Interference suppression of steady-state absorption with increasing radiative power

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We consider nonlinear interference between polarizations induced by resonant monochromatic radiation field in a multilevel system consisting of a ground level split into N sublevels and a nondegenerate excited level. These manifestations have not been discussed previously. This study was performed analytically and on the basis of the numerical calculations. We show that in the absence of collisions in such a system, the magnitude of the energy absorbed reaches a maximum and then falls to zero inversely with the radiative intensity I as the latter increases. For $N \geqslant 4$, the absorption line width near the maximum is smaller than the width for $I \to 0$ by $\leqslant 6\%$. For small splitting Δ, the effect occurs at intensities a factor of γ_1/Δ less than the saturation intensity, where γ_1 is the radiative decay rate of the upper state. The resulting medium bleaching is described by a square root law of beam intensity attenuation with increasing optical depth. Collisions constrain this effect, producing nonzero absorption at large I, and they split the absorption line if the Einstein coefficients of the optical transitions are equal.

1. INTRODUCTION

Various intraatomic coherence phenomena produced by laser radiation in simple schemes of interaction between atoms and laser radiation have been extensively discussed in the literature in recent years. The subject of these investigations has been the interference of probability amplitudes of allowed (optical in most cases) transitions which are coupled through the field-induced polarization of forbidden transitions. Special attention has been paid to three-level Λ-systems with ground and intermediate metastable states.

It has been shown in numerous experimental and theoretical works[1–11] that destructive interference may occur in a Λ-system and in similar more complicated quantum systems, which results under steady-state conditions in the creation of special superposition states[7,12–14] in which light absorption is absent altogether. The population of the upper level combining with the field and the polarizations of the allowed optical transitions are zero in this case, and the population is captured, or trapped, at the lower levels, thus leading to coherent bleaching of the medium.

This phenomenon, termed coherent population trapping, is of a universal nature, and has a great number of practical consequences. For instance, when two-frequency radiation is used in the vicinity of a two-photon resonance, strong dips appear in the absorption and excitation spectra, whose width is less than the homogeneous line widths of the optical transitions;[2,15–17] cooling atoms with light[18] becomes possible down to superlow temperatures, $T \sim 10^{-6}$ K;[19–22] in the process of propagation of CW laser radiation[23,24] or an ultrashort pulse train[25] under the conditions of coherent population trapping, the beam is attenuated not in accordance with the exponential Bouguer–Lambert law but linearly or in some other manner.[24] The list presented can be considerably extended, and a special review could be devoted to coherent population trapping.

However, the effect considered in this paper differs in principle from the phenomenon of coherent population trapping. The point is that situations are possible in which this phenomenon is lacking in Λ- and similar systems, but nonlinear interference effects[26,27] due to field-induced polarization of forbidden transitions and the resulting coupling between polarizations of allowed transitions are realized in full measure. Specifically, in Ref. 28, giant magnitudes of interference shift and line widths of steady-state absorption and fluorescence excitation spectra were theoretically predicted for monochromatic radiation simultaneously resonant with two nearby allowed optical transitions from a doublet ground state to an excited level. In Ref. 29, this analysis was generalized to nonmonochromatic radiation modulated in amplitude and phase by a random totally disconnected Markov process.

One more nontrivial consequence of nonlinear polarization interference consisting in nonlinear dependence of steady-state absorption line width and shift on radiative intensity at sufficiently high intensities of the latter was obtained in Refs. 28 and 29, but was not specially discussed. Such a dependence differs drastically from the familiar square-root intensity dependence of the width taking place under the conditions of saturation of resonant light absorption by a two-level system,[30] with the result that the behavior of the absorbed energy E_{abs} with increasing intensity is qualitatively different. In the absence of collisions, E_{abs} does not tend to a constant value (saturation of absorption), but reaches a maximum and then falls to zero inversely with the intensity. Analysis of the corresponding formulas in Refs. 28 and 29 shows that nonlinear interference effects suppress absorption uniformly with respect to radiation frequency detuning from resonance as the intensity grows. Thus, nonlinear interference of polarizations also leads to bleaching of the medium, as in the case of coherent population trapping, but the mechanism of its action and the peculiarities of its realization are different,

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in spite of the common nature of the two phenomena.

