \sum_{m=1}^K \Omega_{sm} \cos m\Delta_s t, \quad \Omega_{sm} = -d_{21} E_{sm} / \hbar.$$

The solution of equation (2) is known:

$$y(t) = e^{-\int(\gamma - i2V_{21})dt} \left[\int \lambda_{12} e^{\int(\gamma - i2V_{21})dt} dt + \text{const} \right],$$

where

$$\begin{aligned} \text{const} &= 1 - \lambda_{12} \sum_{n_1=-\infty}^{+\infty} \dots \sum_{n_K=-\infty}^{+\infty} \frac{\prod_{m=1}^K J_{n_m}(-Z_{sm})}{\gamma + if_{n_1\dots n_K}}, \\ Z_{sm} &= 2\Omega_{sm} / m\Delta_s, \\ f_{n_1\dots n_K} &= \Omega_{s0} - \sum_{m=1}^K mn_m\Delta_s. \end{aligned}$$

The term const can be neglected, since it is multiplied by $\exp(-\gamma t)$ and for times of the order of 10 becomes small.

Using the equality

$$\exp(\pm iz \sin xt) = \sum_{n=-\infty}^{+\infty} J_n(\pm z) \exp(inxt),$$

get the solution

$$y(t) = \lambda_{12} \sum_{n_1=-\infty}^{+\infty} \dots \sum_{n_K=-\infty}^{+\infty} \sum_{l_1=-\infty}^{+\infty} \dots \sum_{l_K=-\infty}^{+\infty} \frac{B_{n_1\dots n_K l_1\dots l_K} e^{if_{n_1\dots n_K l_1\dots l_K} \Delta_s t}}{(\gamma - if_{n_1\dots n_K})},$$

where

$$\begin{aligned} B_{n_1\dots n_K l_1\dots l_K} &= \prod_{m=1}^K J_{n_m}(-Z_{sm}) J_{l_m}(Z_{sm}), \\ f_{n_1\dots n_K l_1\dots l_K} &= \sum_{m=1}^K m(n_m + l_m). \end{aligned}$$

Find the time dependence of the difference in population levels and the imaginary part of the none-diagonal elements of the density matrix ρ_{12} :

$$\begin{aligned} N_{12}(t) &= \lambda_{12} \sum_{n_1=-\infty}^{+\infty} \dots \sum_{n_K=-\infty}^{+\infty} \sum_{l_1=-\infty}^{+\infty} \dots \\ &\times \sum_{l_K=-\infty}^{+\infty} \left(\frac{B_{n_1\dots n_K l_1\dots l_K} \gamma \cos f_{n_1\dots n_K l_1\dots l_K} \Delta_s t}{\gamma^2 + f_{n_1\dots n_K}^2} \right. \\ &\left. - \frac{B_{n_1\dots n_K l_1\dots l_K} f_{n_1\dots n_K} \sin f_{n_1\dots n_K l_1\dots l_K} \Delta_s t}{\gamma^2 + f_{n_1\dots n_K}^2} \right), \\ \text{Im} \rho_{12}(t) &= \frac{\lambda_{12}}{2} \sum_{n_1=-\infty}^{+\infty} \dots \sum_{n_K=-\infty}^{+\infty} \sum_{l_1=-\infty}^{+\infty} \dots \\ &\times \sum_{l_K=-\infty}^{+\infty} \left(\frac{B_{n_1\dots n_K l_1\dots l_K} \gamma \sin f_{n_1\dots n_K l_1\dots l_K} \Delta_s t}{\gamma^2 + f_{n_1\dots n_K}^2} \right. \\ &\left. + \frac{B_{n_1\dots n_K l_1\dots l_K} f_{n_1\dots n_K} \cos f_{n_1\dots n_K l_1\dots l_K} \Delta_s t}{\gamma^2 + f_{n_1\dots n_K}^2} \right). \end{aligned}$$

The atomic polarization is determined by non-diagonal density matrix elements:

$$P(t) = d_{21} \rho_{12}(t) + \text{c.c.}$$

The polarization components oscillating on the frequencies $\omega_{sj} = \omega_{s0} \pm j\Delta_s$ are given by

$$P_j = -d_{21}/2 \langle i \text{Im} \rho_{12} e^{ij\Delta_s t} \rangle_t,$$

where $\langle \dots \rangle_t$ is an averaging on time.

