

PACS numbers: 33.80. –b; 42.62.Fi
DOI: 10.1070/QE2005v035n11ABEH013032

Nonlinear absorption spectrum in the field of a weakly saturating standing wave

O.V. Belai, D.A. Shapiro, S.N. Yakovenko

***Abstract*. The spectrum of a probe field in a three-level system interacting with a standing wave at the adjacent transition is calculated. The expression for a nonlinear resonance profile is obtained taking into account diffusion in the velocity space in the fourth order of perturbation theory in the standing-wave amplitude.**

Keywords: *plasma, Coulomb collisions, nonlinear spectroscopy, probe field method, three-level system, perturbation theory.*

1. Introduction

Nonlinear spectral resonances in the field of a standing wave have been studied since the 1960s in connection with the development of the theory of a single-frequency gas laser. The velocity distribution of the population of atomic levels in a standing-wave field considerably differs from a simple sum of the spectra of resonance structures induced by counterpropagating travelling waves. This difference is caused by the spatial inhomogeneity of the field in the standing wave, which gives rise to higher spatial harmonics both in the level populations and polarisation of a medium. The solution for a two-level system is obtained in the form of a continued fraction [1, 2], which is expressed in terms of the Bessel function in the case of identical relaxation constants and an exact resonance [3]. However, after averaging over velocities, a smooth profile of the Lamb dip is formed in the lasing spectrum. It was shown that higher spatial harmonics are manifested only in the probe-field spectrum at the adjacent transition. Some structures were calculated numerically in [4] but they were not studied in detail.

The probe-field spectrum at the adjacent transition was measured in the Λ scheme at the lines of Ar II [5]. A strong field with the frequency ω was generated at the laser *mn* ($4p\,^2S_{1/2}-\ 4s\,^2P_{3/2}$) transition at $\lambda = 458$ nm, while the probe field at the frequency ω_μ in the form of travelling wave acted at the *ml* ($4p\,^2S_{1/2} - 3d\,^2P_{3/2}$) transition at $\lambda_\mu = 648$ nm (Fig. 1). Upon the probe-field detuning $\Omega_\mu = k_\mu\Omega/k$, the known field-splitting resonance was observed [6]. Along with this resonance, a new structure was observed at the centre of the spectrum ($\Omega_\mu = 0$) whose position was independent of the strong-field detuning Ω. For $\Omega = 0$, a dip was observed, which can be interpreted as the splitting of the field-splitting peak. As the detuning Ω was increased, the dip transformed to a peak, but its position did not change.

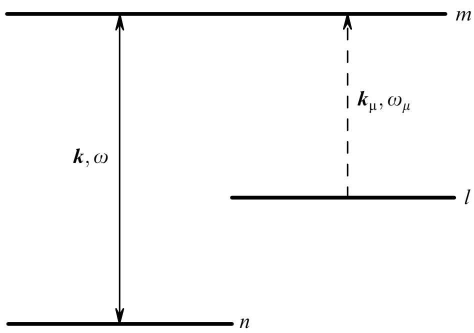

Figure 1. Scheme of the interaction of levels with the field: a strong standing wave (solid straight line) with the frequency ω and the wave vector $\boldsymbol{k}$ and a probe travelling wave (dashed straight line) with the frequency ω_μ and the wave vector $\boldsymbol{k}_\mu$.

It was shown that the new structure corresponds to the resonance of higher spatial harmonics. Its position in the spectrum was explained within the framework of the Feldman – Feld theory [4], whereas the interpretation of the resonance broadening due to ion – ion Coulomb collisions in a plasma required rather cumbersome numerical calculations [7]. The necessity appeared in a simple analytic theory which would describe the dependence of the resonance broadening on the concentration of charged particles and the ion temperature in a discharge.

The aim of this paper is to obtain and analyse the expression for the nonlinear absorption spectrum within the framework of the perturbation theory over the standing-wave amplitude. The higher spatial harmonics are not manifested in the second order of the perturbation theory. Because the second spatial harmonic appears in the fourth order of the perturbation theory, we restricted ourselves in this paper to the expansion of nonlinear absorption up to the fourth order.

2. Equations for the density matrix

The equation describing the behaviour of a three-level system in strong and probe light fields with the wave

O.V. Belai, D.A. Shapiro, S.N. Yakovenko Institute of Automation and Electrometry, Siberian Branch, Russian Academy of Sciences, prosp. akad. Koptyuga 1, 630090 Novosibirsk, Russia;
e-mail: shapiro@iae.nsk.su

Received 11 July 2005
Kvantovaya Elektronika **35** (11) 1027 – 1032 (2005)
Translated by M.N. Sapozhnikov

vectors $\boldsymbol{k}\|\boldsymbol{k}_\mu$, whose interaction with the system is considered as the perturbation $\hat{V} = -\boldsymbol{E}\hat{\boldsymbol{d}}/(2\hbar)$, is obtained from the equation for the density matrix [8]

$$\mathrm{i}\left(\Gamma_{ij} + \frac{\partial}{\partial t} + v\frac{\partial}{\partial x}\right)\rho_{ij} = [\hat{V}, \rho]_{ij}. \quad (1)$$

