Coherent Nonlinear Resonances of Saturated Absorption at Transitions from the Ground $^1S_0$ Atomic State in the Spectroscopy of Unidirectional Waves

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Abstract: The shapes of the saturated absorption resonances at transitions from the ground $^1S_0$ atom state in a gas medium in the field of two unidirectional waves are investigated both analytically and numerically. It is shown for the first time that the spectra of nonlinear resonances are formed by processes in the two-and three-level V-schemes resulting from the degeneracy of the upper P transition state, are qualitatively different at the resonant transition and transitions to the more excited atom states. It is established that the physical causes for differences of these spectra are due to the different degree of branching of the radiation from the excited to the ground state (the degree of openness) of transition. The degree of openness of transition determines the amount of contributions of coherent processes to the resonance shapes both in the system of two levels (coherent population beats) and three levels (nonlinear interference effect).

Keywords: Nonlinear Spectroscopy, Saturated Absorption Resonance, Unidirectional Waves, Closed and Open Transitions

1. Introduction

Nonlinear spectroscopic phenomena under the resonant interaction of several light fields with degenerate atomic systems have been investigated for a long time. The interest in these phenomena is due to a number of specific physical processes in these systems, including the effects of atomic states interference, leading to the narrow resonance structures in the spectra under study (see review [1]). A striking example of coherent phenomena in the transitions from the ground state of atoms are the electromagnetically induced transparency (EIT) resonances [2], which are based on the coherent level population trapping (CPT) phenomenon [3]. However, along with EIT resonances, resonances of the opposite sign were found in such transitions, which were called electromagnetically induced absorption (EIA) resonances. For the first time, EIA resonances were observed precisely in the field of two unidirectional laser waves close in frequency at the closed transition in the Rb atom [4] and

were explained by the spontaneous transfer of magnetic coherence of the excited state levels to the ground state under the conditions of a closed transition [5]. Subsequently, EIA resonances were also observed at a number of other transitions in Rb and Cs atoms [6]. Later in [7], it was reported about the registration by the method of magnetic scanning of EIT and EIA resonances in the field of two counter-propagating laser waves.

Note that the anomalies of the EIA resonances recorded in experiments on degenerate transitions, as, for example, in [6, 7], were not always explained in the framework of the mechanism of [5]. Therefore, to explain such effects, other processes were considered, such as optical pumping and CPT [6], or collisions [8], and not always justifiably, but they gave resonance structures similar to the experimental ones. It was shown in [9] that the main process determining the behavior of the resonance amplitudes in the experiment [7] is the magnetic coherence, induced by the fields of counter-propagating waves at the ground state levels, and

the effect of atomic transition closeness in the resonance shapes is small.

In the recent paper [10], in the development of the concept [5], the authors stated that the cause of narrow peak structures formation in the spectra of saturated absorption resonances (EIA resonances) on closed transitions with any level moment values is the spontaneous transfer of the magnetic coherence induced by light fields at the upper state levels to the lower one. However, there is no analysis of other processes that can form the narrow structures of nonlinear resonance.

In [11] we showed that, in a simple two-level quantum system, the structures of nonlinear resonance in a field of two frequencies can manifest themselves not only as a narrow dip, as was assumed for a long time [12, 13], but also as a narrow absorption peak (resonance EIA). Moreover, the peak width can be less than the lower level width. Such situation is possible at a closed transition, when a spontaneous decay of the upper level occurs along a working transition. The reason for the appearance of a narrow peak structure is the beats of the level populations of a two-level quantum system in the field of two waves with similar frequencies [12].

The practical implementation of a closed two-level system is possible at atomic transitions from the ground nondegenerate state, in particular, at the resonant or intercombination transitions from the ground $^1S_0$ state in the alkaline-earth and similar atoms. However, on these transitions, due to the degeneracy of the upper state, when a nonlinear resonance is detected by the probe field method, the so-called V-scheme of transition level interaction with optical fields is formed. The features of the nonlinear resonance spectrum, in this case, will be determined by the contributions of the processes both in two-and three-level systems. In this case, the shape of the nonlinear resonance will depend on the nature of the light wave polarizations, the magnitude of the upper state level splittings and, in the case of moving atoms, also on the propagation direction of a strong and probe light wave.

Note that the closed V-type transitions, as indicated in [14], are also determinant in the formation of the nonlinear probe field absorption spectrum at resonant transitions of alkali metal atoms (transition with full level moments J-J + 1).

The present paper is devoted to the study of the saturated absorption resonance formation processes in the probe field method of unidirectional laser waves at the transitions from the ground $^1S_0$ atomic state (between levels with full moments J=0-J=1), the dependence of these processes on the level relaxation constants, the magnitude of level splittings and optical field intensities, as well as clarifying the role of transition openness in these resonances formation.

2. Theoretical Model

Consider the problem of the probe field absorption spectrum in a gaseous medium with the V-type of atomic transition in the presence of a unidirectional strong wave field. The scheme of atomic transitions is shown in Figure 1. Such scheme of interaction is realized on the transitions from the $^1S_0$ atomic state. The strong wave is assumed to be plane, monochromatic, linearly polarized (of frequency ω, wave vector k and electric field strength E) and resonant to the atomic *m-n* transition (with transition frequency ω_{mn}). The probe wave is also monochromatic (of frequency ω_μ, wave vector k_μ and electric field strength E_μ) with circular polarization. It is assumed that the absorbing medium is placed in a magnetic field of strength H, which magnitude may vary, and that both waves propagate collinearly to the magnetic field direction (k, k_μ || H). When solving the problem, we take into account that the medium is saturated by the probe wave (at the assumption that it is weak, compared with the strong wave). The gas is assumed to be sufficiently rarefied so that collisions can be neglected. The medium is supposed to be optically thin.

