

NONLINEAR OPTICAL PHENOMENA

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Saturated two-photon absorption of single-frequency radiation in gaseous media

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Abstract. **A saturated nonresonant two-photon absorption (TPA) theory of quasi-monochromatic radiation by an atom is developed in a model of a three-level cascade quantum system. It is shown that doubled-frequency radiation, arising from a multipole interaction of radiation with the dipole-forbidden transition, experiences spatial oscillations at arbitrary intensities of the pump. The influence of the total phase of the waves with the initial and doubled frequencies on TPA is considered under conditions of ring frequency mixing in a medium.**

Keywords: *two-photon absorption, saturation, frequency doubling, nonlinear dispersion.*

1. Introduction

One of the fundamental effects of nonlinear optics – two-photon absorption (TPA) – is used for excitation of highly excited states of atoms having the same parity as the ground state. Two-photon absorption provides the basis for the development of various methods of nonlinear laser spectroscopy [1, 2]. For these and other TPA applications, optimal are the intensities at which absorption becomes saturated. In atomic gases and metal vapours saturation is reached at moderate intensities: from tens of kW cm^{-2} in the case of rarefield gases to tens of MW cm^{-2} in the case of homogeneous line broadening [3 – 5]. However, the TPA theory, with the exception of the last cited work, is based on allowance for the first nonlinear terms in the series expansion of medium polarisation in powers of the field amplitudes and does not describe saturation [6 – 8].

A three-level model of a medium makes it possible to calculate the polarisation without its series expansion, and thus to take into account the TPA saturation. The TPA saturation is considered in [3] as applied to a quantum Λ-system for the case of exact resonance of monochromatic radiation to the forbidden transition. For many atoms, particularly alkali and alkaline earth metals, the TPA is carried out in the Ξ-system (cascade or ladder) with the ground (s) and intermediate (p) states. This variant of saturated single-frequency TPA is considered in [5, 9], where we have shown theoretically that in an optically dense gaseous media optical rectification appears and the second harmonic is effectively generated. Frequency doubling is due to the multipole (electric quadrupole and magnetic dipole) interaction of radiation with the dipole-forbidden transition. However, in [5, 9] we focused our attention on the mechanisms of these effects and their applications, whereas the saturated TPA properties were not discussed in detail.

These properties, in particular, include nonlinear dispersion of a medium, which plays an important role under conditions of saturation of absorption and ring frequency mixing of absorbed radiation and second harmonic [10, 11]. Nonlinear dispersion determines the phase mismatch of waves in a medium (wave asynchronism), which, in the case of the coherent wave mixing, affects the TPA development in space and frequency doubling. This physical mechanism leads to a qualitative change in the generation of doubled-frequency radiation, namely, to spatial oscillations of its intensity (see Section 4). Frequency doubling is considered in [5] under optimal conditions of complete wave matching; therefore, the evaluation of the efficiency of frequency doubling in a more general case of arbitrary asynchronism requires more precise definitions.

In addition, because of the pump depletion due to the second harmonic generation radiation emitted in the medium will be additionally attenuated, depending on the frequency detuning. This can change the profile of the TPA lines as compared with the case when the multipole interaction is weak and frequency doubling is absent. This aspect of the TPA has not been previously discussed elsewhere.

The aim of this paper is to construct the TPA theory taking into account the above-mentioned physical factors and to analyse the effects of absorption saturation, phase relations and second-harmonic generation, as applied to the profiles of the lines and the effective TPA length. However, we will restrict our consideration to the case of nonresonant TPA and will take a closer look at it in the case of homogeneous line broadening. We will consider second-harmonic generation only in the direction of absorbed radiation, because the account for the backward wave does not introduce any qualitatively new aspects related to the mechanisms discussed [12].

2. Medium polarisation

We denote the ground, intermediate and upper states of the Ξ-system under consideration as 0, 1 and 2, respectively. The equations for the density matrix of the medium in the model of relaxation constants are given by [5]

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$$(A_1+\gamma)\rho_0 + A_1\rho_2 = (A_1+\gamma)\rho_0^{(0)} + A_1\rho_2^{(0)}$$

$$+\frac{2}{\hbar}(d_1\mathcal{E}\,\mathrm{Re}\,\mathrm{i}\rho_{10} + W\mathrm{Re}\,\mathrm{i}\rho_{20}),$$

$$(A_2+\gamma)\rho_2 = (A_2+\gamma)\rho_2^{(0)} - \frac{2}{\hbar}(d_2\mathcal{E}\,\mathrm{Re}\,\mathrm{i}\rho_{21} + W\mathrm{Re}\,\mathrm{i}\rho_{20}),$$

$$-\mathrm{i}\dot{\rho}_{10} + \omega_{10}\rho_{10} = \frac{\mathcal{E}}{\hbar}(d_1 n_{01} + d_2\rho_{20}), \quad (1)$$