Here, x is the coordinate along the direction of wave vectors; v is the velocity component in the direction of the wave vectors; subscripts i, and j take the values l, m, n; Γ_{ij} are the radiative constants of the levels and transitions; ρ_{ij} are the elements of the density matrix ρ; $\boldsymbol{E}$ is the field strength in a light wave; $\hat{\boldsymbol{d}}$ is the dipole moment operator; and the brackets denote a commutator. Let us write equations for polarisations ρ_{ml} and ρ_{nl} of the allowed and forbidden transitions

$$\mathrm{i}\left(\Gamma_{ml} + \frac{\partial}{\partial t} + v\frac{\partial}{\partial x}\right)\rho_{ml} = V_{ml}\rho_{ll} + V_{mn}\rho_{nl} - \rho_{mm}V_{ml},$$
$$\mathrm{i}\left(\Gamma_{nl} + \frac{\partial}{\partial t} + v\frac{\partial}{\partial x}\right)\rho_{nl} = V_{nm}\rho_{ml} - \rho_{nn}V_{nl} - \rho_{nm}V_{ml}. \quad (2)$$

We neglect the populations of levels m and n, which were small in the experiment. The population of the level l has the equilibrium distribution $\rho_{ll} = N_l(v) = N_l v_T^{-1}\pi^{-1/2}\times \exp(-v^2 \times v_T^{-2})$, where v_T is the thermal velocity. Let us represent the fields of the travelling and standing waves in the form

$$\boldsymbol{E}_\mu(x,t) = \frac{1}{2}\boldsymbol{E}_\mu \exp(\mathrm{i}k_\mu x - \mathrm{i}\omega_\mu t) + \text{c.c.},$$
$$\boldsymbol{E}(x,t) = \boldsymbol{E}\cos kx \exp(-\mathrm{i}\omega t) + \text{c.c.} \quad (3)$$

The Rabi frequencies of the strong and probe fields are denoted by $G_\mu = \boldsymbol{E}_\mu \boldsymbol{d}_{ml}/(2\hbar)$ and $G_\pm = \boldsymbol{E}\boldsymbol{d}_{mn}/(2\hbar)$, where $\boldsymbol{E}$ and $\boldsymbol{E}_\mu$ are the amplitudes of the strong and probe field strengths; and $\boldsymbol{d}_{ij}$ is the matrix elements of the dipole moment.

By making the substitution

$$\rho_{ml} = \rho_\mu(x,v)\exp(\mathrm{i}k_\mu x - \mathrm{i}\Omega_\mu t),$$
$$\rho_{nl} = \rho_\nu(x,v)\exp(\mathrm{i}k_\mu x + \mathrm{i}\Omega t - \mathrm{i}\Omega_\mu t),$$

we obtain from (2) a closed system of equations for the amplitudes ρ_μ and ρ_ν of polarisations of the allowed and forbidden transitions

$$\left(\Gamma_\mu - \mathrm{i}\Omega_\mu + \mathrm{i}k_\mu v + v\frac{\partial}{\partial x}\right)\rho_\mu$$
$$= -\mathrm{i}[G_+\exp(\mathrm{i}kx) + G_-\exp(-\mathrm{i}kx)]\rho_\nu + \mathrm{i}G_\mu N_l(v),$$
$$\left(\Gamma_\nu + \mathrm{i}\Omega - \mathrm{i}\Omega_\mu + \mathrm{i}k_\mu v + v\frac{\partial}{\partial x}\right)\rho_\nu$$
$$= \mathrm{i}[G_+^*\exp(-\mathrm{i}kx) + G_-^*\exp(\mathrm{i}kx)]\rho_\nu, \quad (4)$$

where $\Gamma_\mu = \Gamma_{ml}$; $\Gamma_\nu = \Gamma_{nl}$. In these equations, only the interaction of ions with the light field is taken into account. However, under the conditions of the problem under study, ion–ion Coulomb interactions accompanied by a change in the ion velocity play an important role. Because Coulomb scattering occurs within small angles, it can be described as diffusion in the velocity space by adding the operator $-D\partial^2/\partial v^2$, to the left-hand sides of Eqns (4), where $D = \nu_c v_T^2/2$ is the diffusion coefficient;

$$\nu_c = \frac{16\sqrt{\pi}NZ^2e^4\Lambda}{3M^2v_T^3}$$

is the effective frequency of ion–ion Coulomb collisions; Ze is the ion charge; M is the ion mass; Λ is the Coulomb logarithm; and N is the total ion density [9]. The expression for the collision frequency uses the ion density because the number of excited ions is small and they are scattered almost exclusively by the ground-state ions.

3. Perturbation theory expansion in a series

Consider first for simplicity the equations neglecting collisions (4) and write them in the matrix form

$$\hat{L}\rho = r, \quad (5)$$

where

$$\rho = \begin{pmatrix}\rho_\mu \\ \rho_\nu\end{pmatrix};\ r = -\frac{\mathrm{i}G_\mu N_l}{\sqrt{\pi}v_T}\exp\left(-\frac{v^2}{v_T^2}\right)\begin{pmatrix}1\\0\end{pmatrix};\ \hat{L} = \hat{L}_0 + \Delta\hat{L};$$

$$\hat{L}_0 = v\frac{\partial}{\partial x} + \begin{pmatrix}\Gamma_\mu - \mathrm{i}(\Omega_\mu - k_\mu v) & 0 \\ 0 & \Gamma_\nu - \mathrm{i}(\Omega_\mu - \Omega - k_\mu v)\end{pmatrix};$$

$$\Delta\hat{L} = \begin{pmatrix}0 & \mathrm{i}G_+\mathrm{e}^{\mathrm{i}kx} + \mathrm{i}G_-\mathrm{e}^{-\mathrm{i}kx} \\ \mathrm{i}G_+^*\mathrm{e}^{-\mathrm{i}kx} + \mathrm{i}G_-^*\mathrm{e}^{\mathrm{i}kx} & 0\end{pmatrix}.$$

It is necessary to calculate the work of the probe field

$$P_\mu(\Omega_\mu) = -2\hbar\omega_\mu \mathrm{Re}\langle \mathrm{i}G_\mu^*\rho_\mu(x,v)\rangle, \quad (6)$$

where the angle brackets denote averaging over the phase space (x, v).