We will consider the problem in the coordinate system with the quantization axis along the wave vector k (k ‖ Z axis). In this system of coordinates the transitions between levels with a change in the magnetic quantum number ΔM= ±1 (for a linear-polarized field) and ΔM= +1, or ΔM= −1 (for a circular polarized field) are allowed (Figure 1).
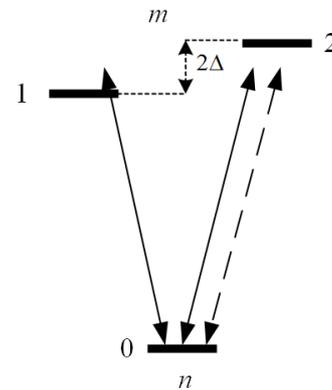
Figure 1. *Scheme of the optical field interaction with the V-type of transition; 2Δ is the level splitting (the solid and dashed lines show the strong and probe field, respectively).*

When solving the problem, we will use the kinetic equations for the density matrix of the atomic system. According to [12], the dynamics of diagonal elements ρ_i and off-diagonal elements ρ_{ik} of the density matrix in the model of relaxation constants is described by the following system of equations:

$$\frac{d\rho_i}{dt} + \Gamma_i \rho_i = Q_i + \sum_k A_{ki} \rho_k - 2\operatorname{Re}(i\sum_j V_{ij}\rho_{ji}) - 2\operatorname{Re}(i\sum_j V_{ij}^\mu \rho_{ji}) \tag{1}$$

$$\frac{d\rho_{ik}}{dt} + (\Gamma_{ik} + i\omega_{ik})\rho_{ik} = -i[V,\rho]_{ik} - i[V^\mu,\rho]_{ik} \tag{2}$$

In Eqs. (1) and (2), $\frac{d}{dt} = \frac{\partial}{\partial t} + \upsilon\nabla$ is the total derivative operator, Γ_i are the level relaxation constants, Γ_{ik} are the transition line half-widths, Q_i are the level excitation rates, V and V^μ are the interaction operators of atom with the strong and probe fields that are defined as: $V = -G\exp(i(kr-\omega t)) + h.c.$, $V^\mu = -G^\mu\exp(i(k_\mu r-\omega_\mu t)) + h.c.$, where $G = dE/2\hbar$, $G^\mu = dE_\mu/2\hbar$, and d is the dipole moment operator. The term $A_{ki}\rho_k$ in the equation (1) determines the spontaneous decay of the upper state m to the lower state n (this process rate is A_{mn}); this term is missing in the equations for the upper level populations.

We note that this formulation of the problem and the obtained solutions are valid both for transitions between excited states and for the case when the lower state n is the ground atom state. In this case, the relaxation constant of the lower state Γ_n is replaced by the average transit width, which is determined by the characteristic light beam transverse size d_0 and the most probable particle velocity v_T.

We seek solutions for the system of equations (1, 2) in the following form (the first harmonic approximation in ε): for

diagonal elements-as $\rho_i = \rho_i^0 + \rho_i^+\exp(i(\varepsilon t - (k_\mu - k)r)) + \rho_i^-\exp(-i(\varepsilon t-(k_\mu-k)r))$; for the off-diagonal elements at allowed transitions-as $\rho_{ik} = R_{ik}\exp(-i(\omega t-kr)) + R_{ik}^\mu\exp(-i(\omega_\mu t-k_\mu r)) + R_{ik}^s\exp(-i(\omega_s t-k_s r))$ and for off-diagonal elements at forbidden transitions-as $\rho_{ik} = r_{ik}^0 + r_{ik}^+\exp(i(\varepsilon t-(k_\mu-k)r)) + r_{ik}^-\exp(-i(\varepsilon t-(k_\mu-k)r))$, where $\varepsilon = \omega_\mu - \omega$, $\omega_s = 2\omega - \omega_\mu$, $k_s = 2k - k_\mu$. The validity of this form of solutions in the stationary case for the transition considered in a wide range of the probe and saturating wave intensities was demonstrated by us in [15].

In the approximation of rotating optical fields, the set of equations (1, 2) for the density matrix under steady-state conditions is reduced to the set of equations for coefficients ρ_i^0, $\rho_i^\pm$, R_{ik}, R_{ik}^μ, R_{ik}^s, r_{ik}^0, $r_{ik}^\pm$. Allowing for the Hermitian character of these coefficients, we will write below only independent equations for the case of linear-polarized strong field. In case of the circular polarized strong field, the equations are obtained from the following ones leading to the zero one of the strong field circular strength component.

In this approximation the populations of the lower (0) and upper (1, 2) levels are described by the following set of equations:

$$\Gamma_n\rho_0^0 = Q_0 + \sum_{k=1,2} A_{k0}\rho_k^0 + \sum_{k=1,2}\left(2\operatorname{Re}(iG_{0k}R_{k0})\right) + 2\operatorname{Re}(iG_{02}^\mu R_{20}^\mu)$$

$$(\Gamma_n + i\varepsilon)\rho_0^+ = \sum_{k=1,2} A_{k0}\rho_k^+ + i(G_{01}R_{10}^s + G_{02}R_{20}^s) + i(G_{02}^\mu R_{20} - G_{20}R_{02}^\mu - G_{10}R_{01}^\mu) \tag{3}$$

$$\Gamma_m\rho_1^0 = Q_1 + 2\operatorname{Re}(iG_{10}R_{01})$$

$$(\Gamma_m + i\varepsilon)\rho_1^+ = i(G_{10}R_{01}^\mu - G_{01}R_{10}^s)$$

$$\Gamma_m\rho_2^0 = Q_2 + 2\operatorname{Re}(iG_{20}R_{02}) + 2\operatorname{Re}(iG_{20}^\mu R_{02}^\mu)$$

$$(\Gamma_m + i\varepsilon)\rho_2^+ = i(G_{20}R_{02}^\mu - G_{02}^\mu R_{20} - G_{02}R_{20}^s)$$

The set of equations for the polarizations on allowed 1-0, 2-0 and forbidden 2-1 transitions has the forms:

$$(\Gamma_{mn} + i\Omega_{10})R_{01} = iG_{01}(\rho_1^0 - \rho_0^0) + iG_{02}r_{21}^0 + iG_{02}^\mu r_{21}^-$$

$$(\Gamma_{mn} + i(\Omega_{10} - \varepsilon))R_{01}^s = iG_{01}(\rho_1^- - \rho_0^-) + iG_{02}r_{21}^- \tag{4}$$

$$(\Gamma_{mn} + i\Omega_{10}^\mu)R_{01}^\mu = iG_{10}(\rho_1^+ - \rho_0^+) + iG_{02}r_{21}^+ + iG_{02}^\mu r_{21}^0$$

$$(\Gamma_{mn} - i\Omega_{20})R_{20} = -iG_{20}(\rho_2^0 - \rho_0^0) - iG_{20}^\mu(\rho_2^+ - \rho_0^+) - iG_{10}r_{21}^0$$

$$(\Gamma_{mn} - i(\Omega_{20} - \varepsilon))R_{20}^s = -iG_{20}(\rho_2^+ - \rho_0^+) - iG_{10}r_{21}^+$$

$$(\Gamma_{mn} - i\Omega_{20}^\mu)R_{20}^\mu = -iG_{20}(\rho_2^+ - \rho_0^+) - iG_{20}^\mu(\rho_2^0 - \rho_0^0) - iG_{10}r_{21}^-$$

$$(\Gamma_{21} + i\omega_{21})r_{21}^0 = i(G_{20}R_{01} - G_{01}R_{20} + G_{20}^\mu R_{01}^\mu) \tag{5}$$

$$(\Gamma_{21} + i(\omega_{21} + \varepsilon))r_{21}^+ = i(G_{20}R_{01}^\mu - G_{01}R_{20}^s)$$

$$(\Gamma_{21} + i(\omega_{21} - \varepsilon))\bar{r}_{21} = i(G^{\mu}_{20}R_{01} - G_{01}R^{\mu}_{20} + G_{20}R^{s}_{01})$$