$$-\mathrm{i}\dot{\rho}_{21} + \omega_{21}\rho_{21} = \frac{\mathcal{E}}{\hbar}(d_2 n_{12} - d_1\rho_{20}),$$

$$\dot{\rho}_{20} + (\Gamma + \mathrm{i}\omega_{20})\rho_{20} = \frac{\mathrm{i}}{\hbar}[\mathcal{E}(d_2\rho_{10} - d_1\rho_{21}) + W n_{02}];$$

$$n_{jl} = \rho_j - \rho_l\ (j,l = 0,1,2),\ \rho_0+\rho_1+\rho_2 = 1,\ \Gamma = A_2/2 + \Gamma_{\mathrm{c}}.$$

Here, ρ_j is the level populations; ρ_{jl} are the off-diagonal elements of the density matrix, or up to a factor, the polarisations of the transitions $j \leftrightarrow l$; $\rho_{0,2}^{(0)}$ are the equilibrium populations of the levels; A_1 and A_2 are the first Einstein coefficients for the dipole-allowed transitions $0 \leftrightarrow 1$ and $1 \leftrightarrow 2$, respectively; Γ_{c} is the constant of the collision broadening of the dipole-forbidden transition $0 \leftrightarrow 2$; γ is the rate of decay of the excited levels; ω_{jl} are the transition frequencies; d_1 and d_2 are the matrix elements of the dipole moment for dipole-allowed transitions; $\mathcal{E}$ is the electric field amplitude of light in the medium; W is the energy of interaction of the field with a dipole-forbidden transition; $\hbar$ is Planck's constant.

The field in the medium $\mathcal{E}$ and the energy W of its interaction with the $0 \leftrightarrow 2$ transition is expressed in the form [4, 5, 9]

$$\mathcal{E} = E_1(z)\cos\Psi_1 + E_2(z)\cos\Psi_2,\quad W = \mu E_2(z)\cos(\Psi_2+\chi); \quad (2)$$

$$\Psi_1 = \omega t - k_1 z + \varphi_1,\quad \Psi_2 = 2\omega t - k_2 z + \varphi_2,$$

$$\mu = \sqrt{m^2 + k_2^2 Q^2/36},\quad \chi = \arctan\frac{k_2 Q}{9m},$$

where E_1, ω, k_1 and φ_1 are respectively the electric field amplitude, frequency, wave number and phase of the radiation, experiencing the TPA; E_2, k_2 and φ_2 are the electric field amplitude, wave number and phase of the second harmonic generated in the medium; m and Q are the matrix elements of the operators of the magnetic dipole and electric quadrupole moments for the dipole-forbidden transition; z is the longitudinal coordinate.

The solution to equations (1) is expressed as

$$\rho_{10} = \sum_{n=-2}^{2} R_{1n}\exp(-\mathrm{i}n\Psi_1),\quad \rho_{21} = \sum_{n=-2}^{2} R_{2n}\exp(-\mathrm{i}n\Psi_1), \quad (3)$$

$$\rho_{20} = r\exp[-\mathrm{i}(\Psi_2+\chi)].$$

Restricting the summation in (3) by $|n| \leqslant 2$ allows one to take into account the fundamental and doubled frequencies. This approximation is quite sufficient, because the intensities of the third and fourth harmonics are two orders of magnitude smaller than the intensities of the first and second harmonics [5].

Substitution of expressions (2), (3) into equations (1) and use of the rotating-wave approximation [13] lead to steady-state equations whose solution is

$$R_{1-2} = \frac{d_1 E_2\exp(-\mathrm{i}\Theta)}{2\hbar(2\omega+\omega_{10})}n_{01},\quad R_{1-1} = \frac{d_1 E_1}{2\hbar(\omega+\omega_{10})}n_{01},$$

$$R_{10} = \frac{d_2 E_2\exp(-\mathrm{i}\chi)}{2\hbar\omega_{10}}r,\quad R_{11} = \frac{E_1}{2\hbar(\omega_{10}-\omega)}$$

$$\times\{d_1 n_{01} + \exp[\mathrm{i}(\Theta-\chi)]d_2 r\},\quad R_{12} = \frac{d_1 E_2\exp(\mathrm{i}\Theta)}{2\hbar\omega_{21}}n_{01},$$

$$R_{2-2} = \frac{d_2 E_2\exp(-\mathrm{i}\Theta)}{2\hbar(2\omega+\omega_{21})}n_{12},\quad R_{2-1} = \frac{d_2 E_1}{2\hbar(\omega+\omega_{21})}n_{12}, \quad (4)$$

$$R_{20} = -\frac{d_1 E_2\exp(-\mathrm{i}\chi)}{2\hbar\omega_{21}}r,\quad R_{21} = \frac{E_1}{2\hbar(\omega-\omega_{21})}$$