The purpose of this paper is to consider the indicated manifestation of nonlinear interference effects for quantum systems with a multiply-split ground state and a nondegenerate upper level interacting with resonant monochromatic radiation in a steady-state mode.

2. STATEMENT OF THE PROBLEM

Let us consider a quantum system consisting of an upper state (1) and a ground state (0) split into $N \geqslant 2$ sublevels. Assume that $\tilde{N}$ sublevels of the ground state are coupled to the excited state by dipole-allowed transitions, whereas such transitions from the remaining $\bar{N}=N-\tilde{N}$ sublevels are forbidden. The splitting will be considered small as compared to the frequency ω_{10} of the optical transition between the lower sublevel of the ground state and the upper state, so radiative relaxation of the split sublevel will be ignored and only collisional population redistribution between them will be taken into account. A monochromatic field of frequency $\omega \simeq \omega_{10}$ and strength $\mathscr{E}$ will be considered to interact with all allowed optical transitions of the system.

The equations for the density matrix of the given system for the steady-state case in the rotating wave approximation, the interaction representation, and the model of relaxation constants have the form

$$\begin{aligned}
&\gamma_1\rho_1=2\sum_l V_lR_l'',\\
&\nu_p\tilde{\rho}_i-\gamma_p\sum_{l\neq i}{}'\tilde{\rho}_l-\gamma_z\sum_l\bar{\rho}_l=A_i\rho_i-2V_iR_i'',\\
&\nu_z\bar{\rho}_j-\gamma_z\sum_{l\neq j}{}'\bar{\rho}_l-\gamma_p\sum_l\tilde{\rho}_l=0,\\
&\rho_1+\sum_l\tilde{\rho}_l+\sum_l\bar{\rho}_l=1,\\
&\left[\frac{1}{2}\gamma_1+\Gamma_i-i(\Omega_i-\delta_i)\right]R_i=iV_i(\tilde{\rho}_i-\rho_1)+i\sum_{l\neq i}{}'V_lr_{li},\\
&[\Gamma_{ij}+i(\Delta_{ij}+\delta_{ij})]r_{ij}=i(V_iR_j-V_jR_i^*).
\end{aligned}\tag{1}$$

Here $R_i = R_i' + iR_i''$ are the off-diagonal elements of the density matrix (polarizations) of the allowed optical transitions, r_{ij} are the polarizations of the low-frequency forbidden transitions between the sublevels i and j of the ground state (the transitions from these sublevels to the excited state are allowed), ρ_1 is the population of the excited state, $\tilde{\rho}_i$ and $\bar{\rho}_j$ are the populations of the ground state sublevels, from which the transitions to the state 1 are respectively allowed and forbidden,

$$V_i=V_i^*=d_{1i}\mathscr{E}/2\hbar$$

are the Rabi frequencies of the allowed optical transitions, d_{1i} are the matrix elements of the dipole moment of these transitions, $\Omega_i=\omega-\omega_{10}+E_i/\hbar$ are the frequency offsets, E_i are the energies of the ground state sublevels, A_i are the Einstein first coefficients for the allowed optical transitions, $\gamma_1=\Sigma_l A_l$ is the radiative decay constant of the upper state, ν_p, ν_z and γ_p, γ_z are the population and the depopulation rates, which are assumed equal for all the splitting sublevels, from which the optical transitions are respectively allowed and forbidden, Γ_i, Γ_{ij} and δ_i, δ_{ij} are the collisional broadening and the shift constants, respectively, for the optical and the forbidden low-frequency transitions, $\Delta_{ij}=(E_i-E_j)/\hbar$ are the natural frequencies of the forbidden low-frequency transitions, or the splitting between the ground state sublevels. The summation of R_i'' and $\tilde{\rho}_i$ is performed from 1 to $\tilde{N}$, the summation of r_{ij} and $\bar{\rho}_j$ is performed from 1 to $\bar{N}$.

We seek the upper state population ρ_1. The intensity of the fluorescence excitation spectrum and, as is easy to show on the basis of (1), the coefficient of field steady-state absorption by the system under consideration, are proportional to ρ_1. By the term "line profile" in what follows, we mean the dependence $\rho_1(\Omega)$.