By using the weakness of interaction of an ion with the strong field, we represent the inverse operator as a series

$$\hat{L}^{-1} = (\hat{L}_0 + \Delta\hat{L})^{-1} = (1 + \hat{L}_0^{-1}\Delta\hat{L})^{-1}\hat{L}_0^{-1}$$
$$= \left[\sum_{m=0}^{\infty}(-\hat{L}_0^{-1}\Delta\hat{L})^m\right]\hat{L}_0^{-1}. \quad (7)$$

Let us expand the functions ρ and r in the basis

$$|n,p\rangle = \frac{1}{\sqrt{\lambda}}\begin{pmatrix}\delta_{p\mu}\\ \delta_{p\nu}\end{pmatrix}\exp(\mathrm{i}nkx), \quad (8)$$

where n is the number of a spatial harmonic and $\lambda = 2\pi/k$. The orthogonality relation for the basis functions has the form

$$\langle n', p'|n, p\rangle = \delta_{n'n}\delta_{pp'}. \quad (9)$$

Because the operator $\hat{L}_0$ is diagonal in this basis, the inverse operator $\hat{K}_0 = \hat{L}_0^{-1}$ is also diagonal and has the eigenvalues K_{np}:

$$\langle n', p'|\hat{L}_0^{-1}|n, p\rangle = \delta_{nn'}\delta_{pp'}K_{np},$$

$$K_{np} = \{\mathrm{i}vnk + \delta_{p\mu}(\Gamma_\mu - \mathrm{i}\Omega_\mu + \mathrm{i}k_\mu v)$$
$$+ \delta_{p\nu}[\Gamma_\nu + \mathrm{i}(\Omega - \Omega_\mu + k_\mu v)]\}^{-1}, \quad (10)$$

where $p = \mu, \nu$. The inverse operator can be conveniently written as the expansion in the projectors

$$L_0^{-1} = \sum_{n,p} |n,p\rangle K_{np} \langle n,p|. \quad (11)$$

Let us introduce the additional notation

$$a^+|n,p\rangle = |n+1,p\rangle, \quad a^-|n,p\rangle = |n-1,p\rangle, \quad (12)$$

$$\hat{U}|n,p\rangle = \delta_{p\nu}|n,\mu\rangle, \quad \hat{\mathscr{D}}|n,p\rangle = \delta_{p\mu}|n,\nu\rangle \quad (13)$$

to express $\Delta\hat{L}$ in terms of the raising and lowering operators a^+ and a^-:

$$\Delta\hat{L} = \mathrm{i}\hat{U}(G_+a^+ + G_-a^-) + \mathrm{i}\hat{\mathscr{D}}(G_+^*a^- + G_-^*a^+). \quad (14)$$

To calculate the work of the probe field, it is necessary to average the matrix element of the inverse operator (7)

$$\langle 0,\mu|\hat{L}^{-1}|0,\mu\rangle = \langle 0,\mu|\left[\sum_{m=0}^{\infty}(-\hat{L}_0^{-1}\Delta\hat{L})^m\right]\hat{L}_0^{-1}|0,\mu\rangle \quad (15)$$

over velocities with the Maxwell distribution. The operator $\Delta\hat{L}$ changes the parity of n because it consists of the operators a^+ and a^-, which increase and decrease, respectively, the harmonic number n by unity.

Let us return to general equations taking into account ion collisions accompanied by a change in their velocity. Compared to the collisionless case considered above, only the operator $\hat{L}_0^{-1}$, inverse to the unperturbed operator, will change. To find this operator, we perform the Fourier transform for the polarisation $\rho_\mu(v)$ and the right-hand side of Eqn (5):

$$-(kn + k_\mu)\frac{\mathrm{d}z(q)}{\mathrm{d}q} + (\Gamma_\mu - \mathrm{i}\Omega_\mu + Dq^2)z(q) = r(q), \quad (16)$$

where $z(q)$ is the Fourier transform of polarisation $\rho_\mu(v)$; and $r(q)$ is the Fourier transform of the right-hand side $r(v)$ of Eqn (5).