$$\times\{-d_2 n_{12} + \exp[\mathrm{i}(\Theta-\chi)]d_1 r\},\quad R_{22} = -\frac{d_2 E_2\exp(\mathrm{i}\Theta)}{2\hbar\omega_{10}}n_{12};$$

$$r = \frac{\mathrm{i}(1+\mathrm{i}\delta_1)}{D}\{V_2 + \sigma\exp[\mathrm{i}(\Theta-\chi)]V_1\}n_{02}^{(0)},\quad n_{jl}^{(0)} = \rho_j^{(0)} - \rho_l^{(0)},$$

$$n_{01} = n_{01}^{(0)} - \xi_1(2-3\xi_2)\frac{U}{D}n_{02}^{(0)},$$

$$n_{12} = n_{12}^{(0)} + \xi_1(1-3\xi_2)\frac{U}{D}n_{02}^{(0)},$$

$$D = 1 + \delta_1^2 + U,\quad U = \xi_1[V_1^2 + 2\sigma\cos(\Theta-\chi)V_1V_2 + V_2^2],$$

$$\delta_1 = \delta - \beta_1 V_1 + \beta_2 V_2^2,\quad \delta = (2\omega-\omega_{20})/\Gamma,$$

$$V_1 = \frac{d_1 d_2 E_1^2}{4\hbar^2\Gamma\Delta},\ V_2 = \frac{\mu E_2}{2\hbar\Gamma},\ \Delta = |\omega_{10}-\omega| \gg \Gamma,$$

$$\sigma = \mathrm{sign}(\omega_{10}-\omega),\ \beta_1 = \sigma\frac{d_1^2-d_2^2}{d_1 d_2},\ \beta_2 = \frac{\Gamma}{\mu^2}\left(\frac{d_1^2}{\omega_{21}} + \frac{d_2^2}{\omega_{10}}\right),$$

$$\xi_1 = \frac{2\Gamma(2A_1+A_2+2\gamma)}{(A_1+\gamma)(A_2+\gamma)},\ \xi_2 = \frac{A_1+\gamma}{2A_1+A_2+2\gamma},$$

$$\Theta = 2\Psi_1 - \Psi_2 = (k_2-2k_1)z + 2\varphi_1 - \varphi_2.$$

Polarisation of the medium taking into account the interaction of the field with a dipole-forbidden transition is defined as [4, 5]

$$P = 2N\mathrm{Re}[d_1\rho_{10} + d_2\rho_{21} + \mu\exp(\mathrm{i}\chi)\rho_{20}]$$

$$\equiv P_0 + P_{\mathrm{s}1}\sin\Psi_1 + P_{\mathrm{c}1}\cos\Psi_1 + P_{\mathrm{s}2}\sin\Psi_2 + P_{\mathrm{c}2}\cos\Psi_2, \quad (5)$$

where N is the concentration of active atoms. Substituting expressions (3) and (4) into (5) gives

$$P_{s1} = \frac{2Nn_{02}^{(0)} d_1 d_2 E_1}{\hbar D \Delta}\{V_1 + \sigma V_2[\cos(\Theta - \chi) - \delta_1 \sin(\Theta - \chi)]\},$$

$$P_{s2} = \frac{2Nn_{02}^{(0)}\mu}{D}\{V_2 + \sigma V_1[\cos(\Theta - \chi) + \delta_1 \sin(\Theta - \chi)]\},$$

$$P_{c1} = \frac{2NE_1}{\hbar}\left(\frac{d_1^2\omega_{10}}{\omega_{10}^2 - \omega^2}n_{01} - \frac{d_2^2\omega_{21}}{\omega^2 - \omega_{21}^2}n_{12}\right)$$

$$-\frac{2Nn_{02}^{(0)} d_1 d_2 E_1}{\hbar D \Delta}\{\delta_1 V_1 + \sigma V_2[\delta_1\cos(\Theta - \chi) + \sin(\Theta - \chi)]\},$$

$$P_{c2} = -\frac{2NE_2}{\hbar}\left(\frac{d_1^2\omega_{10}}{4\omega^2 - \omega_{10}^2}n_{01} + \frac{d_2^2\omega_{21}}{4\omega^2 - \omega_{21}^2}n_{12}\right) \qquad (6)$$

$$-\frac{2Nn_{02}^{(0)}\mu}{D}\{\delta_1 V_2 + \sigma V_1[\delta_1\cos(\Theta - \chi) - \sin(\Theta - \chi)]\},$$

$$P_0 = \frac{2Nn_{02}^{(0)} d_1 d_2 E_2 \Delta}{\hbar\omega_{10}\omega_{21} D}[V_1(\delta_1\cos\Theta - \sin\Theta)$$

$$+\sigma V_2(\delta_1\cos\chi - \sin\chi)].$$

The constant of the medium polarisation, P_0, produces optical rectification of radiation considered in detail in [5].