From the condition of the conservation of the total population of the system at $V_i=0$, $i=1-\tilde{N}$, and Eqs. (1), the relation for the collisional relaxation constants of the levels is

$$[\nu_p-(\tilde{N}-1)\gamma_p][\nu_z-(\bar{N}-1)\gamma_z]=\tilde{N}\bar{N}\gamma_p\gamma_z. \tag{2}$$

Using (2) and eliminating $\bar{\rho}$ and r_{ij} from (1), we transform the original set of equations:

$$\begin{aligned}
&A_i\rho_i-2V_iR_i''=\nu[\tilde{\rho}_i-\beta(1-\rho_1)],\\
&(\tilde{\Gamma}_i-i\tilde{\Omega}_i)R_i-V_i\sum_{l\neq i}{}'V_lD_{li}^*R_l^*=iV_i(\tilde{\rho}_i-\rho_1),\\
&\rho_1+\frac{1}{\beta\tilde{N}}\sum_i\tilde{\rho}_i=1,\\
&\gamma_1\rho_1=2\sum_l V_lR_l'',
\end{aligned}\tag{3}$$

where

$$\begin{aligned}
&\tilde{\Gamma}_i=\frac{1}{2}\gamma_1+\Gamma_i+\sum_{l\neq i}{}'V_l^2D_{li}',\\
&\tilde{\Omega}_i=\Omega+\frac{E_i}{\hbar}-\delta_i+\sum_{l\neq i}{}'V_l^2D_{li}'',\quad \Omega=\omega-\omega_{10},\\
&D_{ij}=D_{ij}'+iD_{ij}''=1/[\Gamma_{ij}-i(\Delta_{ij}+\delta_{ij})],\\
&\beta=\gamma_z/[\nu+\tilde{N}(\gamma_z-\gamma_p)],\quad \nu=\nu_p+\gamma_p.
\end{aligned}\tag{4}$$

Either the last or the next-to-last equation in (3) is redundant, and the use of one or the other depends on the relation between the radiative and collisional relaxation constants.

It can be seen from (1), (3), and (4) that the complex matrix D_{ij}^*, whose nature derives from the polarizations of the forbidden low-frequency transitions r_{ij}, couples the polarizations of the allowed transitions R_i and thus completely determines the nonlinear interference effects in the multilevel system under consideration. The possibility of neglecting this coupling in studying multilevel systems is by no means evident, contrary to the widespread opinion on mutual compensation of the contributions of intraatomic coherence, whereupon only population effects are taken into account. On the contrary, as will be shown in subsequent sections, interference of the polarizations leads, under certain conditions, to unexpected and quite appreciable consequences.

Anticipating the actual calculations, let us write out these conditions, having compared the first and second terms in the second of Eqs. (3). With the estimate

$$\Gamma_i\sim\Gamma_{ij}\sim\Gamma,\quad \Delta_{ij}\sim\Delta,\quad V_i\sim V,\quad \delta_i\sim\delta_{ij}\sim 0,$$

we assume that the contributions of interference add without cancelling one another. Based on (3) and (4), the ratio of the second and first terms can then be represented by the parameter

$$\varkappa_{\rm coh}=\frac{\tilde{N}V^2}{(\gamma_1+2\Gamma)\max(\Gamma,\Delta)}. \tag{5}$$

Nonlinear interference effects obviously become significant for $\varkappa_{\rm coh}\gtrsim 1$. If collisional relaxation prevails ($\Gamma\gg\gamma_1,\Delta$), then the condition $\varkappa_{\rm coh}\gtrsim 1$ reduces to $\tilde{N}V^2/\Gamma^2\gtrsim 1$. As will be shown in Sec. 4, this relation imposes constraints on the field magnitude V^2 that are Γ/γ_1 times more stringent than necessary to saturate the multilevel system considered. In the absence of collisions ($\Gamma=0$),

$$\varkappa_{\rm coh}=\tilde{N}V^2/(\gamma_1\Delta),$$

and nonlinear interference effects may appear at a field intensity lower than the saturation intensity by a factor of $\gamma_1/\Delta\gg 1$, if the effective mean frequency difference between the splitting sublevels is much less than the radiative line widths of the optical transitions, $\Delta\ll\gamma_1$. Physical factors restricting the effect as $\Delta\to 0$ were discussed in Refs. 28 and 29.

