

## Polarization Spectrum of Three-Level Atoms in Weak Polychromatic Fields

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**Abstract**—Differential density matrix equations for a three-level atomic system driven by polychromatic fields are solved numerically. The polarization spectrum of the probe field is obtained. Ultra-narrow resonances in the polarization spectrum on the probe field frequency emerge at multiple harmonics and subharmonics.

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### INTRODUCTION

The effect strong polychromatic radiation has on two- and three-level atomic systems has been studied in great detail in recent years [1–16]. Parametric supernarrow resonances (SNRs) in the polarization spectrum at the frequency of probing fields, the nature of which is at first glance analogous to that of the resonances of multiphoton mixing, were obtained numerically in [3, 4]. However, the analytical formula for the absorption coefficient of probing field (3) and condition (4) of the emergence of SNRs, which cannot be explained by mixing the frequencies of other waves for a two-level system was given in [4, 6]. The absorption coefficient of components of the probe field for a two-level atomic system was obtained in three independent ways: numerically [4]; by solving an infinite system of related recurrence relations [2, 6]; and using an analytical formula for the symmetrical arrangement of field components. The derivation and calculations of this formula were described in [4, 9, 10].

A three-level system with lower ground level 1, in which the  $1 \rightarrow 2$  transition occurred upon irradiation by a strong tri-harmonic field and the  $1 \leftrightarrow 3$  transition was initiated by a one- or two-component weak field, was considered in [16]. The absorption spectrum of the components of the probe bi-harmonic field was obtained by numerically solving differential equations for the density matrix and a subsequent Fourier transform of the time dependence of the nondiagonal element of the density matrix; two types of resonances were obtained simultaneously. Resonances of the first type, considered a manifestation of the nonlinear interference effect, (NIEF) were also observed upon scanning by the one-component probe field. Atomic levels 1, 2 under the action of the strong radiation split into a system of quasi-energy sublevels [2].

The structure of sublevels of split lower level 1 was apparent in the absorption spectrum of the probe field (Fig. 2 in [3]) in the case of the strong bi-harmonic or three-mode field. The width of NIEF resonances was comparable to the uniform width of the transition line. Upon the action of the bi-harmonic field on transition  $1 \leftrightarrow 3$ , resonances of the second type (supernarrow) also emerged in the spectrum [3]. The SNR width does not depend on that of the atomic line or those of the levels. A strong multimode field modulates the population of the lower level with the difference frequency of strong field components  $\Delta_S$ , and these oscillations are not damped. SNRs were observed when frequency difference  $\Delta_P$  of the probe field components was a multiple of  $\Delta_S$ . The condition of SNR emergence was obtained from plots:  $\Delta_P = n\Delta_S/2$ ,  $n = 1, 2, \dots$ . In the three-level scheme, the SNRs coincided with the resonances of multiphoton mixing; i.e., the emergence of the parametric resonance on one of the probe field components could be achieved using a combination of the frequencies of several other field components. Unlike spectroscopy of multiphoton interactions, however, the real population of the quasienergy sublevels of split level 1 and atomic level 3 is observed in double resonance spectroscopy, and there is a pumping field (albeit a weak one) on the frequency of the detected field. The absorption coefficient of components of the probe bi-harmonic field is calculated numerically for the system not only in the SNR points but over the frequency intervals of interaction. It describes the energy structure of the levels that is acquired due to the high-frequency Stark effect.

The aim of this work was to obtain numerically the polarization spectrum of a three-level atomic system on the harmonics of the polychromatic field at transition  $1 \leftrightarrow 3$  upon the impact of a weak polychromatic field on transition  $1 \rightarrow 2$ , and to compare it to the

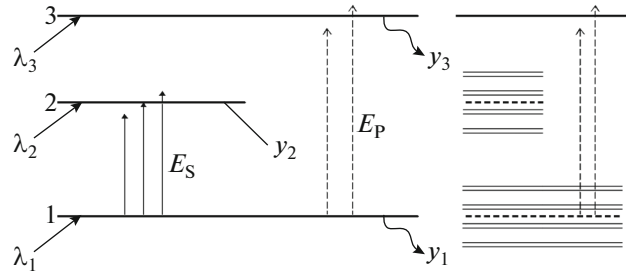


Fig. 1. Scheme of interaction between a three-level atom and a polychromatic field.

results obtained using a second independent analytical approach.