3. Wave equations for the fields and the total phase

From equations (7), given in [11], and formulae (6) we obtain a system of coupled equations for the dimensionless intensity of absorbed radiation V_1, the dimensionless field amplitude with a doubled frequency V_2 and a total phase of the waves Θ (4):

$$\frac{dV_1}{d\zeta} = -\frac{V_1}{D}\{V_1 + \sigma V_2[\cos(\Theta - \chi) - \delta_1\sin(\Theta - \chi)]\},$$

$$\frac{dV_2}{d\zeta} = -\frac{\eta}{D}\{V_2 + \sigma V_1[\cos(\Theta - \chi) + \delta_1\sin(\Theta - \chi)]\},$$

$$\frac{d\Theta}{d\zeta} = -\frac{f_1 n_{01} - f_2 n_{12}}{n_{02}^{(0)}} \qquad (7)$$

$$+\frac{1}{D}\{\delta_1 V_1 + \sigma V_2[\delta_1\cos(\Theta - \chi) + \sin(\Theta - \chi)]\}$$

$$-\frac{\eta}{D}\left\{\delta_1 + \sigma\frac{V_1}{V_2}[\delta_1\cos(\Theta - \chi) - \sin(\Theta - \chi)]\right\};$$

$$f_1 = \frac{3\omega^2\omega_{10}d_1\Delta}{(\omega_{10}^2 - \omega^2)(4\omega^2 - \omega_{10}^2)d_2},\ f_2 = \frac{3\omega^2\omega_{21}d_2\Delta}{(\omega^2 - \omega_{21}^2)(4\omega^2 - \omega_{21}^2)d_1},$$

$$\zeta = Gz,\ G = \frac{8\pi Nn_{02}^{(0)} d_1 d_2\omega}{c\hbar\Delta},\ \eta = \frac{\mu^2\Delta}{2\Gamma d_1 d_2},$$

where c is the speed of light.

It follows from the third equation in (7) that the rate of change of phase in a medium is determined by the populations of the levels (the first term in the right-hand side) and the total (interference) interaction of radiation with frequencies ω and 2ω with an atom (the second and third terms, respectively). At low and high radiation intensities the level populations are constant, and the first term is a constant dimensionless wave detuning giving a linear variation of the phase along the length. The second and third terms contain the phase and determine its complex behaviour when radiation propagates in the medium. The presence of $\propto V_2^{-1}$ in the third term determines the phase jumps and positive values of the field amplitude with the doubled frequency [11].

Equations (7) can be generalised to the case of inhomogeneous broadening by making the replacement $\delta \to \delta - k_2 v/\Gamma$ (v is the absorbing molecule velocity projection onto the z axis) and by averaging the right-hand sides of equations (7) over the Maxwell velocity distribution. As a result, we obtain

$$\frac{dV_1}{d\zeta} = -\frac{\sqrt{\pi}\,\Gamma V_1}{k_2\bar{v}\sqrt{1+U}}\{[V_1 + \sigma V_2\cos(\Theta - \chi)]\mathrm{Re}\,w(Z)$$

$$-\sigma V_2\sqrt{1+U}\sin(\Theta - \chi)\,\mathrm{Im}\,w(Z)\},$$

$$\frac{dV_2}{d\zeta} = -\frac{\sqrt{\pi}\,\Gamma\eta}{k_2\bar{v}\sqrt{1+U}}\{[V_2 + \sigma V_1\cos(\Theta - \chi)]\mathrm{Re}\,w(Z)$$

$$+\sigma V_1\sqrt{1+U}\sin(\Theta - \chi)\,\mathrm{Im}\,w(Z)\}, \qquad (8)$$

$$\frac{d\Theta}{d\zeta} = -\frac{f_1 n_{01}^{(0)} - f_2 n_{12}^{(0)}}{n_{02}^{(0)}} + \frac{\sqrt{\pi}\,\Gamma}{k_2\bar{v}\sqrt{1+U}}\Big\{\{[(2 - 3\xi_2)f_1$$

$$+(1 - 3\xi_2)f_2]U + \sigma(\eta V_1 + V_2)\sin(\Theta - \chi)\}\mathrm{Re}\,w(Z)$$

$$-\sqrt{1+U}\left[\eta - V_1 + \sigma\left(\eta\frac{V_1}{V_2} - V_2\right)\cos(\Theta - \chi)\right]\mathrm{Im}\,w(Z)\Big\};$$

$$w(Z) = \frac{i}{\pi}\int_{-\infty}^{\infty}\frac{\exp(-x^2)dx}{Z - x} \approx \sum_{n=1}^{4}\frac{a_n}{b_n + Z},$$

$$Z = \frac{\Gamma}{k_2\bar{v}}(\delta_1 + i\sqrt{1+U}),\ \bar{v} = \sqrt{\frac{2k_B T}{m_a}},\ k_2 \approx \frac{2\omega}{c},$$

where k_B is the Boltzmann constant; T is the temperature; m_a is the mass of the absorbing atoms. The expressions for the complex coefficients a_n and b_n in the approximate representation of the probability integral of the complex argument [14] are given in [15, 16].