Note that for nonlinear interference effects to manifest themselves, only two closely spaced levels suffice in general, but the parameter $\varkappa_{\rm coh}$ may then be $\tilde{N}$ times smaller, in principle.

The meaning of the parameter β in the chosen model of collisional population redistribution is easily understood if we make an additional simplifying assumption: $\nu_p=\nu_z$, $\gamma_p=\gamma_z$. Using the definition of β in (4) and relation (2), we get

$$\beta=1/(\tilde{N}+\sqrt{\bar{N}\tilde{N}}).$$

In the special cases $\bar{N}=0$ and $\tilde{N}=\bar{N}$, the magnitude of β is equal to $1/\tilde{N}$ and $1/2\tilde{N}$, respectively. The factor $1/\beta\tilde{N}$ multiplying the sum in the third of Eqs. (3) characterizes the fraction of the total population of the system corresponding to all lower states of allowed optical transitions. For the special cases cited above,

$$1/\beta\tilde{N}=\begin{cases}1, & \bar{N}=0,\\ 2, & \tilde{N}=\bar{N}.\end{cases}$$

For $\tilde{N}\ll\bar{N}$, we have $1/\beta\tilde{N}=(\bar{N}/\tilde{N})^{1/2}\gg 1$.

3. STEADY-STATE ABSORPTION UNDER COLLISIONLESS CONDITIONS

In the absence of collisions,

$$\Gamma_i=\Gamma_{ij}=\delta_i=\delta_{ij}=\nu=D'_{ij}=0,\quad D''_{ij}=1/\Delta_{ij},$$

$$\tilde{\Gamma}_i=(1/2)\gamma_1,\quad \tilde{\Omega}_i=\Omega+E_i/\hbar+\sum_{l\neq i}{}'V_l^2/\Delta_{li}.$$

Sublevels of the ground state not coupled to state 1 by allowed transitions drop out of consideration, so we should set $\tilde{N}=N$, $\beta=1/N$ in this Section. Since $A_i\propto d_{1i}^2\omega_{1,0i}^3$ (see, for instance, Ref. 31) and $|\Delta_{ij}|\ll\omega_{10}$, we have

$$V_i^2/A_i\equiv\mathscr{V}=\text{const}$$

to high precision. Using the relations presented, we get an exact solution of system (3):

$$\rho_1=(\gamma_1\mathscr{V}/N)/[(\Omega-\Omega_0)^2+\Gamma_0^2],$$

$$\Omega_0=-\frac{1}{N}\sum_i\left(E_i+\mathscr{V}\sum_{l\neq i}{}'\frac{A_l}{\Delta_{li}}\right),$$

$$\Gamma_0^2=\frac{1}{4}\gamma_1^2+2\gamma_1\frac{\mathscr{V}}{N}+\frac{1}{N}\sum_i E_i^2-\Omega_0^2+\frac{2\mathscr{V}}{N}\times\sum_i E_i\sum_{l\neq i}{}'\frac{A_l}{\Delta_{li}}+\frac{2\mathscr{V}^2}{N}\sum_i\left(\sum_{l\neq i}{}'\frac{A_l}{\Delta_{li}}\right)^2, \tag{6}$$

$$\hbar=1.$$

The dependence $\rho_1(\Omega)$ (6) has a Lorentzian shape with center frequency Ω_0 and half-width Γ_0. As in the case of a three-level Λ-system,[28,29] the quantities Ω_0 and Γ_0 for high radiative power levels are proportional to the intensity:

$$I=(c/8\pi)\varepsilon^2=(c\hbar A_i/2\pi d_{1i}^2)\mathscr{V}.$$

It is easy to show that this is entirely due to nonlinear interference effects.