In equations (3-5), Γ_n and Γ_m are the relaxation constants of the lower and upper states, Γ_{mn} is the uniform transition line half-width, Γ_{21} is the relaxation constant of the polarization between the magnetic sublevels (magnetic coherence) of the upper state; $\Omega_{ik} = \omega - \omega_{ik}$ and $\Omega^{\mu}_{ik} = \omega_{\mu} - \omega_{ik}$ are the detunings of the strong and probe field frequencies from the transition frequencies ω_{ik} between the sublevels of m and n states. Further, we assume that, in the absence of optical fields, the level populations are: $\rho_0 = N_n$, $\rho_1 = \rho_2 = N_m$.

Taking into account that the atom motion leads to a change in the equations: $\Omega_{ik} \rightarrow \Omega_{ik} - kv$, $\Omega^{\mu}_{ik} \rightarrow \Omega^{\mu}_{ik} - k_{\mu}v$, and $\varepsilon \rightarrow \varepsilon - (k_{\mu} - k)v$, where v is the atom velocity vector. Below, we will consider a case of unidirectional ($k \approx k_{\mu}$) waves with close frequencies.

The probe field absorption line shape (calculated per atom) was determined as

$$\alpha/(\alpha_0 N_{nm}) = -\Gamma_{mn}\langle Re(i(R_{20}^{\mu}G^{\mu}_{02}))/|G^{\mu}|^2\rangle, \qquad (6)$$

where the designation $\langle...\rangle$ means averaging over the Maxwellian distribution of particle velocities and $\alpha_0 = 4\pi\omega_{mn}d^2/c\hbar\Gamma_{mn}$ is the resonance absorption cross section. The probabilities A_{ki} of the magnetic sublevel decay for each spontaneous channel were taken to be the same and equal to A_{mn}.

3. Results of Solutions of the Original Model Equations

The exact analytical solutions of the stationary system of equations (3)-(5) are complex and not very informative. Therefore, these equations were solved further numerically. To understand the physical processes determining the characteristics of nonlinear resonance spectra, in some cases, the analytical solutions of this system of equations were obtained in the weak probe field approximation and the first nonlinear corrections in a strong field.

In the numerical simulation, the following transition constant values were used: $\Gamma_m = 5.5 \cdot 10^7 c^{-1}$, $\Gamma_n = (10^{-2} \div 10^{-1})\Gamma_m$, $\Gamma_{mn} = (\Gamma_m + \Gamma_n)/2$, $A_{mn} = a_0\Gamma_m$, where a_0 is the branching parameter of radiation from the upper state. At the resonant transition the branching parameter is $a_0 = 1$, since the upper state decay follows the working transition (the transition is closed). For the transitions from the ground to more excited states or between excited states, the branching parameter is $a_0 < 1$ (open transitions), because there are always channels of spontaneous decay of the upper working level to the other underlying levels. The relaxation constant of the polarization between the upper state sublevels is equal to the constant Γ_m. The Doppler line width is assumed to be $kv_T = 5.2 \cdot 10^9 c^{-1}$, the range of variation in the particle velocities in the integration was $\pm 3kv_T$ with the step of $\Delta kv_T = (10^{-3} \div 10^{-4})kv_T$. The saturation parameters of strong κ_s and probe κ_p fields are chosen in the form: $\kappa_s = 2(dE/2\hbar)^2\gamma_{mn}/(\Gamma_{mn}\Gamma_m\Gamma_n)$, $\kappa_p = 2(dE_{\mu}/2\hbar)^2\gamma_{mn}/(\Gamma_{mn}\Gamma_m\Gamma_n)$, where E, E_{μ} are the strengths of the strong and probe field circular components, d

is the reduced matrix element of the m-n transition dipole moment, and $\gamma_{mn} = \Gamma_m + \Gamma_n - A_{mn}$. The values of saturation parameters were varied in the ranges $\kappa_s \leq 50$ and $\kappa_p \leq \kappa_s$. Further, the results are presented for the probe field saturation parameter value $\kappa_p = 10^{-3}$.

## 3.1. Numerical Calculation of the Resonance Line Shapes

The results of numerical calculations of the saturated absorption line shape of the circularly polarized probe wave are shown in Figure 2 for the resonant (closed) transition, and in Figure 3 for the transition to the more excited state (open transition with parameter $a_0 = 0.5$) for several splittings of the upper state levels and values: $\Omega_s = 0$, $\Gamma_n = 0.02\Gamma_m$, $\kappa_s = 1$, $\kappa_p = 10^{-3}$.

In the absence of level splitting, a resonance is formed on the Doppler absorption line contour of the probe wave near the frequency detuning $\Omega_{\mu} = 0$ shaped as a traditional wide dip and a narrow structure of a small amplitude in the line center (curves 1). At the resonant (closed) transition the structure appears as a narrow absorption peak (Figure 2), and, at the open transitions, the structure manifests itself as a narrow transmission peak (Figure 3). The forms of the resonance spectra near the line center, in this case, are due to the contributions of the processes in two-and three-level transition schemes. In case of level splitting exceeding the transition width ($\Delta > \Gamma_{mn}$), the resonances are formed in the two-and three-level schemes are divided in the frequency scale and appear in their pure shapes (curves 2 and 3).