The solution of this equation can be written in the form

$$z(q) = -\frac{E_{n\mu}(q)}{kn + k_\mu}\int_C^q E_{n\mu}^{-1}(q_1)r(q_1)\mathrm{d}q_1, \quad (17)$$

where

$$E_{n\mu}(q) = \exp\left(\frac{Dq^3/3 + q\Gamma_\mu - \mathrm{i}q\Omega_\mu}{kn + k_\mu}\right); \quad (18)$$

C is a constant determined by the boundary conditions. The boundary condition for $z(q)$ is the requirement $z(\pm\infty) \to 0$. Taking into account the behaviour of $E(q)$ at infinity, we can conclude that, if $kn + k_\mu > 0$, then C should be set equal to $+\infty$ to provide the convergence of the integral in (17). Correspondingly, in the opposite case $kn + k_\mu < 0$, we should set $C = -\infty$. Note that $E_{n\mu}(-q) = E_{n\mu}^{-1}(q)$, so that the general solution has the form

$$z(q) = -\frac{1}{kn + k_\mu}\int_C^q E_{n\mu}(q)E_{n\mu}(-q_1)r(q_1)\,\mathrm{d}q_1, \quad (19)$$

where $C = \mathrm{sign}(kn + k_\mu) \times \infty$;

$$\mathrm{sign}\,x = \begin{cases} +1, & x > 0 \\ -1, & x < 0. \end{cases}$$

It follows from Eqns (4) that the general solution of a similar equation for the ν component can be obtained from the solution for the μ component (18) by the substitution $\Gamma_\mu \to \Gamma_\nu$, $\Omega_\mu \to \Omega_\mu - \Omega$, i.e.,

$$E_{n\nu}(q) = \exp\left[\frac{Dq^3/3 + q\Gamma_\nu - \mathrm{i}q(\Omega_\mu - \Omega)}{kn + k_\mu}\right]. \quad (20)$$

Then,

$$\rho(q) = -\int_C^q\left[\sum_{n,p}\frac{|n,p\rangle E_{np}(q)E_{np}(-q_1)\langle n,p|}{nk + k_\mu}\right]r(q_1)\,\mathrm{d}q_1. \quad (21)$$

The result of the action of the inverse operator on the right-hand side of Eqn (5) will be the s-fold integral, where s is the perturbation theory order.

4. Nonlinear absorption spectrum

The Fourier transform of the right-hand side of Eqn (5) in the Doppler limit ($v_T \to \infty$) is the delta function

$$r(q) = \frac{\mathrm{i}}{\sqrt{2\pi}}\int_{-\infty}^{\infty}|0,\mu\rangle\exp(-\mathrm{i}qv)\mathrm{d}v = \mathrm{i}\sqrt{2\pi}\,\delta(q)|0,\mu\rangle. \quad (22)$$

The expression in the zero order of the perturbation theory contains the step function $\Theta(q)$:

$$(\hat{L}_0 + Dq^2)^{-1}r(q) = \mathrm{i}k_\mu^{-1}\sqrt{2\pi}\Theta(-q)E_{0\mu}(q)|0,\mu\rangle. \quad (23)$$

Under the action of the operator $\Delta\hat{L}$, the zero harmonic $|0,\mu\rangle$ transforms to the two adjacent harmonics $|1,\nu\rangle$ and $|-1,\nu\rangle$. The operator $(\hat{L}_0 + Dq^2)^{-1}$ acting on the linear combination of harmonics changes only coefficients at them, without adding new harmonics. The subsequent action of the operator $\Delta\hat{L}$ again adds the adjacent harmonics $|2,\mu\rangle$, $|0,\mu\rangle$, and $|-2,\mu\rangle$. The expression for the required quantity P_μ contains $\rho_\mu(x,v)$ averaged over x, which is equivalent to the scalar multiplication of the sum of harmonics by the zero harmonic $\langle 0,\mu|$. Because of the orthogonality relation, only the term $\langle 0,\mu|0,\mu\rangle = 1$ will be nonzero.

The generation of harmonics is illustrated in Fig. 2. To obtain the contribution of a harmonic to $P_\mu^{(s)}$, it is necessary to make s steps from $|0,\mu\rangle$ to $|0,\mu\rangle$ along one of the routes shown in Fig. 2, by moving each time to the right-upward or to the right-downward to an adjacent node. The total number of such routes in the s order of the perturbation theory is

$$N = C_s^{s/2} = s!\left(\frac{s}{2}!\right)^{-2}$$

(if s is odd, then $N = 0$ because it is impossible to come back to the zero harmonic for the odd number of steps). For $s = 2$, 4, and 6, the number of routes N is 2, 6, and 20, respectively.

In the Stokes case, when $k_\mu < k$, a part of integrals vanish. In the second order of the perturbation theory, the integral containing the factor $|G_-|^2$ and corresponding to

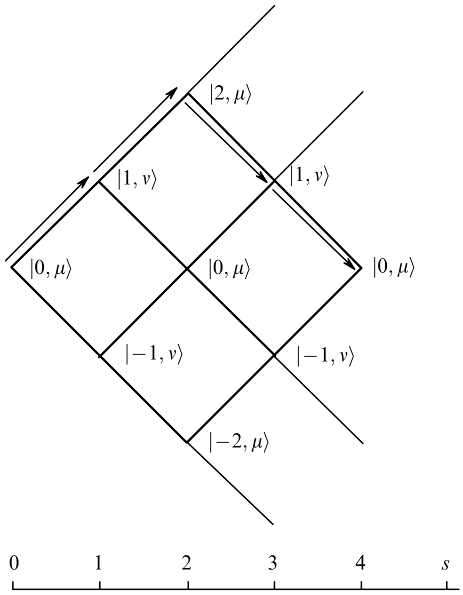

Figure 2. Increase in the number of harmonics due to the s-fold action of the operator $\Delta\hat{L}$. The arrows indicate the route for which the contribution of harmonics to the probe-field work vanishes.

