

## Polarization of two-level atom in a weak polychromatic field

A G Antipov, A A Kalinichev, S A Pulkin, S V Saveleva, A C Sumarov,  
S V Uvarova and V I Yakovleva

Saint Petersburg State University, St.Petersburg, Russia

E-mail: usvik2009@yandex.ru

**Abstract.** Numerical solutions have been obtained for 2-level atom driven by a polychromatic field. Two different methods of analytical solution were proposed for any case of position of the field's components.

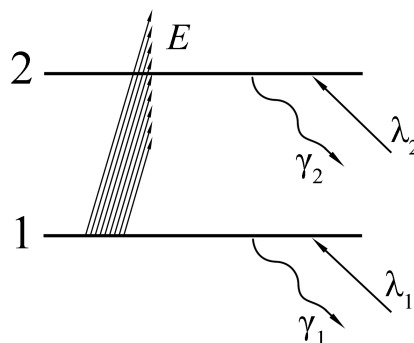
### 1. Introduction

The aim of the present study was to investigate the polarization spectrum of two-level homogeneously broadened atomic system in a weak polychromatic field.

Let the medium of two-level atoms is subjected to the action of quasi-resonant polychromatic field consisted of  $(2M + 1)$  equidistant monochromatic components:

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

where  $\omega_{s0} = (\omega_{2M+1} + \omega_1)/2$  is the median frequency of the polychromatic field,  $\Delta_s = \omega_{j+1} - \omega_j$  is the frequency distance between components and is the amplitude of m-th component (Figure 1).



**Figure 1.** The diagram of two-level atom subjected to the action of polychromatic field  $E(t)$ .

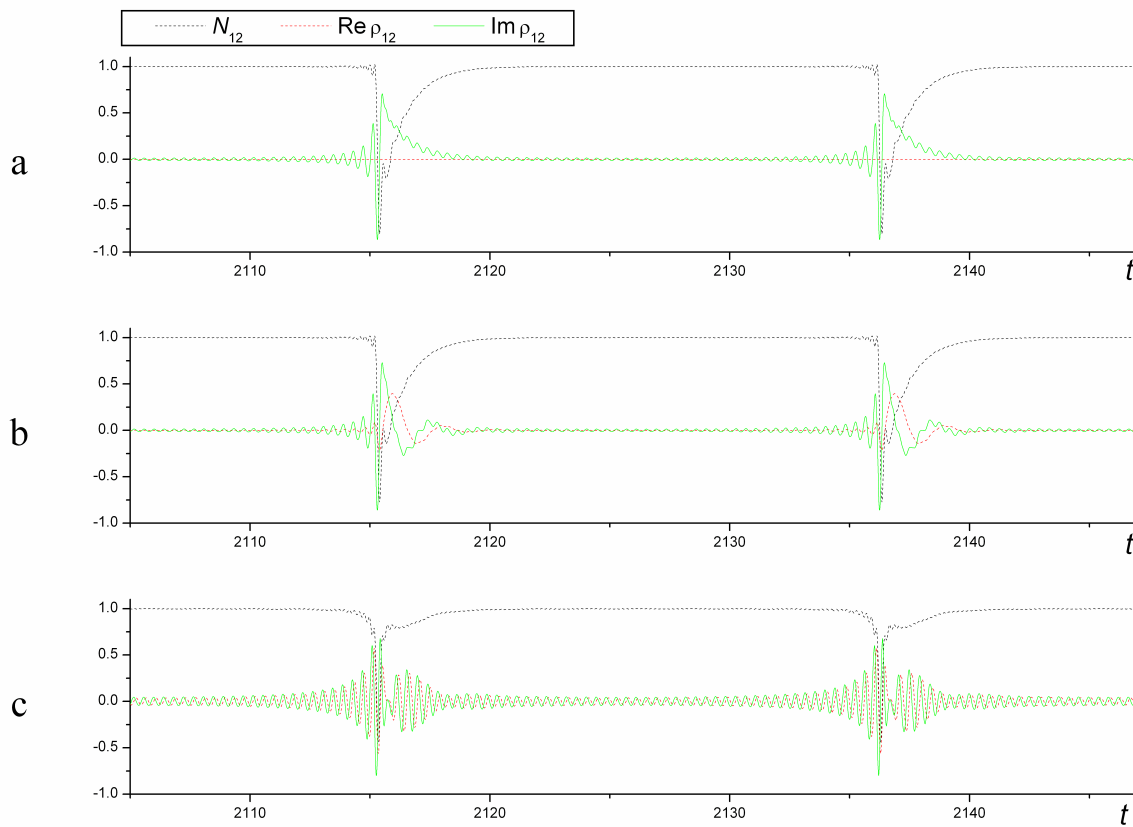
The density matrix equations for two-level system in quasi-resonant rotating wave approximation have the following form:

$$\begin{aligned}\frac{d\rho_{12}}{dt} &= -(\Gamma + i\delta)\rho_{12} + iV_{21}^*N_{12} \\ \frac{dN_{12}}{dt} &= \lambda_{12} - \gamma N_{12} - 4\text{Im}(V_{21}\rho_{12}),\end{aligned}\quad (2)$$

where  $\lambda_{12} = \lambda_1 - \lambda_2$  is the level pump difference,  $V_{21} = \frac{1}{2}\Omega_{s0} + \sum_{m=1}^M \Omega_{sm} \cos m\Delta_s t$  is the matrix element of interacting energy in dipole approximation,  $\Omega_{sm} = -\frac{d_{21}E_{sm}}{\hbar}$  is the Rabi frequency,  $d_{21}$  is the dipole moment of the transition,  $N_{12} = \rho_{11} - \rho_{22}$  is the population difference,  $\delta = \omega_{s0} - \omega_{21}$  is the shift from field median frequency to a transition frequency  $\omega_{21}$ ,  $\gamma = \gamma_1 = \gamma_2$  and  $\Gamma$  are the longitudinal and transverse relaxation constants respectively.

## 2. Computations

The first way to solve differential equation system (2) is to apply to it direct numerical methods. Runge-Kutta method gives the population difference  $N_{12}$  and the non-diagonal element of density matrix as the discrete functions of time (Figure 2). Frequency dependencies can be obtained by Fourier transform.



**Figure 2.** Population difference  $N_{12}$ , real and imaginary parts of non-diagonal density matrix element  $\rho_{12}$  as functions of time at some values of shift  $\delta = \omega_{s0} - \omega_{21}$ .  $M = 50$ ,  $\Delta_s = 0.3\Gamma$ ,

$$\Omega_{sm} = 0.2\Gamma, \quad m = 0, \pm 1, \dots, \pm M, \quad \lambda_{12} = 1; \quad \text{a) } \delta = 0, \quad \text{b) } \delta = 3\Gamma, \quad \text{c) } \delta = 13\Gamma.$$

The second way to solve (2) which is close to Stenholm method of chain ratios [2] is to decompose the unknown functions in harmonic basis.

The system of differential equations (2) simplifies for the special symmetric case of zero shift  $\delta = 0$  and equal relaxation constants  $\gamma = \Gamma$ . Introducing new variable  $y = N_{12} + i2 \operatorname{Im} \rho_{12}$  one can reduce equation system (2) to a single equation:

$$\frac{dy}{dt} = -(\gamma - i2V_{21})y + \lambda_{12}. \quad (3)$$

Substituting into (3) harmonic decomposition  $y(t) = \sum_{j=-\infty}^{+\infty} a_j e^{ij\Delta_s t}$ , we arrive to an infinite

linear equation system for unknown coefficients  $(\gamma + ij\Delta_s)a_j - i \sum_{m=-M}^M \Omega_{sm} a_{j-m} = \lambda \delta_{0,j}$ ,

$j = 0, \pm 1, \pm 2, \dots$ , where  $\delta_{0,j}$  is the Kronecker symbol.

Due to linear growth of respective matrix diagonal elements as  $j$  increases while non-diagonal elements remain the same one can realize a reduction, i.e. to consider finite system of equations instead of infinite one. The values of diagonal elements are defined by  $\Delta_s$ , while the values of non-diagonal ones are defined by  $\Omega_{sm}$ , thus at small ratio  $\Omega_{sm} / \Delta_s$  the effectiveness of reduction increases. While coefficients  $a_j$  are computed, we can obtain the population difference  $N_{12}(t) = \operatorname{Re} y(t)$  and the imaginary part of non-diagonal density matrix element  $\operatorname{Im} \rho_{12}(t) = \operatorname{Im} y(t) / 2$  as functions of time.