The sums in (6) can be calculated analytically by invoking the model of equidistant lines of equal intensities (the Elsasser model[32]) used in atmospheric optics. According to this model,

$$A_i=\gamma_1/N,\quad V_i=V,\quad E_i=\Delta(i-1),\quad \Delta_{ij}=\Delta(i-j),$$

$$d_{1i}=d,\quad \mathscr{V}=NV^2/\gamma_1.$$

The corresponding calculations give

$$\rho_1 = V^2/[(\Omega-\Omega_0)^2+\Gamma_0^2],$$

$$\Omega_0 = -\frac{1}{2}(N-1)\Delta,$$

$$\Gamma_0^2 = \frac{1}{4}\gamma_1^2 + \frac{1}{12}(N^2-1)\Delta^2 - (N-3)V^2 + 8S_N\frac{V^4}{\Delta^2}, \quad (7)$$

$$S_N = \frac{1}{N}\sum_{n=1}^{N-1}\left(\sum_{k=n}^{N-n}\frac{1}{k}\right)^2,$$

$$S_2=\frac{1}{2}, \quad S_3=\frac{3}{2}, \quad S_4=\frac{251}{144}\approx 1.743,$$

$$S_5=\frac{145}{72}\approx 2.014,\ldots, \quad S_\infty = 2\zeta(2)\approx 3.290,$$

where $\zeta(n)$ is the Riemann zeta function. For $N=2$, expressions (7) coincide with formulas (2b) in Ref. 28 at $V_1=V_2$. Note that for systems with equidistant splitting sublevels, the interference contributions at the line center cancel completely. Thus, the location of the center becomes independent of the field intensity. At the same time, there is no such cancellation for the line width; moreover, the interference contribution to the width (the last term in the expression for Γ_0^2 (7)) grows approximately sixfold with increasing sublevel number, up to $8S_\infty V^4/\Delta^2$ for $N \gg 1$.

Remarkably, the next-to-last term in the expression for Γ_0^2 (7) (linear in intensity), which is responsible for ordinary power broadening of the line[30] for $N=2$, vanishes at $N=3$ and becomes negative at $N \geqslant 4$. As a consequence, in a system with four or more sublevels of the ground state, interference line narrowing, which was not noted before, is possible with increasing field intensity. The half width Γ_0 is smallest at

$$V^2 = V_{\min}^2 = \frac{1}{16}\frac{(N-3)\Delta^2}{S_N}, \quad N \geqslant 4, \quad (8)$$

and equals

$$(\Gamma_0)_{\min} = \frac{1}{2}\left\{\gamma_1^2 + \frac{1}{3}\Delta^2\left[N^2-1-\frac{3}{8}\frac{(N-3)^2}{S_N}\right]\right\}^{1/2}. \quad (9)$$

The largest relative narrowing $[\Gamma_0(V=0)-(\Gamma_0)_{\min}]/\Gamma_0(V=0)$ occurs for $\Delta^2 \gg 3\gamma_1^2$ and ranges from 0.7% for $N=4$ to 5.9% for $N \gg 1$, i.e., it increases with N.

In accordance with the qualitative behavior of the maximum energy absorbed as a function of the radiative intensity described in Sec. 1, the quantity $\rho_1(\Omega=\Omega_0)\equiv\rho_{1m}{}^{(7)}$ grows linearly with intensity at small V,

$$I = \frac{c\hbar^2}{2\pi d^2}V^2 = \frac{2N\hbar\omega}{3\pi\gamma_1\lambda\!\!\!^{-2}}V^2, \quad \lambda\!\!\!^{-} = c/\omega, \quad (10)$$

reaches a maximum

$$\rho_{1m} = \rho_{1\,\max} = \left\{\frac{2}{\Delta}\left[2S_N\left(\gamma_1^2+\frac{\Delta^2}{3}\right)\right]^{1/2} - N+3\right\}^{-1} \quad (11)$$

at

$$I = I_{\max} = \frac{N\hbar\omega\Delta}{6\pi\gamma_1\lambda\!\!\!^{-2}}\left(\frac{\gamma_1^2+\Delta^2/3}{2S_N}\right)^{1/2}, \quad (12)$$

and then falls to zero as the reciprocal of the intensity.