the route $|0,\mu\rangle \to |1,\nu\rangle \to |0,\mu\rangle$ vanishes. In the fourth order, the integral containing the factor $|G_-|^4$ also vanishes and one integral with $|G_+|^2$ and four integrals with factors $|G_+G_-|^2$ remain. Of these four integrals, one is also zero, namely, the integral for the route $|0,\mu\rangle \to |1,\nu\rangle \to |2,\mu\rangle \to |1,\nu\rangle \to |0,\mu\rangle$, which is indicated by the arrows in the scheme. In the second order of the perturbation theory, only one integral remains, and four integrals remain in the fourth order. They correspond to the following routes in Fig. 2:

$$\begin{aligned}
&|0,\mu\rangle \to |-1,\nu\rangle \to |0,\mu\rangle,\\
&|0,\mu\rangle \to |-1,\nu\rangle \to |-2,\mu\rangle \to |-1,\nu\rangle \to |0,\mu\rangle,\\
&|0,\mu\rangle \to |-1,\nu\rangle \to |0,\mu\rangle \to |-1,\nu\rangle \to |0,\mu\rangle,\\
&|0,\mu\rangle \to |-1,\nu\rangle \to |0,\mu\rangle \to |1,\nu\rangle \to |0,\mu\rangle,\\
&|0,\mu\rangle \to |1,\nu\rangle \to |0,\mu\rangle \to |-1,\nu\rangle \to |0,\mu\rangle.
\end{aligned} \tag{24}$$

Let us write the obtained multiple integrals for $G_+ = G_- = G$:

$$\Delta P_\mu = \delta P_\mu^{(2)} + \delta P_\mu^{(4)}, \tag{25}$$

$$\delta P_\mu^{(2)} = \frac{-G^2}{k_\mu^2(k_\mu - k)} \int_\infty^0 \mathrm{d}q_4 F_{0,-1}(q_4) \times \int_{-\infty}^{q_4} \mathrm{d}q_3 F_{-1,0}(q_3)\Theta(-q_3), \tag{26}$$

$$\begin{aligned}
\delta P_\mu^{(4)} &= \frac{G^4}{k_\mu^2(k_\mu - k)^2(k_\mu - 2k)} \int_\infty^0 \mathrm{d}q_4 F_{0,-1}(q_4)\\
&\times \int_{-\infty}^{q_4} \mathrm{d}q_3 F_{-1,-2}(q_3) \int_{-\infty}^{q_3} \mathrm{d}q_2 F_{-2,-1}(q_2)\times\\
&\times \int_{-\infty}^{q_2} \mathrm{d}q_1 F_{-1,0}(q_1)\Theta(-q_1) + \frac{G^4}{k_\mu^3(k_\mu - k)^2}\\
&\times \int_\infty^0 \mathrm{d}q_4 F_{0,-1}(q_4) \int_{-\infty}^{q_4} \mathrm{d}q_3 F_{-1,0}(q_3)\\
&\times \int_\infty^{q_3} \mathrm{d}q_2 F_{0,-1}(q_2) \int_{-\infty}^{q_2} \mathrm{d}q_1 F_{-1,0}(q_1)\Theta(-q_1)\\
&+ \frac{G^4}{k_\mu^3(k_\mu - k)(k_\mu + k)} \int_\infty^0 \mathrm{d}q_4 F_{0,-1}(q_4) \int_\infty^{q_4} \mathrm{d}q_3 F_{-1,0}(q_3)\\
&\times \int_\infty^{q_3} \mathrm{d}q_2 F_{0,1}(q_2) \int_\infty^{q_2} \mathrm{d}q_1 F_{1,0}(q_1)\Theta(-q_1)\\
&+ \frac{G^4}{k_\mu^3(k_\mu - k)(k_\mu + k)} \int_\infty^0 \mathrm{d}q_4 F_{0,1}(q_4) \int_\infty^{q_4} \mathrm{d}q_3 F_{1,0}(q_3)\\
&\times \int_\infty^{q_3} \mathrm{d}q_2 F_{0,1}(q_2) \int_\infty^{q_2} \mathrm{d}q_1 F_{-1,0}(q_1)\Theta(-q_1),
\end{aligned} \tag{27}$$

where

$$F_{mn}(q) = E_{mp_1}(-q)E_{np_2}(q); \quad F_{mn}(q) = F_{nm}(-q);$$

$p_1 = \mu$ if m is even and $p_1 = \nu$ in the opposite case; p_2 is defined in terms of n similarly, and E_{np} are described by expressions (18) and (20). We can show that integrals in the fourth-order perturbation theory can be simplified and reduced to integrals of a lower multiplicity, i.e., to the products of single, twofold, and threefold integrals. Due to such a simplification, the integrals can be comparatively easily calculated numerically. The order of sequence of the integrals in sum (27) corresponds to that of the routes in (24). The resonance of higher spatial harmonics is caused by the term in (27) corresponding to the only route passing through the second harmonic: $|0,\mu\rangle \to |-1,\mu\rangle \to |-2,\mu\rangle \to |-1,\mu\rangle \to |0,\mu\rangle$.