### ANALYTICAL APPROACH

Let us consider a case in which a medium consisting of three-level atoms with common lower level 1

(Fig. 1), transition frequencies  $\omega_{21}$  and  $\omega_{31}$ , dipole moments of transitions  $d_{21}$  and  $d_{31}$ , and longitudinal and transverse relaxation constants  $\gamma$  and  $\Gamma$  is affected upon transitions  $1 \rightarrow 2$  and  $1 \leftrightarrow 3$  by weak equidistant polyharmonic fields  $E_S$  and  $E_P$ :

$$E_S(t) = \frac{1}{2} \left( \left[ E_{s0} + \sum_{m=1}^{M_1} E_{sm} (e^{im\Delta_s t} + e^{-im\Delta_s t}) \right] e^{i\omega_{s0}t} + \text{c.c.} \right),$$

$$E_P(t) = \frac{1}{2} \left( \left[ E_{p0} + \sum_{m=1}^{M_2} E_{pm} (e^{im\Delta_p t} + e^{-im\Delta_p t}) \right] e^{i\omega_{p0}t} + \text{c.c.} \right),$$

where  $\Delta_S = \omega_{sm+1} - \omega_{sm}$  and  $\Delta_P = \omega_{pm+1} - \omega_{pm}$  are the frequency distances between the field components,

and  $\omega_{s0} = \frac{1}{2}(\omega_{sM_1} + \omega_{s-M_1})$  and  $\omega_{p0} = \frac{1}{2}(\omega_{pM_2} + \omega_{p-M_2})$  are the average field frequencies.

To solve the problem, we use the formalism of the density matrix in the fixed-atom and rotating-wave approximations:

$$\frac{d}{dt} \rho_{11} = \lambda_1 - \gamma_1 \rho_{11} - 2 \text{Im}(V_{21} e^{i\delta_{12}t} \rho_{12}) - 2 \text{Im}(V_{31} e^{i\delta_{13}t} \rho_{13}), \quad (1)$$

$$\frac{d}{dt} \rho_{22} = \lambda_2 - \gamma_2 \rho_{22} + 2 \text{Im}(V_{21} e^{i\delta_{12}t} \rho_{12}), \quad (2)$$

$$\frac{d}{dt} \rho_{33} = \lambda_3 - \gamma_3 \rho_{33} + 2 \text{Im}(V_{31} e^{i\delta_{13}t} \rho_{13}), \quad (3)$$

$$\frac{d}{dt} \rho_{12} = -\Gamma_{12} \rho_{12} + i(V_{21}^* e^{i\delta_{12}t} (\rho_{11} - \rho_{22}) - V_{31}^* e^{i\delta_{13}t} \rho_{23}^*), \quad (4)$$

$$\frac{d}{dt} \rho_{13} = -\Gamma_{13} \rho_{13} + i(V_{31}^* e^{i\delta_{13}t} (\rho_{11} - \rho_{33}) - V_{21}^* e^{i\delta_{12}t} \rho_{23}), \quad (5)$$

$$\frac{d}{dt} \rho_{23} = -\Gamma_{23} \rho_{23} - i(V_{21} e^{i\delta_{12}t} \rho_{13} - V_{31}^* e^{i\delta_{13}t} \rho_{12}^*), \quad (6)$$