In the two-level system (0 and 2) a resonance is formed by wave fields with identical circular polarizations. Moving atoms contribute to this resonance, for which field frequency detunings satisfy the following conditions: $\Omega_{20} = \omega - \omega_{20} - kv = 0$ and $\Omega^{\mu}_{20} = \omega_{\mu} - \omega_{20} - kv = 0$. Therefore, a resonance occurs near the field frequency difference $\varepsilon = \omega_{\mu} - \omega = 0$ and is manifested in the shape of a traditional population dip. At the same time, in the center of the dip at $\varepsilon = 0$, as in the absence of level splitting, a narrow structure is formed as a peak of absorption at the closed transition (Figure 2), or a peak of enlightenment at the open transition (Figure 3).
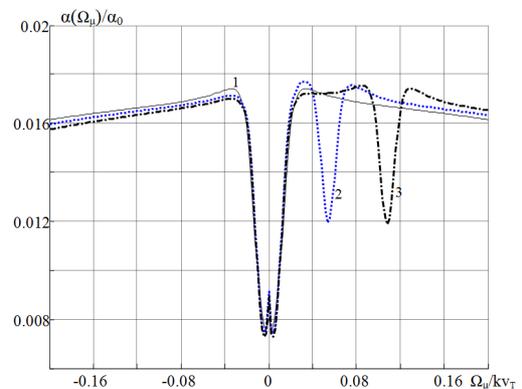
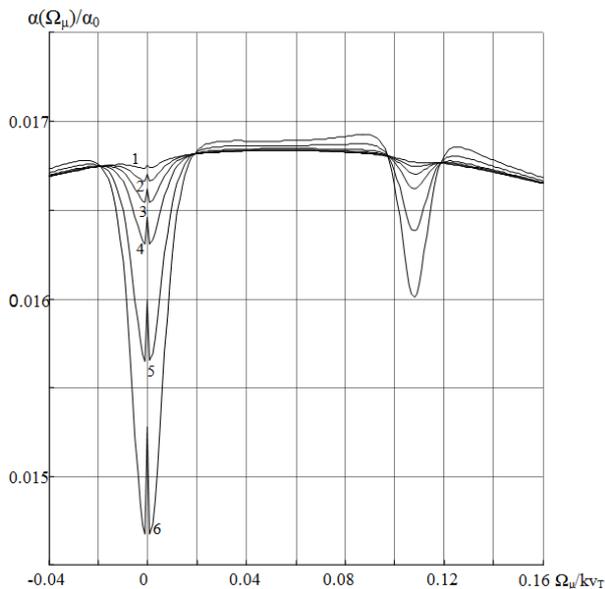

Figure 2. The resonance shapes at transition from the ground $^1S_0$ to the first excited P state at $a_0 = 1$, $\Omega_s = 0$, $\kappa_s = 1$, $\kappa_p = 10^{-2}$, $\Gamma_n = 0.02\Gamma_m$ and level spittings: $\Delta = 0$ (1), $5\Gamma_{mn}$ (2), $10\Gamma_{mn}$ (3).
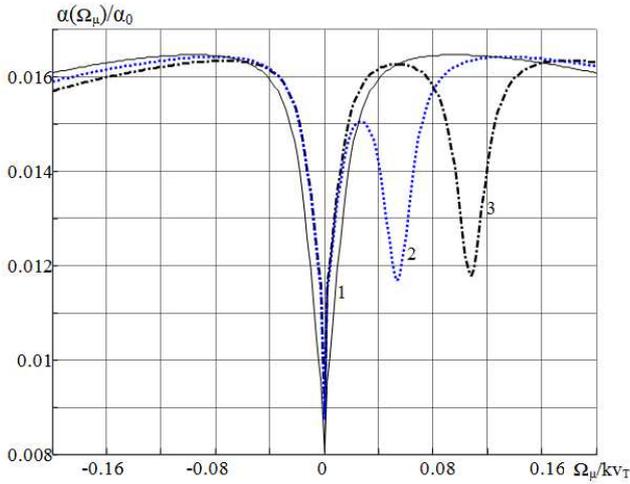
Figure 3. *The resonance shapes at transition from the ground $^1S_0$ to the highly excited P state at $a_0 = 0.5$, $\Omega_s = 0$, $\kappa_s = 1$, $\kappa_p = 10^{-2}$, $\Gamma_n = 0.02\Gamma_m$ and level spittings: $\Delta = 0$ (1), $5\Gamma_{mn}$ (2), $10\Gamma_{mn}$ (3).*

In the three-level system a resonance is formed by the wave fields with different circular polarizations. In this case, the atoms, for which the field frequency detunings are as follows: $\Omega_{10} = \omega - \omega_{10} - kv = 0$ and $\Omega^\mu_{20} = \omega_\mu - \omega_{20} - kv = 0$, contribute to the resonance. This resonance arises near the field frequency differences $\varepsilon = \omega_\mu - \omega = \omega_{21} = 2\Delta$ and also manifests itself as a dip, but with other parameters. In this case, the dip profiles at the resonant transition and transitions to the more excited states differ both in their amplitude and width, as well as in the shape of their wings. At the resonant transition, in wings of a dip the maxima characteristic for coherent processes are formed, while there are no such structures at transitions to excited states. This fact testifies about the distinction of the processes creating a resonance spectrum on the considered types of transitions.

Figure 4. *The resonance shapes at transition from the ground $^1S_0$ to the first excited P state at $a_0 = 1$, $\Omega_s = 0$, $\kappa_p = 10^{-2}$, $\Gamma_n = 0.02\Gamma_m$, $\Delta = 10\Gamma_{mn}$ and different values of κ_s: $\kappa_s = 2 \cdot 10^{-3}$ (1), $5 \cdot 10^{-3}$ (2), 10^{-2} (3), $2 \cdot 10^{-2}$ (4), $5 \cdot 10^{-2}$ (5), 10^{-1} (6).*

The action of the strong field intensity on the shape of the saturated absorption probe wave line on the resonant transition is shown in Figures 4, 5, where the strong field saturation parameter κ_s changes from 0.002 to 0.1. Under the conditions of Figures 4, 5 a growth of the strong wave intensity leads to the linear growth in the amplitudes of both dips and peak structure, but poorly influences on the widths of dip and peak structure. Thus, the central dip amplitude exceeds the amplitude of the frequency shifted dip by a factor of $2 \div 2.5$ and the peak structure amplitude by a factor of $3 \div 4$, but the dip widths differ only by $\sim 20\%$. The full widths of resonances change also by the same value with a change of parameter κ_s. At the same time, the changes in the width of the narrow peak structure are not found.

It should be noted that an increase in the strong field intensity leads to a qualitative change in the shape of the frequency shifted resonance: at saturation parameters $\kappa_s > 0.01$, the wide peaks arise in the dip wings (curves 4-6), whose amplitudes and widths, as well as the positions of their maxima, depend on the κ_s value. This fact indicates a change in the nature of the process that forms the resonance spectrum at values $\kappa_s > 0.01$. It will be shown below that, under these conditions, the field splitting of the lower state levels will occur, leading to a change in the resonance shape.