The dependences $\Delta P_\mu(\Omega_\mu)$ for different G and Ω are presented in Fig. 3. We used the calculation parameters that were close to the experimental ones. In the left part of the figure are shown the dependences for $\Omega = 0$ with the standing-wave amplitude increasing from top to bottom. Beginning with some amplitude, the top of the field-splitting resonance becomes flat and then a dip appears, which corresponds to the second spatial harmonic. The observed dip is broader than for the case $D = 0$. In the right part of the figure are presented the dependences obtained for the same amplitude of the standing wave but its different detunings, which increase from top to bottom. One can see that the dip transforms to a peak located at the line centre, which does not change its position upon detuning. The peak amplitude decreases with increasing detuning. This behaviour qualitatively agrees with the experiment.

To elucidate the physical meaning of individual terms in (25), we calculate integrals in (26) and (27) for $D = 0$:

$$\delta P_\mu^{(2)} = \frac{2G_+^2(k_\mu - k)}{[k_\mu(-\Gamma_\mu + \Gamma_\nu + \mathrm{i}\Omega) + k(\Gamma_\mu - \mathrm{i}\Omega_\mu)]^2}, \tag{28}$$

$$\delta P_\mu^{(4)} = \frac{6G_+^4(k - k_\mu)^2 k_\mu}{[k_\mu(-\Gamma_\mu + \Gamma_\nu + \mathrm{i}\Omega) + k(\Gamma_\mu - \mathrm{i}\Omega_\mu)]^4} + \frac{G_-^2 G_+^2 k_\mu^2[k_\mu^2(\Gamma_\mu - \Gamma_\nu - \mathrm{i}\Omega) + 6k^2(\Gamma_\mu - \mathrm{i}\Omega_\mu)]}{2k^2[k_\mu(-\Gamma_\mu + \Gamma_\nu + \mathrm{i}\Omega) + k(\Gamma_\mu - \mathrm{i}\Omega_\mu)]^3(\Gamma_\mu - \mathrm{i}\Omega_\mu)^2}$$

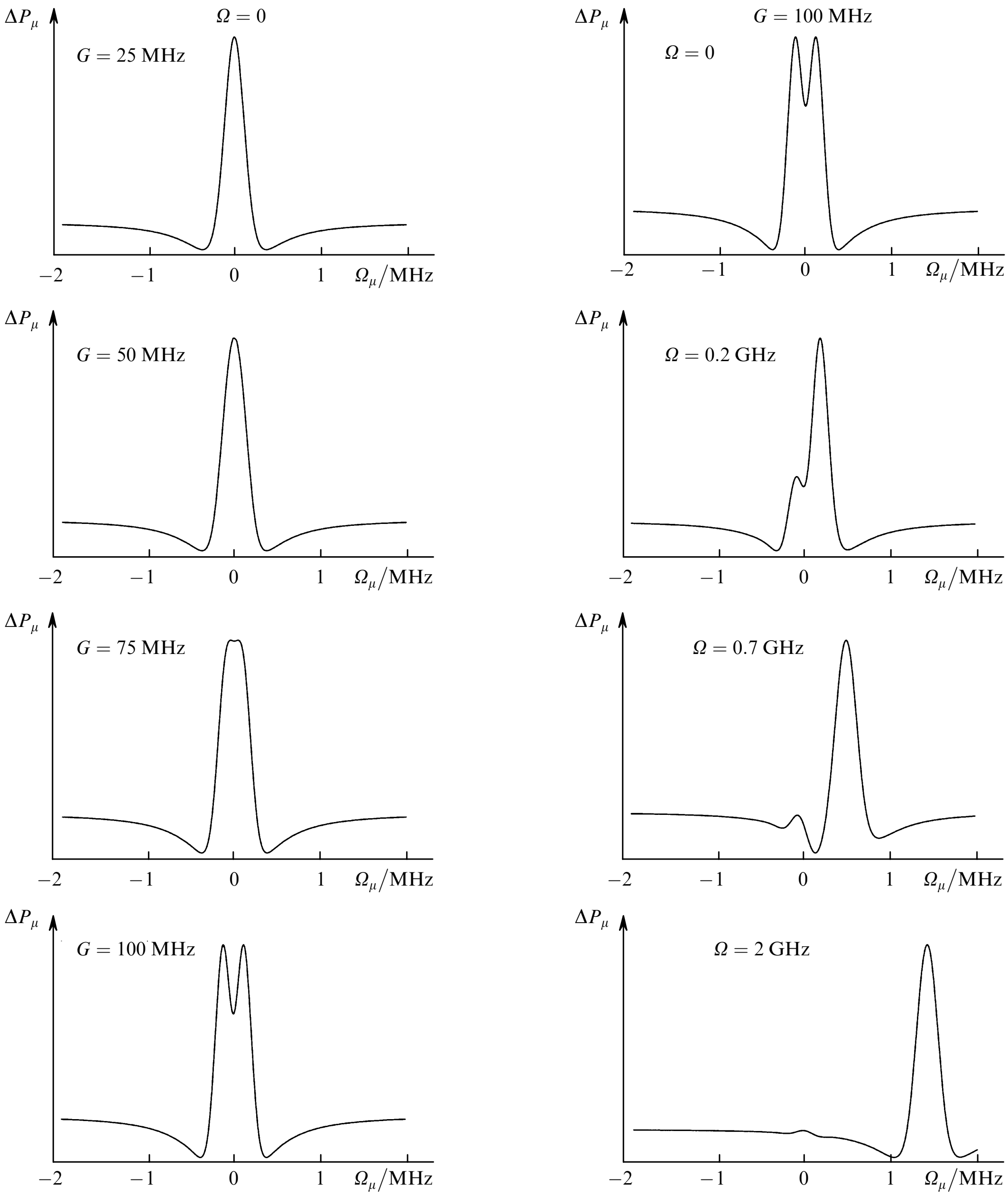

Figure 3. Dependences of $\Delta P_\mu(\Omega_\mu)$ for different G (on the left) and Ω (on the right).