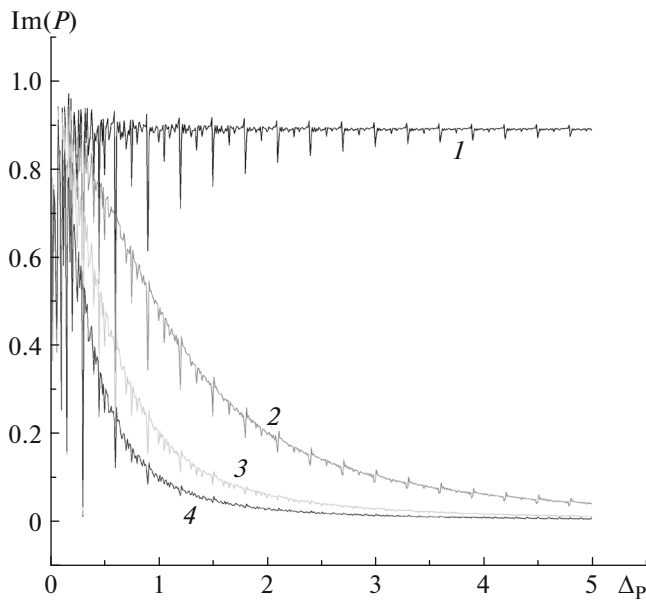
where the matrix elements of the Hamiltonian of the interaction between transitions  $V_{21} = \frac{1}{2} \Omega_{s0} + \sum_{m=1}^{M_1} \Omega_{sm} \cos m\Delta_s t$  and  $V_{31} = \frac{1}{2} \Omega_{p0} + \sum_{m=1}^{M_2} \Omega_{pm} \cos m\Delta_p t$  in dipole approximations  $\Omega_{sm} = -\frac{d_{21} E_{sm}}{\hbar}$  and  $\Omega_{pm} = -\frac{d_{31} E_{pm}}{\hbar}$ ;  $\delta_{12} = \omega_{s0} - \omega_{21}$  and  $\delta_{13} = \omega_{p0} - \omega_{31}$  are the frequencies of the detuning of the average frequencies of the fields from frequencies of the transition; and  $\lambda_j$  is pumping on the  $j$ -th level, where  $j = 1, 2, 3$ .

We solve the system of differential equations (1)–(6) with respect to  $\rho_{13}$  and calculate the polarization of the medium according to the formula

$$P(t) = d_{31} \rho_{13} e^{i\omega_{31}t} + \text{c.c.} \quad (7)$$

The polarization spectrum is found using the Fourier transform of the time dependence of the nondiagonal element of the density matrix  $\rho_{13}$ :

$$P_{\omega_{pj}} = \frac{d_{31}}{T} \int_0^T \rho_{13}^*(t) e^{-i(-1)^j J \Delta_p t} dt, \quad (8)$$



**Fig. 2.** Frequency dependence of the imaginary part of the polarization of a three-level atomic system at the frequency of the harmonics of the probe field on the intermode distance at system parameters  $M_1 = 50$ ,  $\Omega_{sm} = 0.2\Gamma$ ,  $\Omega_{pm} = 0.001\Gamma$ , and  $\Delta_s = 0.3\Gamma$ . The plot number coincides with the number of the harmonic of probe field  $m - 1$ .

where integers  $J = -M_2, -M_2 + 1, \dots, M_2$  and  $T$  is the length of integration.

## RESULTS AND DISCUSSION

The system of differential equations of density matrix (1)–(6) was solved and the polarization spectrum of components of probe field (8) was calculated numerically on a computer. The polarization spectrum of the probe field contained SNRs. We can see from the plots of the dependences of the polarization spectrum of the polyharmonic field on frequency distance  $\Delta_p$  that SNR resonances occur at points of the multiplicity and subcriticality of the intermode distance of the field upon transition  $1 \leftrightarrow 3$  to the frequency distance between components of the equidistant field upon transition  $1 \rightarrow 2$  (Fig. 2):

$$\Delta_p = \frac{g}{r} \Delta_s, \quad (9)$$

where  $g$  and  $r$  are integers.

The analytical solution to the system of differential equations of the density matrix is separated into two stages [17]:

(1) Solving the equations describing the dynamics of a two-level system in a polychromatic field, and calculating the difference between the diagonal elements

of the density matrix using equations in the zero approximation over a weak field.

(2) Calculating the nondiagonal element of the density matrix of three-level systems with allowance for the effect of the probe field using the solutions of the first stage.