Calculations of the nonlinear resonance shapes in a wider range of changes in the strong field intensities (up to saturation parameter values $\kappa_s \sim 50$) showed that, at values $\kappa_s > 0.5$, the dependences of amplitude and width of the central dip become close to the root-like, whereas the dependence of the frequency shifted dip amplitude remains linear. In this case, a nonlinear increase in the width of this dip occurs, but more slower than that of the central dip. So, at value $\kappa_s = 10$, the dip amplitudes differ by $\sim 20\%$, and their widths differ more than twice. At the same time, a growth of the strong wave intensity leads to an increase in the amplitudes and widths of the peaks in the frequency-shifted dip wings and an increase in the distance between their maxima close to the root law.

Amplitude of the peak structure at this saturation parameters changes also nonlinearly and reaches a maximum value at $\kappa_s = 1 \div 2$. The maximum peak-to-dip contrast ($\sim 30\%$) is observed at values $\kappa_s \sim 0.5 \div 0.7$. At the same time, a change in the strong wave intensity does not effect practically on the width of the narrow peak structure. Note that, under the identical conditions, the pica amplitude, in case of level splitting, significantly exceeds the pica amplitude in the absence of splitting (Figure 2).

Calculations of the nonlinear resonance shape on the open transition (at $a_0 = 0.5$) showed that the amplitudes and widths of the dips formed in the two-and three-level systems in the range of saturation parameters $\kappa_s \sim 10^{-2} \div 50$ are almost identical and depend identically on κ_s: the amplitudes rise linearly at $\kappa_s < 1$ and according to the root law at $\kappa_s \gg 1$, while the widths across the whole range have a root-like dependence.
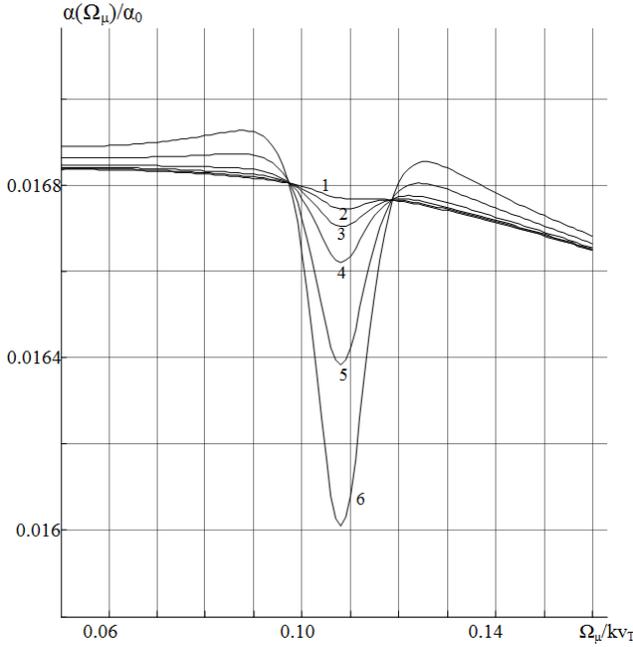
Figure 5. *The fragment of the coherent part of resonance (of Figure 4) with the higher spectral resolution.*

The amplitude dependence of the narrow (dip) structure on the saturation parameter κ_s, as well as at the closed transition, is also nonlinear with a maximum near the values of $\kappa_s \sim 2$. At the same time, the maximum of the narrow structure contrast, with respect to the dip, is realized at small values of

parameter $\kappa_s \sim 0.1$ and reaches almost $\sim 100\%$. The structure contrast decreases with an increase in κ_s. It is important to note that, at the open transition, the growth of the parameter κ_s in the range of $\kappa_s \sim 10^{-2} \div 10$ leads to an increase in the narrow dip structure width by more than twice, unlike the closed transition, where the strong field intensity does not effect on the width of the narrow peak structure.

Numerical and analytical calculations show that the parameters of narrow resonance structures (peak and dip) are defined by the lower level relaxation constant Γ_n. The reduction of constant value Γ_n leads to an increase in the amplitudes and a decrease in the widths of narrow resonance structures.

3.2. Analytical Calculations of Resonance Line Shapes

To identify the physical processes that form the structures of the saturated absorption resonance spectra in unidirectional waves, shown in Figures 2, 3, based on numerical solutions of the equations (3–5), we will consider the approximate analytical solutions of these equations for two-and three-level atomic transitions.

3.2.1. The Resonance Shape in the Two-Level System

In the system of two levels (0 and 2, Figure 1) the saturated absorption resonance shape can be obtained from equations (3-5) for a weak probe field in the approximation of the first nonlinear corrections in a strong field and a large Doppler broadening ($\Gamma_{mn} << kv_T$) in the following form [4]:

$$\alpha(\varepsilon)\Big/(\alpha_0 N_{nm}) = \sqrt{\pi}\,\frac{\Gamma_{mn}}{kv_T}\exp\left[-(\Omega_\mu/kv_T)^2\right]\left\{1-2|G|^2\,\mathrm{Re}\left[\frac{1}{2\Gamma_{mn}-i\varepsilon}\left(\frac{\Gamma_m+\Gamma_n-A_{mn}}{\Gamma_m\Gamma_n}+\frac{a_m}{\Gamma_m-i\varepsilon}+\frac{a_n}{\Gamma_n-i\varepsilon}\right)\right]\right\}, \quad (7)$$

where: $a_m = \dfrac{\Gamma_m+A_{mn}-\Gamma_n}{\Gamma_m-\Gamma_n}$, $a_n = \dfrac{\Gamma_m-A_{mn}-\Gamma_n}{\Gamma_m-\Gamma_n}$.

It follows from expression (7) that, in the probe-wave absorption spectrum a resonance consisting of three spectral components with different widths and amplitudes is formed near the frequency detuning $\varepsilon = 0$ ($\Omega_\mu = \Omega$). The component described by the first term in the nonlinear part of expression (7) is due to the strong field saturation, it forms in the spectrum a traditional populational dip with the half-width of $2\Gamma_{mn}$. The other two components (the second and third terms) are due to the level population beatings and have half-widths Γ_m and Γ_n, respectively. The amplitudes of components are determined by coefficients a_m and a_n. It follows from the expressions for these coefficients that, in case of the ratio of constants $\Gamma_m > \Gamma_n$, coefficient a_m is always positive, and the sign of coefficient a_n, determining the sign (form) of the

narrow resonance component, depends on the relation between the relaxation constants and Einstein coefficient A_{mn}. The narrow structure manifests itself in the spectrum as an additional dip when the constants are related as $\Gamma_m\text{-}A_{mn} > \Gamma_n$ ($a_n > 0$) and in the form of a peak when $\Gamma_m\text{-}A_{mn} < \Gamma_n$ ($a_n < 0$). In case of ratio $\Gamma_m\text{-}A_{mn} = \Gamma_n$ the narrow structure in the resonance spectrum is absent, since $a_n = 0$.