$$+\frac{G_-^2 G_+^2 k_\mu^2 [k k_\mu(-7\Gamma_\mu + 2\Gamma_\nu + 2\mathrm{i}\Omega + 5\mathrm{i}\Omega_\mu)]}{2k^2[k_\mu(-\Gamma_\mu + \Gamma_\nu + \mathrm{i}\Omega) + k(\Gamma_\mu - \mathrm{i}\Omega_\mu)]^3(\Gamma_\mu - \mathrm{i}\Omega_\mu)^2}$$

$$-\frac{2\mathrm{i}G_-^2 G_+^2 (k - k_\mu)^3}{k[\Gamma_\nu + \mathrm{i}(\Omega - \Omega_\mu)]\{\mathrm{i}[k\Gamma_\mu + k_\mu(-\Gamma_\mu + \Gamma_\nu + \mathrm{i}\Omega)] + k\Omega_\mu\}^3}. \quad (29)$$

A similar expression was obtained earlier in papers [5, 10]. The term (28) of the second order in the standing-wave amplitude describes the resonance centred at the frequency $\Omega_\mu = k_\mu \Omega / k$. Resonances at other frequencies appear only in the fourth order. The first term in (29) is similar to the second-order term (28) and describes the increase in the field splitting with increasing G_+^2. The second and third terms in (29) describe a nonlinear structure with the width Γ_μ at the line centre with $\Omega_\mu = 0$. This can be seen from the factor $(\Gamma_\mu - \mathrm{i}\Omega_\mu)^{-2}$, which is contained only in these terms. The latter term in (29) appears due to the spatial modulation of the coherence of the forbidden nl transition, resulting in the appearance of a nonlinear structure with the width Γ_ν of the forbidden transition width at the two-photon resonance frequency $\Omega_\mu = \Omega$.

In the anti-Stokes case, the expression for the work of the probe field in a medium with a large Doppler broadening is simplified because only one integral corresponding to the lower route in Fig. 2 remains:

$$\delta P_\mu^{(4)} \propto \int_\infty^0 \mathrm{d}q_4 F_{0,-1}(q_4) \int_\infty^{q_4} \mathrm{d}q_3 F_{-1,-2}(q_3)$$

$$\times \int_C^{q_3} \mathrm{d}q_2 F_{-2,-1}(q_2) \int_\infty^{q_2} \mathrm{d}q_1 F_{-1,0}(q_1)\Theta(-q_1), \quad (30)$$

where $C = \mathrm{sign}(k_\mu - 2k) \times \infty$. The nonlinear absorption spectrum is shown in Fig. 4. One can see that the amplitude of the resonance, as in the Stokes case, decreases with

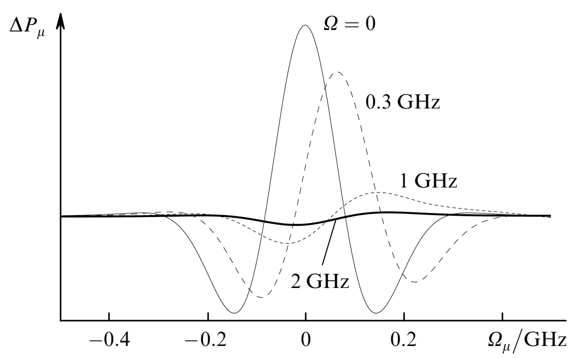

Figure 4. Dependences of $\Delta P_\mu(\Omega_\mu)$ for $k_\mu/k = 1.4$ and different Ω.

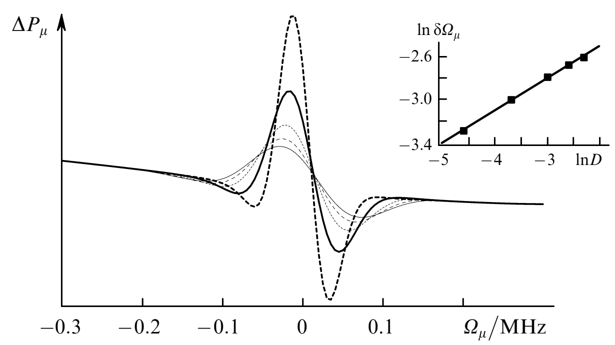

Figure 5. Peaks of higher spatial harmonics for $\Gamma_\nu = \Gamma_\mu = 1$ GHz, $D/D_0 = 0.1, 0.075. 0.05, 0.025, 0.001$ (in the order of increasing the peak amplitude) and $D_0 = 10^{13}$ m^2 s^{-3}. The insert shows the dependence of the width of the peak on the diffusion coefficient.

increasing detuning. The profile of the nonlinear resonance is of alternating sign. One can show that the area under the nonlinear resonance contour of higher spatial harmonics (27) or (30) is zero.