Equations (7) and (8) are a generalisation of equations (25) and (26) from [5] to the case when the total phase Θ changes in the process of wave propagation in the medium. To assess the impact of the variable phase on the TPA, we will write equation (7) for the case of the constant phase $\Theta = 2\varphi_1 - \varphi_2 \equiv \Phi + \chi$ (i.e., complete wave matching):

$$\frac{dV_1}{d\zeta} = -\frac{V_1}{D}[V_1 + \sigma V_2(\cos\Phi - \delta_1\sin\Phi)],$$

$$\frac{dV_2}{d\zeta} = -\frac{\eta}{D}[V_2 + \sigma V_1(\cos\Phi + \delta_1\sin\Phi)]. \qquad (9)$$

A further simplification of the situation is the case when the multipole interaction of radiation with the dipole-forbidden transition is absent ($\mu, \eta = 0$) and second harmonic is not generated ($V_2 = 0$). Then, we obtain from equations (9) and formulae (4)

$$\frac{dV_1}{d\zeta} = -\frac{V_1^2}{1 + \delta^2 - 2\beta_1 V_1 + \vartheta V_1^2}, \quad \vartheta = \xi_1 + \beta_1^2. \tag{10}$$

The transcendental solution to equation (10) is

$$\zeta = (1 + \delta^2)\left[\frac{1}{V_1} - \frac{1}{V_1(0)}\right] + \vartheta[V_1(0) - V_1] - 2\beta_1 \ln\frac{V_1(0)}{V_1}. \tag{11}$$

At exact resonance ($\delta = 0$), formula (11) reduces to an explicit expression for the coordinate dependence of the absorbed radiation intensity:

$$V_1(\zeta) = \frac{Q + \vartheta V_1^2(0) - V_1(0)\zeta - 1}{2\vartheta V_1(0)} = \frac{2V_1(0)}{Q - \vartheta V_1^2(0) + V_1(0)\zeta + 1}, \tag{12}$$

$$Q = \sqrt{[1 + V_1(0)\zeta - \vartheta V_1^2(0)]^2 + 4\vartheta V_1^2(0)}.$$

Equation (12) up to notation coincides with expression (45) from paper [3], obtained for TPA in the quantum Λ-system.

For small and large radiation intensities at the entrance to the medium equation (12) simplifies to:

$$V_1(\zeta) = \begin{cases} \dfrac{V_1(0)}{1 + \zeta V_1(0)}, & V_1(0) \ll 1, \\ \dfrac{V_1(0)}{1 + \zeta/[\vartheta V_1(0)]}, & V_1(0) \gg 1. \end{cases} \tag{13}$$

It follows from expression (13) that in the case of single-frequency TPA slow hyperbolic attenuation of radiation takes place. In the case of TPA of two waves with different frequencies, attenuation is exponential. This difference was noted previously for unsaturated TPA [7]. The fact that when passing from small to large intensities the behaviour of the absorption does not change is caused by an increase in the absolute values of the effective frequency detuning δ_1 (4) due to the dynamic Stark effect and the escape of radiation from resonance.

Dimensionless length (optical thickness) $\zeta_{1/2}$, at which the radiation intensity decreases by half compared with the intensity at the entrance to the medium, is determined from the expression (12):

$$\zeta_{1/2} = \frac{1}{V_1(0)} + \frac{\vartheta}{2} V_1(0).$$

The minimum length $\zeta_{1/2}$ is reached at a saturation parameter $V_{opt} = \sqrt{2/\vartheta}$, and for $V_1(0) = V_{opt}$ have $\zeta_{1/2} = \sqrt{2\vartheta}$.