For transitions with a long-living low level ($\Gamma_m >> \Gamma_n$) the coefficients are defined as: $a_m \approx 1+a_0$, $a_n \approx 1-a_0-\Gamma_n/\Gamma_m$, where $a_0 = A_{mn}/\Gamma_m$ is the radiation branching parameter.

For the transitions with the branching parameter $a_0 < 1-\Gamma_n/\Gamma_m$ (open transitions), the probe wave absorption line shape is defined from (7) as:

$$\alpha(\varepsilon)\Big/(\alpha_0 N_{nm}) = \sqrt{\pi}\,\frac{\Gamma_{mn}}{kv_T}\exp\left[-(\Omega_\mu/kv_T)^2\right]\left[1-2|G|^2\,\mathrm{Re}\left(\frac{1-a_0}{\Gamma_n}\frac{1}{2\Gamma_{mn}-i\varepsilon}+\frac{1-a_0}{\Gamma_m}\frac{1}{\Gamma_n-i\varepsilon}\right)\right]. \quad (8)$$

In this case the resonance appears shaped as a wide dip with a half-width of $2\Gamma_{mn}$ and a narrow dip within the wide one with a half-width of Γ_n, while the amplitudes of the narrow and wide dips are identical. This amplitude

relationship is observed in the numerical calculations of resonance shapes for the open transition at small saturation parameters ($\kappa_s \leq 0.1$).

In case of transitions with the branching parameter $a_0 = 1$

(closed transitions), the probe wave absorption line shape is

$$\alpha(\varepsilon)\Big/(\alpha_0 N_{nm}) = \sqrt{\pi}\,\frac{\Gamma_{mn}}{kv_{\mathrm{T}}}\exp\left[-(\Omega_\mu/kv_{\mathrm{T}})^2\right]\left\{1-2|G|^2\,\mathrm{Re}\left[\frac{1}{2\Gamma_{mn}-i\varepsilon}\left(\frac{1}{\Gamma_m}+\frac{2}{\Gamma_m-i\varepsilon}-\frac{\Gamma_n}{\Gamma_m}\frac{1}{\Gamma_n-i\varepsilon}\right)\right]\right\}. \tag{9}$$

In this case, the resonance manifests itself being shaped as a wide (with a half-width of $2\Gamma_{mn}$) dip, within which a narrow absorption peak with a half-width of Γ_n is formed. The narrow peak amplitude is about 3 times smaller than the dip structure amplitude. This fact is in agreement with the above-presented numerical calculation data for the peak contrast at small saturation parameters κ_s.

Note that in the approximation of the first nonlinear corrections the amplitudes of the main dip and narrow structures depend linearly on the strong field intensity, whereas the widths of the dip and its structures are independent from the field intensity. This fact was noted above at the discussion of the numerical results presented in Figures 2, 3. When accounting the next strong-field corrections, the dependences of the parameters of the main dip (see [12, 13]) and its narrow structures [16] on the strong field intensity behave differently. When the constant ratio is $\Gamma_n \ll \Gamma_m$, the narrow structure amplitudes depend nonlinearly on the saturation parameter κ_s with a maximum at values $\kappa_s \approx 1 \div 2$ [16]. In this case, the maxima of structure contrasts are realized at values $\kappa_s \approx 0.5 \div 0.7$.

The width dependences of the narrow resonance structures on the strong field intensity are significantly different for open and closed transition types. At the open transition the half-width of the dip structure changes linearly with saturation parameters κ_s (at $\kappa_s \leq 1$) as: $\Gamma_{\mathrm{w}} \approx \Gamma_n(1+\kappa_s)$.

The width growth of this structure, close to linear, is observed in the given above numerical calculations at the strong field saturation parameters $\kappa_s \ll \sim 1$. At the closed transition, the half-width dependence of the peak structure has the other form: $\Gamma_{\mathrm{w}} = \Gamma_n(1+\kappa_s)/(1+2\kappa_s)$. Here, the growth of parameter values in the range of $\kappa_s = 0 \div 1$ leads to the reduction of the structure half-width in the range: $\Gamma_w = (1 \div 0.7)\,\Gamma_n$.

The absence of the strong-field intensity influence on the narrow peak structure width in the calculations under conditions of Figure 2 is caused by the insufficient spectral resolution. At an increase in the spectral resolution ($\sim$ by an order of magnitude) in calculations, a narrowing of the peak structure with growth of the strong field intensity was registered.

3.2.2. The Resonance Shape in the Three-Level System

In the system of three splited levels (Figure 1), the nonlinear resonance shape can be obtained from the set of equations (3-5) in the approach of a weak probe field and disregarding polarization at the combination frequency in the form:

$$\alpha(\varepsilon)/\alpha_0 = \Gamma_{mn}\,\mathrm{Re}\left\langle \frac{\Gamma_{21}-i(\omega_{12}+\varepsilon)}{(\Gamma_{mn}-i\Omega_{1\mu})\left[\Gamma_{21}-i(\omega_{12}+\varepsilon)\right]+|G|^2}\left[\begin{array}{c}(\rho_0^0-\rho_2^0)-\\[4pt]-\dfrac{|G|^2(\rho_0^0-\rho_1^0)}{(\Gamma_{21}-i(\omega_{12}+\varepsilon))(\Gamma_{mn}+i\Omega_1)}\end{array}\right]\right\rangle, \tag{10}$$

where ρ_0^0, ρ_1^0 and ρ_2^0 are determined from the system (3-5) solutions at only the strong field action as [12]: $\rho_0^0 - \rho_1^0 = N_{nm}\left[1-\dfrac{\kappa_s\Gamma_{mn}^2}{\Gamma_s^2+\Omega_1^2}\right]$, $\rho_1^0 = N_m + \left[\dfrac{N_{nm}}{\Gamma_m T_{mn}}\dfrac{\kappa_s\Gamma_{mn}^2}{\Gamma_s^2+\Omega_1^2}\right]$, $\rho_2^0 = N_m$,

$\Gamma_s = \Gamma_{mn}\sqrt{1+k_s}$, $\kappa_s = 2|G|^2 T_{mn}/\Gamma_{mn}$, $T_{mn} = \gamma_{mn}/\Gamma_n\Gamma_m$, $\gamma_{mn} = \Gamma_m - A_{mn} + \Gamma_n$, $N_{nm} \equiv N_n\text{-}N_m$.