The polarization is defined by non-diagonal density matrix elements:

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

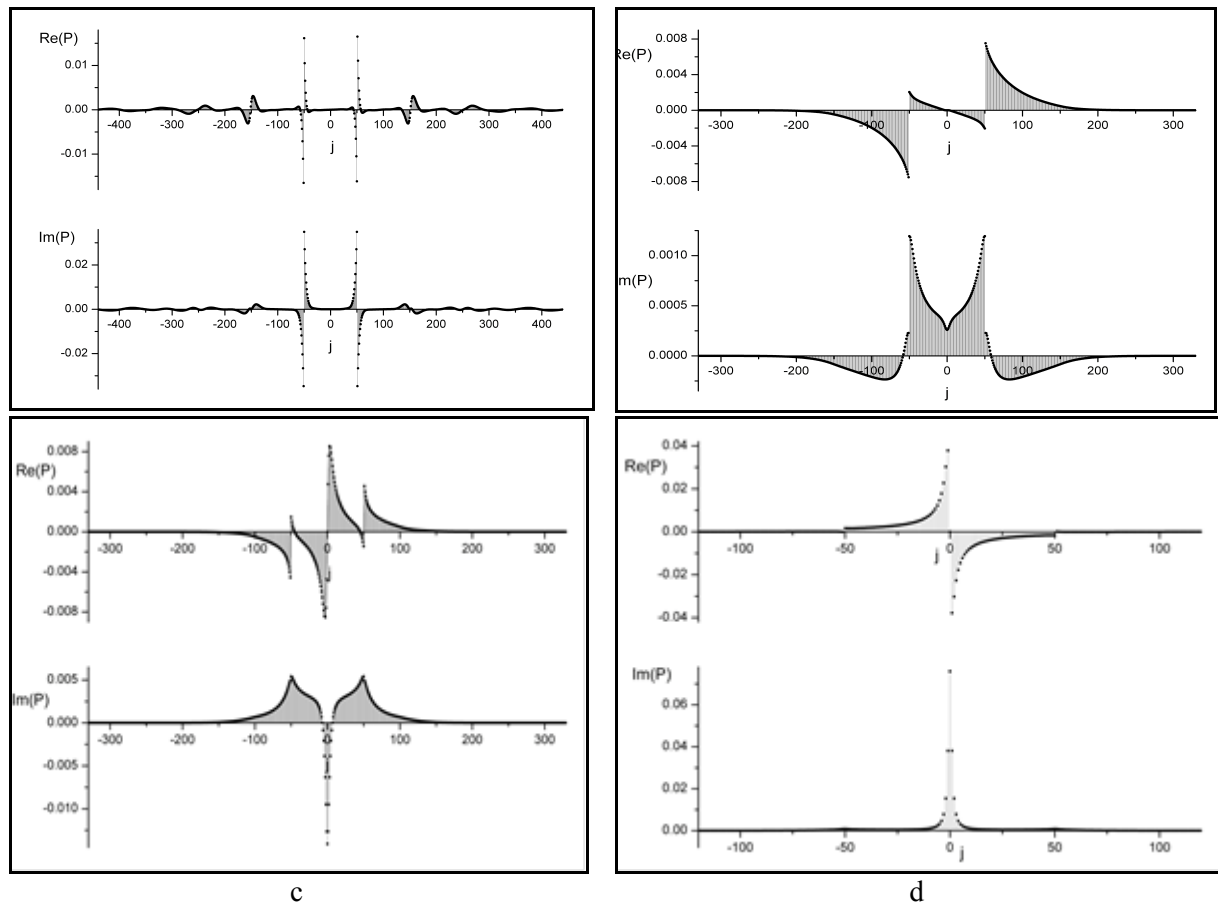
Spectral components of polarization oscillating on frequencies are equal to

$$P_{\pm j} = -\frac{d_{21}}{2} \langle \rho_{12} e^{\pm ij\Delta_s t} \rangle_t. \quad (4)$$

In isotropic medium a direction of polarization vector coincides with the field orientation and the susceptibility is given by

$$\chi_j = \frac{P_j}{E_{sj}} = -\frac{P_j d_{21}}{\Omega_{sj} \hbar}$$

The real and the imaginary parts of susceptibility define the dispersion and the absorption coefficient respectively.



**Figure 3.** The real and the imaginary parts of polarization as functions of component number  $j$  at different distances between components  $\Delta_s$ .  $M = 50$ ,  $\Omega_{sm} = 0.2\Gamma$ ,  $m = 0, \pm 1, \dots, \pm M$ ,  $\lambda_{12} = 1$ ,  $\delta = 0$ ; a)  $\Delta_s = 0.01$ , b)  $\Delta_s = 0.2$ , c)  $\Delta_s = 0.3$ , d)  $\Delta_s = 1$ .