The analytical solution in the approximation of the monochromaticity of the field components yields the zero (point) width of super-narrow resonance (SNR).

The conditions in [17] for the emergence of super-narrow resonances, which coincide with conditions (9) obtained from the plots of numerical calculations, are determined.

The authors have written applied programs that allow us to calculate spectral dependences numerically with any parameters of three-level systems and components of equidistant polychromatic fields.

## CONCLUSIONS

Nonlinear multiphoton processes are ignored in linear comb spectroscopy. We showed that at an amplitude of a separate component of a weak field commensurate with the value of the intermode distance of the polychromatic field, the nonlinear effects play a decisive role in the emergence in the spectrum of the polarization of a three-level atomic system at the frequency of a probe field of super-narrow resonances whose width is independent of those of the levels and the atomic transition and is determined by the spectral width of the components of the fields acting on the atoms.

Our results can be used in nonlinear comb spectroscopy inside a spectral line with homogeneous broadening.

## REFERENCES

1. Gaida, L.S. and Pul'kin, S.A., *Opt. Spektrosk.*, 1989, vol. 67, p. 761.
2. Toptygina, G.I. and Fradkin, E.E., *J. Exp. Theor. Phys.*, 1990, vol. 70, p. 428.
3. Vitushkin, L.F., Lazaryuk, S.V., Pul'kin, S.A., et al., *Opt. Spektrosk.*, 1992, vol. 73, p. 880.
4. Vitushkin, L.F., Gaida, L.S., Zeilikovich, I.S., et al., *Bull. Russ. Acad. Sci.: Phys.*, 1992, vol. 56, no. 8, p. 1164.
5. Agarwal, G.S., Zhu, Y., Gauthier, D.J., et al., *J. Opt. Soc. Am. B*, 1991, vol. 8, no. 5, p. 1163.
6. Vitushkin, L.F., Korotkov, V.I., Lazaryuk, S.V., et al., *Opt. Spektrosk.*, 1993, vol. 74, p. 786.
7. Toptygina, G.I. and Fradkin, E.E., *Opt. Spektrosk.*, 1993, vol. 75, p. 228.
8. Lukin, M.D., Fleischhauer, M., Zibrov, A.S., et al., *Phys. Rev. Lett.*, 1997, vol. 79, no. 16, p. 2959.
9. Yoon, T.H., Pulkin, S.A., Park, J.R., et al., *Phys. Rev. A*, 1999, vol. 60, p. 605.

10. Antipov, A.G., Pulkin, S.A., Sumarokov, A.S., Uvarova, S.V., and Yakovleva, V.I., *Opt. Spectrosc.*, 2015, vol. 118, no. 6, p. 945.
11. Antipov, A.G., Kalinichev, A.A., Pulkin, S.A., et al., *J. Phys.: Conf. Ser.*, 2016, vol. 735, p. 012029.
12. Pul'kin, S.A., Savel'eva, M.Yu., Fradkin, E.E., and Uvarova, S.V., *Opt. Spectrosc.*, 2007, vol. 103, no. 6, p. 981.
13. Pulkin, S.A., Yoon, T.H., Kuz'min, A.I., and Uvarova, S.V., *Opt. Spectrosc.*, 2008, vol. 105, no. 2, p. 288.
14. Antipov, A.G., Matveeva, N.I., Pul'kin, S.A., and Uvarova, S.V., *Opt. Spectrosc.*, 2016, vol. 121, p. 879.
15. Sumarokov, A.S., Uvarova, S.V., Antipov, A.G., et al., *Russ. J. Phys. Chem. B*, 2017, vol. 11, no. 1, p. 59.
16. Pul'kin, S.A., Uvarova, S.V., and Fradkin, E.E., *Opt. Spectrosc.*, 2002, vol. 93, no. 2, p. 167.
17. Uvarova, S.V., The study of polarization and susceptibility spectra of atoms in strong light fields, *Cand. Sci. (Phys.–Math.) Dissertation*, St. Petersburg: St. Petersburg State Univ., 2008.

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