In this case, when averaging over the velocities of

expression (10), the first term forms the Doppler absorption line contour, and the features of the resonance spectrum, both in unidirectional and counter-waves, will be determined by the second (coherent) term.

In case of the unidirectional waves from expression (10) at a large Doppler broadening $\Gamma_{mn}\sqrt{1+k_s} \ll kv_{\mathrm{T}}$ and the field frequency detunings of $\Omega_\mu \ll kv_{\mathrm{T}}$, $\Omega \ll kv_{\mathrm{T}}$, the resonance shape is as follows:

$$\alpha(\delta)/(\alpha_0 N_{nm}) = \sqrt{\pi}\,\frac{\Gamma_{mn}}{kv_{\mathrm{T}}}\left\{1-2|G|^2\,\mathrm{Re}\left[\frac{\Gamma_{mn}(\Gamma_m-A_{mn})(\Gamma_m-i\delta)}{\Gamma_m\Gamma_n\Gamma_s\Delta_1}+\frac{1-\beta_V}{\Delta_2}\right]\right\}, \tag{11}$$

where $\delta = \varepsilon + \omega_{12} = \Omega_{20}^\mu - \Omega_{10}$; $\Delta_1 = (\Gamma_m-i\delta)\left[(\Gamma_s+\Gamma_{mn})-i\delta\right]+|G|^2$;

$\Delta_2 = (\Gamma_m-i\delta)\left[2\Gamma_{mn}-i\delta\right]+|G|^2$;

$\beta_V = \dfrac{\kappa_s\Gamma_{mn}^2}{\Gamma_s^+ + i\delta}\left[\dfrac{1}{(\Gamma_s^+ - i\delta)}-\dfrac{(\Gamma_{mn}^1-i\delta)}{2\Gamma_s\Gamma_s^0}\right]$;

$\Gamma_s^\pm = \Gamma_s \pm \Gamma_{mn}^1$; $\Gamma_s^0 = \Gamma_s + \Gamma_{mn}$; $\Gamma_{mn}^1 = \Gamma_{mn}+\dfrac{|G|^2}{\Gamma_m-i\delta}$.

Here, the determinants Δ_1 and Δ_2 determine the field dependence of the resonance structure widths, including conditions of the spectral splitting of these structures into

components. The value of β_V is small; at frequency detunings $\delta < \Gamma_{mn}$ this value is approximated as $\beta_V \approx \kappa_s/8$ for small saturation parameters $\kappa_s < 1$, and as $|\beta_V| < 0.5$ for $\kappa_s \gg 1$. Therefore, in a wide range of saturation parameters, including those in the range of $\kappa_s \sim 1$, the contribution of this term to the resonance shape can be neglected.

It follows from (11) that the resonance is formed by the contributions of two terms: the first term in square brackets is due to the saturation of the level populations by a strong field and is of incoherent character, and the second term is due to the coherent processes in the system of three transition levels. At the closed transition (at $\Gamma_m = A_{mn}$), the contribution of the population term is absent, and the nonlinear resonance shape will be determined only by coherent processes.

In the approach of the first nonlinear corrections on the strong field, the resonance shape from (11) takes the form:

$$\alpha(\delta)/(\alpha_0 N_{nm}) = \sqrt{\pi}\frac{\Gamma_{mn}}{kv_T}\left\{1-2|G|^2\operatorname{Re}\left[\frac{\Gamma_m - A_{mn}}{\Gamma_m\Gamma_n}\frac{1}{2\Gamma_{mn}-i\delta} - \frac{1}{2\Gamma_{mn}-\Gamma_m}\left(\frac{1}{2\Gamma_{mn}-i\delta} - \frac{1}{\Gamma_m-i\delta}\right)\right]\right\}, \tag{12}$$

In this case, the nonlinear resonance shape is formed by the sum of three Lorentzians with different amplitudes and widths and is centered at the same frequency (at $\delta = 0$). The first Lorentzian represents a contribution of the incoherent saturation process of the level populations and the second, and third Lorentzians are due to the contributions of two coherent two-quantum processes: the step and two-photon absorption [12, 13], respectively, in the three-level transition system.

As noted above, for the closed transition (at $\Gamma_m = A_{mn}$), the contribution of the incoherent process in (12) is absent, and the nonlinear resonance shape is determined by the sum (interference) of contributions of the step and two-photon processes. It represents a purely nonlinear interferential effect (NIEF) in the three-level V-scheme of transition [12]. In this case, the nonlinear resonance manifests itself as a dip near the detuning of frequencies $\delta = 0$ (result of domination of the two-photon absorption in this frequency region) and two small-amplitude peaks equally spaced from the line center at values $\delta = \pm\sqrt{3}\Gamma_m$. Similar nonlinear resonance shapes are observed in Figure 5 at small saturation parameters $\kappa_s \leq 0.02$ (curves 1–4).

With an increase in the degree of transition openness (reduction of value a_0), the peak amplitudes decrease and they are not shown in the resonance form at $a_0 \leq 0.9$. In this case, when forming a resonance spectrum, according to (12), the contribution of the incoherent saturation process of level populations by a strong field is crucial and it leads to the traditional dip with a half-width of $2\Gamma_{mn}$.

Let us consider the strong field intensity effect on a resonance shape at the closed transition in more detail. In this case, the resonance shape is caused by coherent processes and, at saturation parameters $\kappa_s \leq 1$, it is representable from (11) as follows:

$$\alpha(\delta)/(\alpha_0 N_{mb}) = \sqrt{\pi}\frac{\Gamma_{mn}}{kv_T}\left\{1-2|G|^2\operatorname{Re}\left[\frac{i}{2\delta_0}\left(\frac{1}{\Gamma_+ -i\delta_-} - \frac{1}{\Gamma_+ -i\delta_+}\right)\right]\right\}, \tag{13}$$

where: $\delta_0 = \left|\sqrt{|G|^2 - \Gamma_-^2}\right|$; $\delta_\pm = \delta \pm \delta_0$; $\Gamma_\pm = \Gamma_{mn} \pm \Gamma_m/2$.

It follows from (13) that the strong field leads to a change in the widths of spectrum structures at values $|G|^2 - \Gamma_-^2 < 0$ and to a splitting of the spectrum structures into components at values $|G|^2 - \Gamma_-^2 > 0$. When the level relaxation constants are $2\Gamma_{mn} = \Gamma_m + \Gamma_n$, the magnitude δ_0 is determined as $\delta_0^2 = |G|^2 - \Gamma_n^2/4$, and the spectral splitting will be observed at saturation parameters $\kappa_s > \kappa_r = \Gamma_n^2/(2\Gamma_{mn}\Gamma_m)$.