In weak fields with small distances between components where  $\Delta_s$  is less or equal to  $\Omega_{sm}$ , the imaginary part of polarization have positive values for  $-M < j < M$ , therefore these components are absorbed (Figure 3 a, b, c). At  $|j| > M$  the amplitude of imaginary part of polarization for  $M-1$ -th component is negative and its absolute value is almost equal to the absolute value of  $M+1$ -th component, thus new components appear at frequencies where acting field is absent. In spite of the only weak field acts non-linear processes involving new harmonics forming at  $|j| > M$  arise.

At some parameters of atom and field the amplification exists on the transition frequency i.e. the imaginary part of polarization is negative.

At large distances between field components  $\Delta_s > \Omega_{sm}$  the dependence of polarization imaginary part on component number  $j$  looks like a Lorentz contour (Figure 3 d) as in the case of homogeneous broadening and one-component weak field near the absorption line. A dispersion has abnormal characteristics near the transition line and normal characteristics aside of it.

In the common case ( $\delta \neq 0$ ) considering  $N_{12}$ ,  $\text{Re} \rho_{12}$ ,  $\text{Im} \rho_{12}$  in density matrix equations (2) as vectors in harmonic space we obtain the following operator equation for population difference:

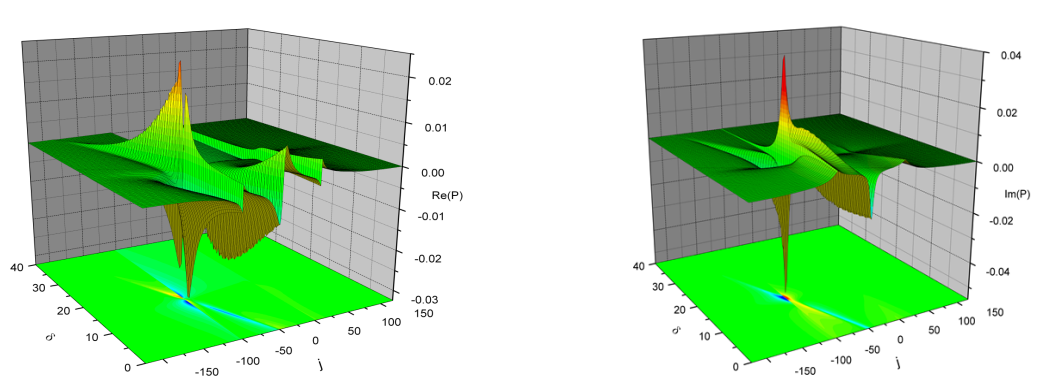
$$\left( \frac{d}{dt} + \gamma + 4V \left( \left( \frac{d}{dt} + \Gamma \right) + \delta^2 \left( \frac{d}{dt} + \Gamma \right)^{-1} \right)^{-1} V \right) N_{12} = \lambda.$$

Using reduction to solve the last equation numerically we can find  $N_{12}$  as decomposition on finite number of harmonics. The imaginary and real parts of non-diagonal element of density matrix are expressed in terms of population difference  $N_{12}$  :