5. Discussion of results

Although expressions (26) and (27) for the spectrum are rather cumbersome, they have a simple structure. The integrands are the products of Green functions describing diffusion in the velocity space and relaxation of polarisations of the allowed and forbidden transitions. These expressions allow one to estimate the width of the resonance of higher spatial harmonics and to find the dependence of ΔP_μ on parameters in the limiting case of the diffusion time shorter compared to the relaxation time: $(Dk^2)^{-1/3} \ll \Gamma_{\mu,\nu}^{-1}$.

Consider the case with the zero detuning $\Omega = 0$. By making the change of integration variables $t_i = (D/k)^{1/3} q_i$ ($i = 1 - 4$) in integrals (26) and (27), in terms of which $\Delta P_\mu(\Omega_\mu)$ is expressed, we obtain the dependence of ΔP_μ on D

$$\Delta P_\mu(\Omega_\mu) = D^{-4/3} f\left(\frac{\Omega_\mu}{k^{2/3}D^{1/3}}, \frac{k_\mu}{k}\right), \tag{31}$$

where f is a function of two dimensionless variables. Because Ω_μ enters into (31) only in the above combination, the width (FWHM) of the peak of higher spatial harmonics is $\delta\Omega_\mu = C_1 D^{1/3} k^{2/3}$. To verify this estimate, we plotted a family of curves $\Delta P_\mu(\Omega_\mu)$ (Fig. 5) and found the width of the peak near $\Omega_\mu = 0$ for different D. The FWHM of the peak as a function of D is shown in the insert. The slope angle was 0.31, in accordance with the estimate. The coefficient C_1 proved to be 0.15. Such dependence can be simply qualitatively explained.

The peak broadens due to a change in the velocity after Coulomb collisions. During the time t, an ion is scattered by a random angle ϑ, so that $\langle\vartheta\rangle = 0$ and $\langle\vartheta^2\rangle = \nu_c t$. The ion, which was initially located in the antinode of a standing wave and had the zero velocity projection on the wave-vector direction and the transverse velocity close to the thermal velocity, acquires the longitudinal velocity $\delta u \sim (\nu_c t)^{1/2} v_T$ for the time t and propagates over the distance $\delta x \sim \nu_c^{1/2} t^{3/2} v_T$. When the phase shift caused by the Doppler frequency shift $nk\delta x$ of the nth harmonic becomes equal to π, the phase is disrupted and the effect of this harmonic disappears. The characteristic time of this process is $t \sim (nkv_T)^{-2/3} \nu_c^{-1/3}$. The resonance at the centre appears because of the presence of higher even spatial harmonics with $n = 2, 4, \ldots$. Therefore, the estimate should be performed by considering the influence of harmonics with $n = 2$; in this case, the FWHM of the peak is $\delta\Omega_\mu \sim D^{1/3} k^{2/3}$. For $\delta\Omega_\mu \sim \Gamma_\mu$, the dip in the resonance is 'blurred' and the width of the peak in the absence of resonance is approximately doubled.

6. Conclusions

We have obtained analytic expressions for the nonlinear absorption spectrum taking into account diffusion in the velocity space. The contours of the nonlinear resonance of higher spatial harmonics were calculated for the experimental situation and anti-Stokes case. The broadening of the resonance caused by the Coulomb interaction was found. The expressions obtained by using the perturbation theory agree with the known limiting case [5] and numerical calculations [7]. It was shown that the Coulomb scattering in a plasma qualitatively explains experimental data.

Acknowledgements. The authors thank M.G. Stepanov for useful discussions. This work was partially supported by the State Program 'Leading Scientific Schools' (Grant No NSh-439.2003.2).

References

1. Stenholm S., Lamb W.E. *Phys. Rev.*, **181**, 618 (1969).
2. Feldman B.J., Feld M.S. *Phys. Rev. A*, **1**, 1375 (1970).
3. Rautian S.G., Sobel'man I.I. *Zh. Eksp. Teor. Fiz.*, **44**, 934 (1963) [*Sov. Phys. JETP*, **17**, 635 (1963)].
4. Feldman B.J., Feld M.S. *Phys. Rev. A*, **5**, 899 (1972).
5. Babin S.A., Podivilov E.V., Potapov V.V., Churkin D.V., Shapiro D.A. *Zh. Eksp. Teor. Fiz.*, **121**, 807 (2002) [*JETP*, **94**, 694 (2002)].
6. Babin S.A., Stepanov M.G., Churkin D.V., Shapiro D.A. *Zh. Eksp. Teor. Fiz.*, **125**, 1029 (2004) [*JETP*, **98**, 953 (2004)].
7. Belai O.V., Shapiro D.A. *Pis'ma Zh. Eksp. Teor. Fiz.*, **79**, 257 (2004) [*JETP Lett.*, **79**, 203 (2002)].
8. Rautian S.G., Smirnov G.I., Shalagin A.M. *Nelineinye rezonansy v spektrakh atomov i molekul* (Nonlinear Resonances in Spectra of Atoms and Molecules) (Novosibirsk: Nauka, 1979).
9. Babin S.A., Shapiro D.A. *Phys. Rep.*, **241**, 119 (1994).
10. Popov A.K. *Vvedenie v nelineinuyu spektroskopiyu* (Introduction to Nonlinear Spectroscopy) (Novosibirsk: Nauka, 1983).