At the ratio of constants $\Gamma_n \ll \Gamma_m$, the value of κ_r is determined as $\kappa_r = \Gamma_n^2/\Gamma_m^2 \ll 1$. It means that the spectral splitting in the considered transition scheme should occur already at small strong field saturation parameters.

At values of parameters $\kappa_s < \Gamma_n^2/(2\Gamma_{mn}\Gamma_m)$ $(4|G|^2 < \Gamma_n^2)$, the resonance shape can be represented from (13) as the difference of two Lorentzians with different amplitudes and widths which are centered at the one frequency:

$$\alpha(\delta)/(\alpha_0 N_{nm}) = \sqrt{\pi}\frac{\Gamma_{mn}}{kv_T}\left\{1+2|G|^2\operatorname{Re}\left[\frac{1}{2\delta_1}\left(\frac{1}{\Gamma_+ + \delta_1 - i\delta} - \frac{1}{\Gamma_+ - \delta_1 - i\delta}\right)\right]\right\}, \tag{14}$$

where $\delta_1 = \left|\sqrt{\Gamma_n^2/4 - |G|^2}\right|$. At small saturating fields (at $\Gamma_n^2 \gg 4|G|^2$), δ_1 is approximated as $\delta_1 \approx \Gamma_n(1 - 2|G|^2/\Gamma_n^2)/2$, and the resonance shape takes the form:

$$\alpha(\delta)/(\alpha_0 N_{nm}) = \sqrt{\pi}\frac{\Gamma_{mn}}{kv_T}\left\{1+\frac{2|G|^2/\Gamma_n}{1-2|G|^2/\Gamma_n^2}\operatorname{Re}\left[\left(\frac{1}{2\Gamma_{mn}-|G|^2/\Gamma_n-i\delta} - \frac{1}{\Gamma_m+|G|^2/\Gamma_n-i\delta}\right)\right]\right\} \tag{15}$$

Herein, the first term in (15) describes the stepwise absorption process of two photons in the three-level system and forms a peak structure in the line contour, while the second term is caused by the two-photon absorption process and forms a dip. An increase in the saturating field intensity leads to a broadening of the two-photon structure and to a narrowing of the stepped spectral structure, as well as to the growth of the amplitudes of these structures under the linear law. In this case, the absorption line contour as a result of the addition (interference) of two coherent processes reflects (according to [12]) the nonlinear interference effect (NIEF) in the considered level system.

In case of saturation parameters $\kappa_s > \Gamma_n^2 / (2\Gamma_{mn}\Gamma_m)$ ($4|G|^2 > \Gamma_n^2$) the nonlinear resonance shape is as follows:

$$\alpha(\delta)/(\alpha_0 N_{nm}) = \sqrt{\pi} \frac{\Gamma_{mn}}{kv_T} \left\{1 - \frac{|G|^2}{\delta_0}\left[\frac{\delta + \delta_0}{\Gamma_+^2 + (\delta + \delta_0)^2} - \frac{\delta - \delta_0}{\Gamma_+^2 + (\delta - \delta_0)^2}\right]\right\}, \tag{16}$$

In this case, the resonance is formed by the difference of two dispersive counters, that are displaced at value of $2\delta_0$, where $\delta_0 = \left|\sqrt{|G|^2 - \Gamma_n^2/4}\right|$ determines the field splitting of the lower level. At the same time, the resonance is represented as a dip in the center (at $\delta = 0$) and maxima in the line wings at

$$\delta = \pm\sqrt{\Gamma_+^2 + \delta_0^2} = \pm\sqrt{\Gamma_m(\Gamma_m + \Gamma_n) + |G|^2} . \tag{17}$$

The dip amplitude depends linearly on the strong field intensity. The resonance width, as a distance between the maxima from (17), has a linear relation at saturation parameters $\kappa_s <\sim 1$ and acquires a root relation for large values of $\kappa_s \gg 1$. Let us note that the linear relations of amplitude and width of coherent resonance are observed in the calculated resonance shapes at a change of the saturation parameter up to values $\kappa_s <\sim 10$.

Thus, this resonance on a closed transition has an exclusively coherent nature, while the wings of the resonance counter contain the structures characteristic for coherent processes due to the interference of two-quantum processes (NIEF) and to the level splitting effect by a strong field.

4. Conclusion

Thus, the presented results of studying the shapes of the saturated absorption resonances on $^1S_0$-$^{1,3}P_1$ transitions, as from the ground and excited atom states, show that the nonlinear resonance spectra, as well as the processes forming them in the unidirectional waves, depend on the level relaxation constants, value of level splitting and, especially, on the nature (degree of openness) of the atomic transition. Moreover, the influence of relaxation constants on the nonlinear resonance shape has a quantitative character, and the influence of the level splitting and, especially, the degree of the transition openness is manifested in a qualitative way.

In the absence of the upper level splitting, a nonlinear resonance occurs at the frequency of atomic transition shaped as a traditional dip and is formed mainly by noncoherent population processes in two-and three-level transition schemes. Coherent processes are weakly expressed and manifest themselves only as a resonance at the $^1S_0$-$^{1,3}P_1$ closed transition (resonant or intercombinational transition from the ground to the first excited state). When level splitting exceed the transition line width, the resonances are formed in two-and three-level schemes are separated in their frequency and are shown in the pure shape.

In case of unidirectional waves in the absorption spectrum of a probe wave in the system of two levels, the wide population dip and the narrow structure with the lower level width, caused by the coherent beats of level populations, are formed. Moreover, at the resonant (closed) transition, the narrow structure appears as peak-shaped, and on transitions to overlying levels (open transitions), the structure appears as dip-shaped. The nonlinear resonance shape in the system of three levels also turns out to be dependent on the nature of transition. At the open transition, the resonance is due mainly to the incoherent saturation process of level populations and it manifests itself as a traditional dip, and, at the closed transition, the resonance is formed exclusively by coherent processes and has an interference shape.

At the end of article, we will note one more feature of the closed transitions $^1S_0$-$^{1,3}P_1$ in case of two counter-waves. In this case, in the system of three levels a cross-resonance is formed. In case of an open transition, the cross-resonance manifests itself as a dip and has an incoherent population pattern; and, in case of a closed transition, it appears as a peak and has an exclusively coherent nature with the interference spectrum structure [18]. Here, a situation is possible when the saturated absorption resonance at a closed transition will be represented only by the cross-coherent resonance.

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