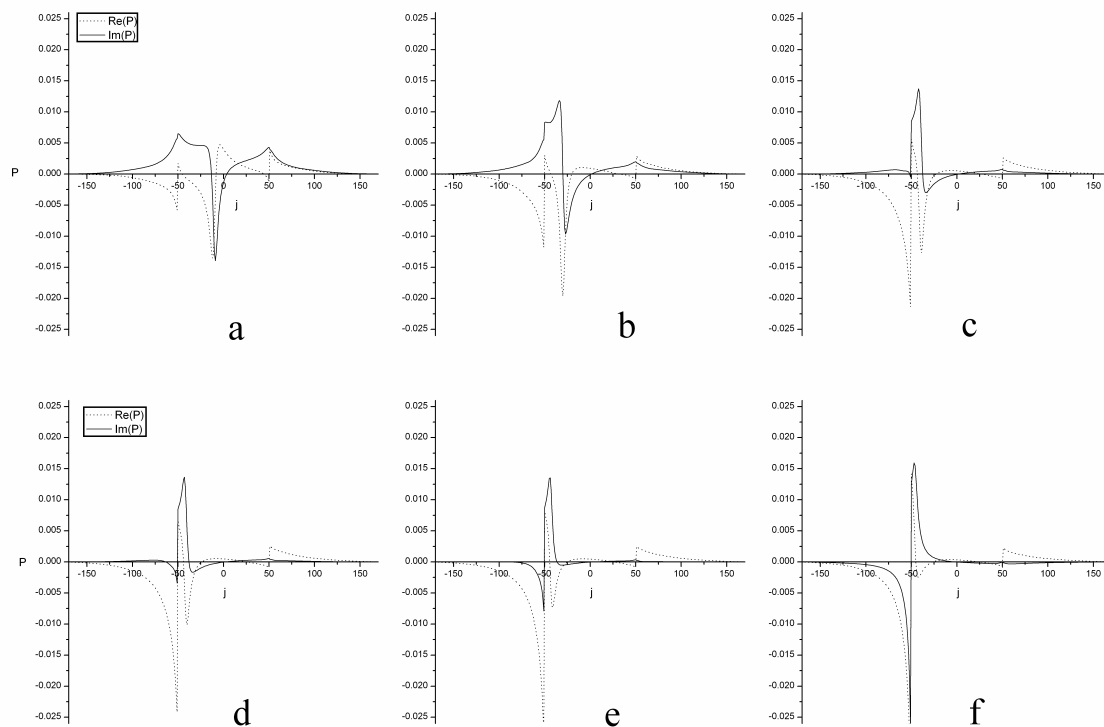
$$\begin{aligned} \text{Im} \rho_{12} &= \left( \left( \frac{d}{dt} + \Gamma \right) + \delta^2 \left( \frac{d}{dt} + \Gamma \right)^{-1} \right)^{-1} V N_{12} \\ \text{Re} \rho_{12} &= \delta \left( \frac{d}{dt} + \Gamma \right)^{-1} \left( \left( \frac{d}{dt} + \Gamma \right) + \delta^2 \left( \frac{d}{dt} + \Gamma \right)^{-1} \right)^{-1} V N_{12} \end{aligned}$$

Polarization can be computed using (4).

Figures 4 and 5 show the evolution of amplification resonances as the frequency shift  $\delta$  changes. A projection on the plane  $\delta 0j$  demonstrates the central resonance ( $\delta = 0, j = 0$ ) follows the transition frequency. The obtained numerical data show that at shifts  $\delta < M\Delta_s = 15$  the resonance is slightly pulled by the median frequency of acting field. At  $\delta > 15$  the resonance on the contrary is slightly pushed away from the transition frequency by the acting field. Near  $\delta = M\Delta_s = 15$  while the left margin of acting field spectrum is about the transition frequency one can see a transformation of one amplification resonance into another with a lesser characteristic frequency.



**Figure 4.** The real  $\text{Re}(P)$  and the imaginary  $\text{Im}(P)$  parts of polarization as functions of component number  $j$  and shift  $\delta$  of transition frequency from the median field frequency.  $M = 50$ ,  $\Delta_s = 0.3\Gamma$ ,  $\Omega_{sm} = 0.2\Gamma$ ,  $m = 0, \pm 1, \dots, \pm M$ ,  $\lambda_{12} = 1$ .



**Figure 5.** The real and the imaginary parts of polarization as functions of component number  $j$  at different shifts  $\delta$ .  $M = 50$ ,  $\Delta_s = 0.3\Gamma$ ,  $\Omega_{sm} = 0.2\Gamma$ ,  $m = 0, \pm 1, \dots, \pm M$ ,  $\lambda_{12} = 1$ ; a)  $\delta = 3$ , b)  $\delta = 9$ , c)  $\delta = 12$ , d)  $\delta = 12.5$ , e)  $\delta = 13$ , f)  $\delta = 14$ .

The results obtained by the both considered methods are in good agreement with each other [3].

### 3. Conclusions

In comb-spectroscopy the nonlinear multi-quantum processes usually are not considered.

We showed that even in weak fields the nonlinear effects take place and this nonlinearity should be taken into account.

When median field frequency does not coincide with the transition frequency the resonance mostly follows the transition frequency. But some frequency pulling or pushing is observed depending on the transition frequency inside the field component bounds or not. This phenomenon can be considered as an evidence of nonlinear interaction of the weak field components.

The results could be used for nonlinear optics and spectroscopy [4].

### References

- [1] Vitushkin L F *et al* 1993 *Optika i spektroskopiya* **74** pp. 786-794
- [2] Stanholm S 1984 *Foundations of laser spectroscopy* (New York: Wiley)
- [3] Antipov A G, Pulkin S A, Sumarov A S, Uvarova S V, Yakovleva V I, *Optics and Spectroscopy* **118** pp. 945-948
- [4] Pulkin S A *et al* 2015 *Russian Journal of Physical Chemistry B* **9** pp. 571-573