4. Numerical experiment

Numerical calculations are performed for sodium vapours in a buffer gas. As the states 0, 1 and 2, we take respectively the states 3s, 3p and 3d. The wavelengths of the transitions $0 \leftrightarrow 1$, $1 \leftrightarrow 2$ and $0 \leftrightarrow 2$ are, respectively, equal to 5889.9, 8194.8 and 3426.88 Å [17]. Assume $N = 5\times10^{14}$ cm^3, $d_1 = 9.1\times10^{-18}$, $d_2 = 1.16\times10^{-17}$ [18], $\mu = 2.8\times10^{-20}$ esu, $\Gamma = 5.65\times10^9$ s^{-1} (the buffer gas pressure is ~30 Torr), $\gamma = \Gamma/3$, $A_1 = 5.9\times10^7$, $A_2 = 4.8\times10^7$ s^{-1} [18], $\chi = \pi/4$, $\rho_0^{(0)} = 1$, $\rho_1^{(0)} = \rho_2^{(0)} = 0$. In this case $\sigma = 1$, $\xi_1 = 11.85$, $\xi_2 = 0.494$, $\beta_1 = -0.490$, $\beta_2 = 0.563$, $\eta = 0.591$, $G = 0.256$ cm^{-1}, $f_1 = 0.478, f_2 = 0.528$, $V_{opt} = 0.407$, $\zeta_{1/2} = 4.92$, $z_{1/2} = 19.2$ cm. The saturation intensities I_{s1} and I_{s2} for the absorbed wave and waves with the doubled frequency, determined from the equality $V_{1,2} = 1$, are equal to 12.8 and 21.6 MW cm^{-2}, respectively, under these conditions.

The coefficient G (7) is directly proportional to the concentration N of absorbing atoms, and other parameters depend on N only through the additive components of the relaxation constants Γ and γ. In the presence of a buffer gas with a sufficiently high pressure, these additives are relatively small, and therefore the transition to other concentrations does not require recalculation of the dependences $V_{1,2}(\zeta)$ – it requires only an appropriate renormalisation of the optical thickness ζ, which does not affect the form of equations (7).

The dependences $V_{1,2}(\zeta)$ and $\Theta(\zeta)$, obtained by solving equations (7) numerically at small and large saturation parameters $V_1(0)$, are shown in Fig. 1. One can see from Figs 1b and f that the frequency-doubled radiation experiences spatial oscillations whose period varies only slightly with increasing intensity of the absorbed radiation: for $V_1(0) = 0.001$ the period is 13.3, whereas for $V_1(0) = 5$, it is somewhat smaller and is equal to 10.3. Note that in the case of three-wave stimulated Raman scattering (SRS), spatial oscillations of the wave amplitudes (mainly anti-Stokes waves) also take place at certain values of the wave mismatch [11]. Oscillations in a low-intensity field are observed at the wings of the gain line of the doubled-frequency field, and its generation is maximal at zero frequency detuning δ. As the intensity increases, the generation maximum $V_2(\zeta)$ is displaced from the line centre to the wings (Fig. 1e) and is asymmetric for positive and negative frequency detunings. This asymmetry is due to the fact that because of the phase relations the profiles of the absorption and gain lines [right-hand sides of the first two equations (7)] are the sum of the Lorentzian and dispersion profiles, and the effective detuning δ_1 (4) depends on the intensities of the fields.

It follows from equations (7) and (9) that at $V_1 \gg V_2$ the TPA coefficient is proportional to V_1^2. Therefore, weak-field absorption (Fig. 1a) varies considerably slower than that of saturated TPA (Fig. 1d). However, the ratio V_2/V_1 is higher for the weak field. Upon saturation the intensity V_1 experiences weakly pronounced spatial oscillations in the line wings (Fig. 1d).

At $V_1(0) \ll 1$ the phase Θ in the medium changes abruptly, starting from small values of ζ, and then its behaviour establishes: a jump as the sign of the frequency detuning changes and spatial oscillations with a relatively small amplitude (Fig. 1c). For positive detunings ($\delta > 0$), the phase is negative, while for $\delta < 0$ it is positive. Increasing the intensity of radiation absorbed qualitatively changes the behaviour of the phase (Fig. 1f). At small ζ, the phase retains its original value; then, starting from certain (decreasing in the absolute value) optical thicknesses, the phase decreases linearly with distance, remaining always negative, and experiences jumps. Spatial oscillations of the phase in this case are absent.

Calculations show that variation of the initial total phase of the waves at the entrance to the medium Φ does not affect V_1 and V_2. Dependences $\Theta(\Phi)$ for different $V_1(0)$ are similar in form to those shown in Fig. 2c in work [11]. The main feature of the behaviour of the phase Θ lies in the fact that it undergoes jumps at $\zeta = 0$ and when Φ changes in a region of some Φ_0, Φ_0 being shifted in a positive direction with increasing initial intensity $V_1(0)$. A qualitative explanation of the