After averaging on time the real and the imaginary parts of polarization have the forms:

$$\begin{aligned} \text{Re}(P_j) &= \frac{\gamma \lambda_{12} d_{21}}{4} \sum_{n_1=-\infty}^{+\infty} \dots \sum_{n_K=-\infty}^{+\infty} \sum_{l_1=-\infty}^{+\infty} \dots \\ &\times \sum_{l_K=-\infty}^{+\infty} \left(\frac{B_{n_1\dots n_K l_1\dots l_K} \delta_{j, f_{n_1\dots n_K l_1\dots l_K}}}{\gamma^2 + f_{n_1\dots n_K}^2} \right. \\ &\left. - \frac{B_{n_1\dots n_K l_1\dots l_K} \delta_{j, -f_{n_1\dots n_K l_1\dots l_K}}}{\gamma^2 + f_{n_1\dots n_K}^2} \right), \end{aligned} \quad (3)$$

$$\begin{aligned} \text{Im}(P_j) &= -\frac{\lambda_{12} d_{21}}{4} \sum_{n_1=-\infty}^{+\infty} \dots \sum_{n_K=-\infty}^{+\infty} \sum_{l_1=-\infty}^{+\infty} \dots \\ &\times \sum_{l_K=-\infty}^{+\infty} \left(\frac{B_{n_1\dots n_K l_1\dots l_K} f_{n_1\dots n_K} \delta_{j, f_{n_1\dots n_K l_1\dots l_K}}}{\gamma^2 + f_{n_1\dots n_K}^2} \right. \\ &\left. - \frac{B_{n_1\dots n_K l_1\dots l_K} f_{n_1\dots n_K} \delta_{j, -f_{n_1\dots n_K l_1\dots l_K}}}{\gamma^2 + f_{n_1\dots n_K}^2} \right), \end{aligned}$$

where $\delta_{j, \pm f_{n_1\dots n_K l_1\dots l_K}}$ is the Kronecker symbol.

For weak fields and small intermodal distances when Δ_s is less or equal to Ω_{sm} , the imaginary part of polarization has positive meanings for harmonics with number $-4 < j < 4$, where laser field exist. For $|j| > 4$

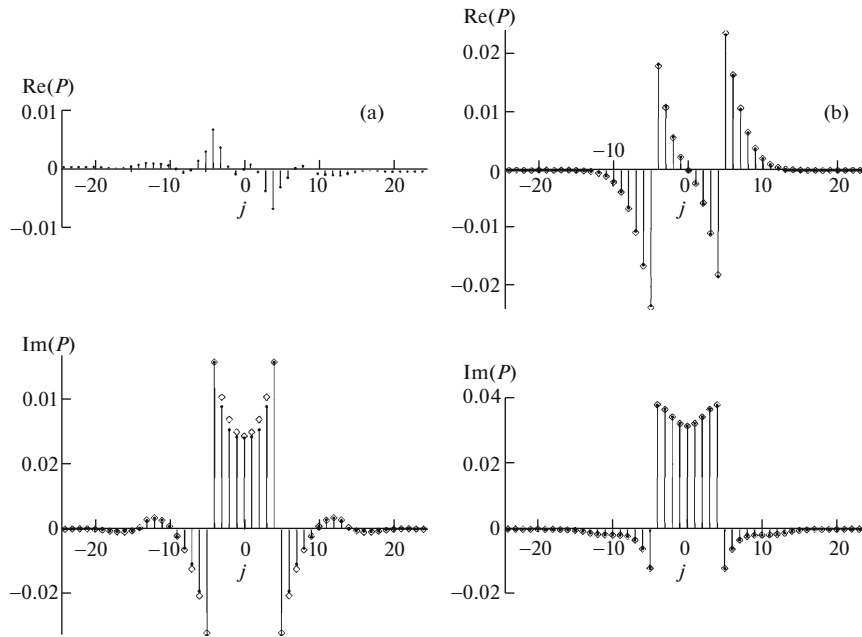


Fig. 2. The real $\text{Re}(P)$ and the imaginary $\text{Im}(P)$ parts of polarization as functions of spectral number j . $K = 4$, $\Omega_{s0} = \Omega_{sm} = 0.2\Gamma$, $\lambda_{12} = 1$, $\delta = 0$, $\Delta_s = 0.01$ (a), $\Delta_s = 0.2$ (b). The diamond points show the results of numerical solutions of the equation system (1), the small round points show the analytical solution (3) of equation (2).

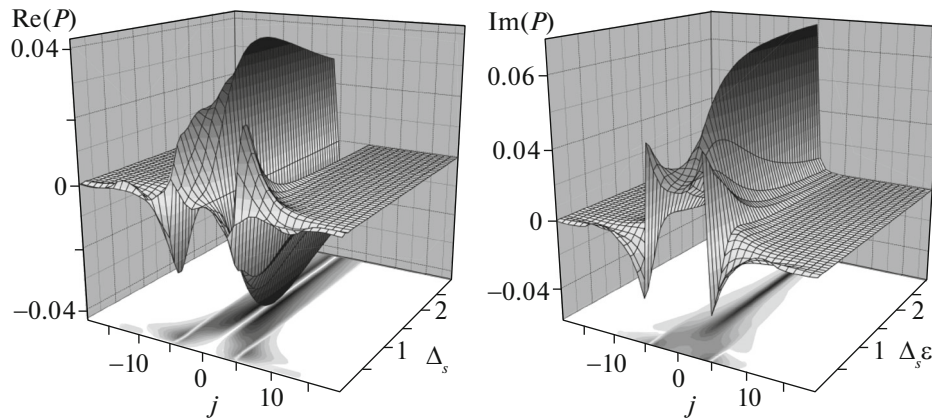


Fig. 3. The dependence of real part of polarization $\text{Re}(P)$ and imaginary part $\text{Im}(P)$ on the number j and inter-mode distance Δ_s . $K = 4$, $\Omega_{s0} = \Omega_{sm} = 0.2\Gamma$, $\lambda_{12} = 1$, $\delta = 0$.

the imaginary part of polarization has negative value, which for $j = 5$ is $3/4$ values for the imaginary part of polarization for $j = 4$ and to order one value for $-2 < j < 2$ (Fig. 2a), i.e. the new components appear at frequencies where there was no harmonics. Despite the fact that there are weak fields, nonlinear processes taking place with the emergence of new harmonics (Fig. 2).

For large inter-mode distances $\Delta_s > \Omega_{sm}$ the dependence of imaginary part of polarization of the harmonic number is in the form of the “bell” (Figs. 3

and 4b) in the case of a homogeneously broadened Lorentzian contour and a weak field near the line of absorption and dispersion relation. The refractive index determined by the real part of the polarization and associated with the imaginary from the Kramers–Kronig has an anomalous appearance near the transition line and normal—away.

The analytical solution are good agreement with calculations based on numerical methods of Runge–Kutta for solving differential equations (1) and then

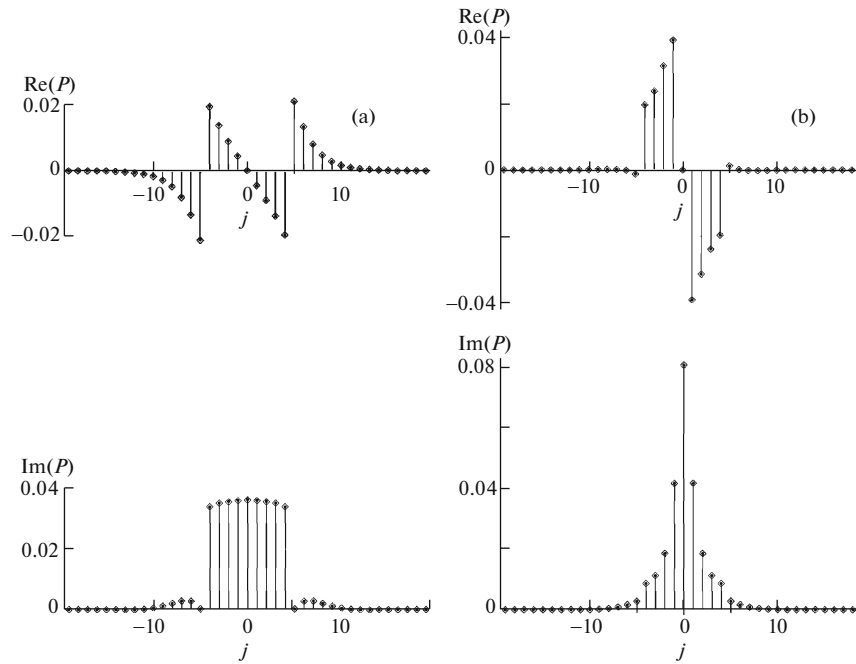


Fig. 4. The real $\text{Re}(P)$ and the imaginary $\text{Im}(P)$ parts of polarization as functions of spectral number j . $K = 4$, $\Omega_{s0} = \Omega_{sm} = 0.2\Gamma$, $\lambda_{12} = 1$, $\delta = 0$, $\Delta_s = 0.3$ (a), $\Delta_s = 1$ (b). The diamond points show the results of numerical solutions of the equation system (1), the small round points show the analytical solution (3) equation (2).

integrating the temporal dependence of the polarization $P(t)$ [3, 4] (Fig. 4).

CONCLUSION

The probability of nonlinear coherent processes is determined by the laser field. If the one strong field acts on medium, the criterion of a strong field is well known. It's ratio of the square of the Rabi frequency Ω_{s0} to the product level width γ and the line width Γ is greater than:

$$\Omega_{s0}^2 / \gamma\Gamma > 1.$$

If the ratio is less than unity, then the non-linear process can be neglected.

The criterion of strong field in polychromatic field associated with the ratio of the square of the Rabi frequency, not only to the level widths (lines), as in the case of a strong field, but also to the frequency (intermode) distance between the components of a polychromatic field. And even with the weak components of the field and a small mode spacing appear nonlinear interaction effects. Phenomenon connected with

mutual influence of the weak field components that are close to each other.

Thus, for certain parameters polychromatic weak field has the same nonlinear effect as strong field. And can be expected splitting of each level on a two-tier system of an infinite system of quasi-energy sublevels [5] and the emergence of multiphoton transitions between them.

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