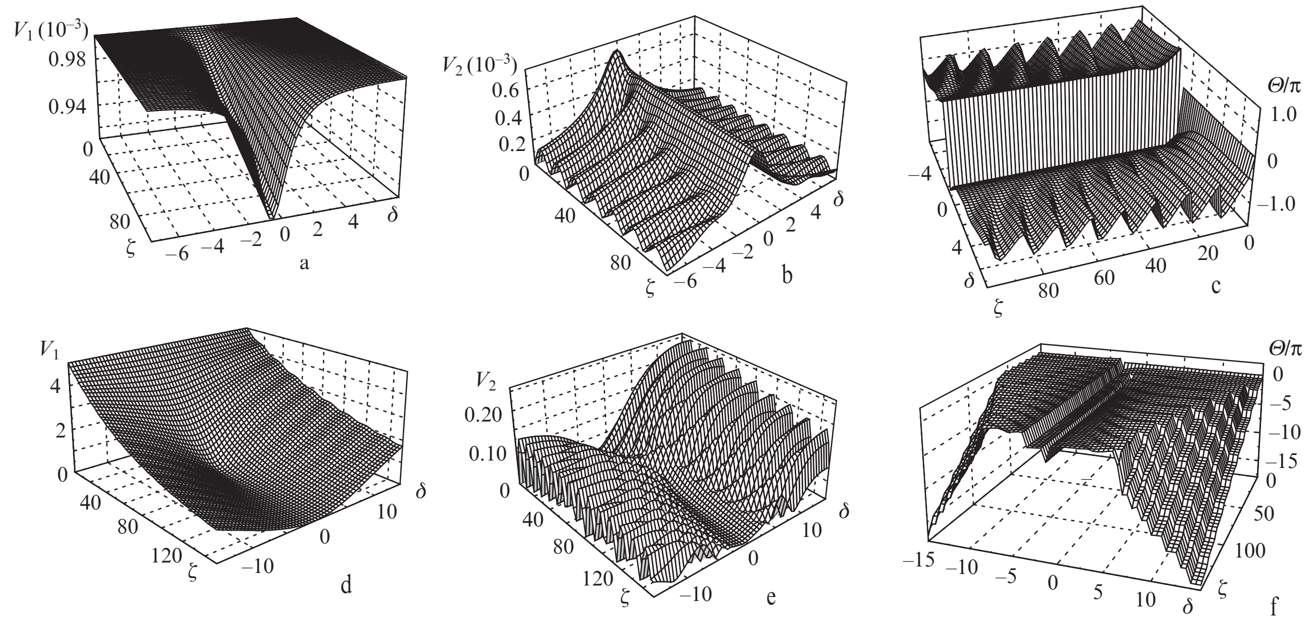

Figure 1. Behaviour of the dimensionless intensity of absorbed radiation V_1 (a, d), the dimensionless field amplitude V_2 with the doubled frequency (b, e) and the total phase of the wave Θ (c, f) as a function of the optical thickness ζ and the dimensionless frequency detuning δ for $V_1(0) = 0.001$ (a – c) and 5 (d – f), $V_2(0) = 10^{-12}$, $\chi = \pi/4$ and $\Phi = 0$.

phase jumps is the same as in the case of three-wave SRS [11]. At high intensities there are weakly pronounced spatial oscillations of the phase $\Theta(\zeta)$.

Comparison of numerical solutions of approximate equations (9) and (10) with the solution of equations (7), taking into account the phase change in the medium, is shown in Fig. 2. It follows from Figs 2a and c that the analytic solution to (11) for the extremely simplified equation (10) to within a few percent coincides with the solution obtained with the help of the exact equations (7). This is explained by relatively small amplitude of the generated field V_2 with the doubled frequency (Fig. 2d) and by small attenuation of the pump due to this generation. In particular, at $\zeta = 60$ and $\delta = 10$ the ratio of the wave intensities is $I_2/I_1 = V_2^2/(2\eta V_1) \approx 0.01$. In the case of fixed phase Θ, solutions to equations (9) for V_1 and V_2 are significantly different from the corresponding solutions to

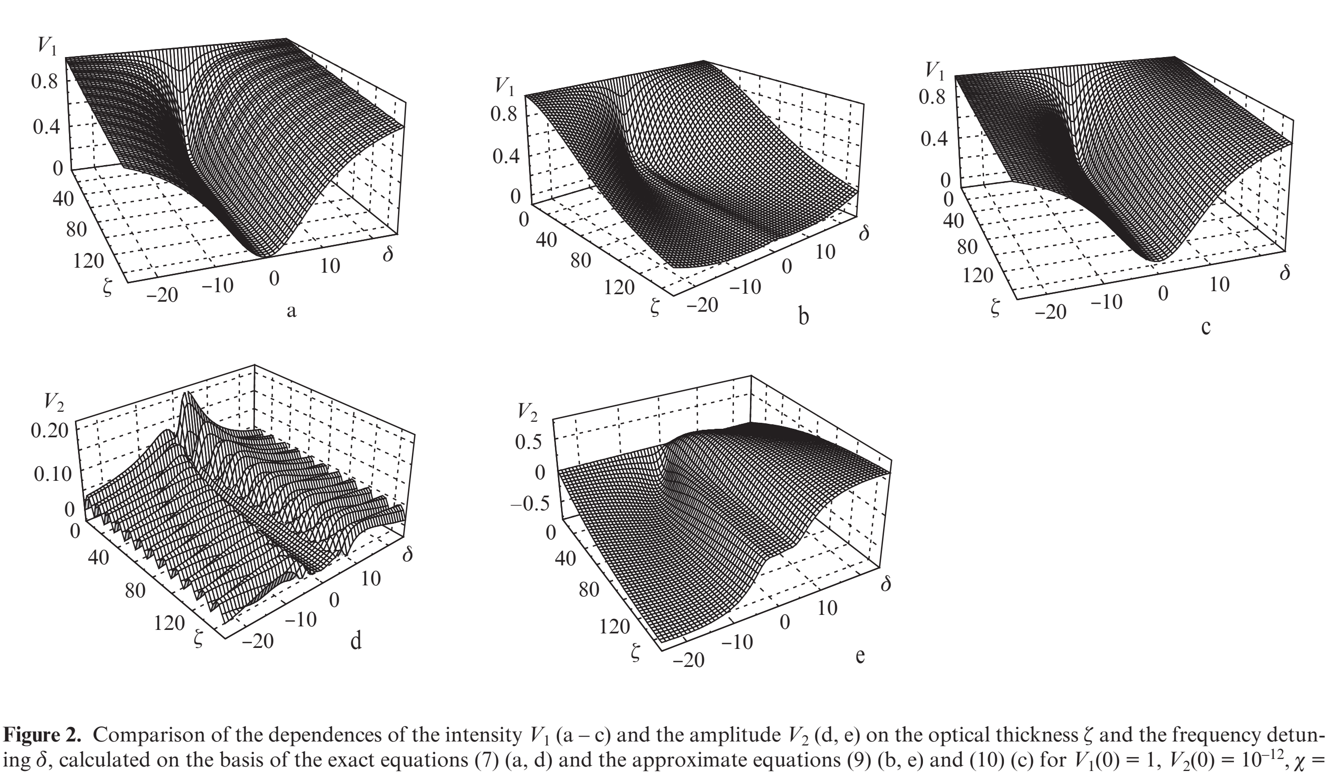

Figure 2. Comparison of the dependences of the intensity V_1 (a – c) and the amplitude V_2 (d, e) on the optical thickness ζ and the frequency detuning δ, calculated on the basis of the exact equations (7) (a, d) and the approximate equations (9) (b, e) and (10) (c) for $V_1(0) = 1$, $V_2(0) = 10^{-12}$, $\chi = \pi/4$ and $\Phi = 0$.

equations (7), i.e., the profile of the TPA lines in Fig. 2b is much wider and has a singularity near $\delta = 0$. Field generation with the doubled frequency (Fig. 2e) is qualitatively different from generation in the case of variable phase. The difference is that the amplitude of V_2 changes the sign (explanation of this fact is given in [11]), does not undergo spatial oscillations and is several times more at the line wings compared to the amplitude shown in Fig. 2d. The intensity ratio I_2/I_1 in this case is 0.25. Thus, neglecting the mutual phase adjustment of the waves in a medium leads to an overestimation of the calculated doubled-frequency radiation intensity by more than an order of magnitude.

5. Conclusions

Thus, this paper develops the most complete theory of nonresonant TPA of quasi-monochromatic radiation with allowance for saturation of absorption, for generation of doubled-frequency radiation due to the electric quadrupole and magnetic dipole interactions of radiation with the dipole-forbidden atomic transition and for a change in the total phase of the waves during their propagation caused by nonlinear dispersion of a gaseous medium.

Using numerical calculations we have shown that significant TPA occurs when the optical thicknesses of the medium exceed $\zeta > 20$, generation of fields with the doubled frequency arises at $\zeta > 5$, and its amplitude undergoes spatial oscillations both at small and at high intensities of the absorbed radiation. Its effectiveness in the metal vapours is relatively small, and the ratio of the intensities of the waves with doubled and initial frequencies do not exceed a few percent. This fact (which is not obvious in advance) ('negative' result) allows one to use a simple analytical expression (11) for the quantitative description of saturated TPA applied to the radiation medium.

In the case when of interest is the second-harmonic generation, the calculations must take into account the change in the total phase during the wave propagation in the medium, i.e., use equations (7) or (8). Neglecting this factor in equations (9) leads to significant qualitative and quantitative differences in the behaviour of the waves in comparison with their behaviour, reproducible by using a complete TPA description.

It is shown that in contrast to the three-wave SRS [11], the intensities of the waves involved in the TPA do not depend on the initial phase Φ. Such a dependence manifests itself only in the complex behaviour of the total phase in the medium Θ, dependent to a large extent on the radiation intensity.

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