Let us compare $I_{\max}$ (12) with the saturation intensity $I_{\rm sat}$ for the case of light absorption by a two-level system, given by

$$V_{\rm sat}^2 = \frac{3\pi\gamma_1\lambda\!\!\!^{-2}}{2N\hbar\omega}I_{\rm sat} = \frac{1}{2}\gamma_1^2. \quad (13)$$

We have

$$\frac{I_{\max}}{I_{\rm sat}} = \frac{\Delta}{2\gamma_1^2}\left(\frac{\gamma_1^2+\Delta^2/3}{2S_N}\right)^{1/2}. \quad (14)$$

It can be seen from (14) that if the separation between neighboring sublevels is less than the radiative line widths of allowed transitions ($\Delta^2 \ll 3\gamma_1^2$), then

$$\frac{I_{\max}}{I_{\rm sat}} = \frac{1}{2\sqrt{2S_N}}\frac{\Delta}{\gamma_1} \ll 1. \quad (14')$$

The comparison of (14′) with the estimates of the extent of the manifestation of nonlinear interference effects made in Sec. 2 on the basis of formula (5) for $\varkappa_{\rm coh}$ shows that these effects are N times weaker in a multilevel system than the estimate involving the parameter $\varkappa_{\rm coh}$; however, it dominates under the conditions specified.

Now let us follow the effects of unequally spaced sublevels and scatter among the Einstein coefficients for different transitions on interference suppression of absorption. With this purpose, we performed numerical calculations according to Eqs. (6) for $N=20$, in which $\Delta_{i+1,i}$ and A_i were assumed to be random quantities with various prescribed probability distributions. The half-width Γ_0 and the values of ρ_{1m} for the center of the average profile, whose position coincided with Ω_0 (7) to high accuracy, were averaged over 1000 realizations. This was sufficient for the rms deviation of Γ_0 and ρ_{1m} not to exceed a few percent. The deviation of the line center position, on the other hand, was more than ten percent, suggesting a high sensitivity of Ω_0, which is subject to considerable interference shifts,[28] to chance coincidences of the energies of split sublevels, resulting in a sharp increase in the polarization coupling parameter. Numerical results are presented in Fig. 1, where the results for equidistant lines of equal intensities [curves *1*, Eqs. (7)] and for $D_{ij}=0$, where nonlinear interference effects are absent (curves *9*), are also given for the sake of comparison.

A comparison of curves *2*, *4*, and *6* in Fig. 1a with curve *1*, illustrating the effect of $\Delta_{i+1,i}$ for preset equal constants of excited state radiative decay, shows that an increase in the spread results in a decrease in the maximum amplitudes ρ_{1m} and $\rho_{1\max}$ independent of I, as well as a shift of $\rho_{1\,\max}$ towards lower intensities. This behavior of $\rho_{1m}(I)$ can be explained in a natural way by an increase in the fraction of small splittings $\Delta_{i+1,i}$, for which interference affects the line profile most noticeably. The addition of the spread in the A_i values to this factor further enhances the effect, but less than the spread in the splitting (see curves 2–7 in Fig. 1a). The suppression of absorption is most significant for the exponential probability distribution of $\Delta_{i+1,i}$. In this case, the relative contribution of small splittings is greater than for rectangular distributions (curve 8 in Fig. 1a). The spread in the splitting values and Einstein coefficients leads to the smearing of interference power line narrowing. This smearing becomes stronger as the spread increases, and eventually the narrowing is completely eliminated. If so, stronger line broadening occurs with increasing intensity, particularly in comparison with the case when interference is absent. This can be illustrated by the ratio of the slopes of curves 8 and 9 in Fig. 1b, which can reach $\simeq 100$. In other words, in the presence of a sufficiently large number of close sublevels of the ground state, power broadening of the line with regard to polarization interference may be two orders of magnitude greater than in the presence of the population saturation effect alone.

An important consequence of interference suppression of absorption is medium bleaching in the process of propagation of CW laser radiation. Let us deduce a law describing laser beam intensity attenuation as a function of the absorption length on the basis of (7). Its differential form may be written as

$$dI = -n\hbar\omega\gamma_1\rho_1(\Omega,I)dx, \tag{15}$$

where n is the number density of absorbing atoms, and dx is the element of absorption length. Using (10) and integrating (15) with $\Omega=\Omega_0$, we obtain for motionless atoms

$$a_0\ln(I/I_0) - a_1(I-I_0) + a_2(I^2-I_0^2) = -k_0x,$$

$$k_0 = 6\pi n\frac{\lambda^2}{N}, \quad a_0 = 1+\frac{1}{3}(N^2-1)\left(\frac{\Delta}{\gamma_1}\right)^2, \tag{16}$$

$$a_1 = 6\pi\left(1-\frac{3}{N}\right)\frac{\lambda^2}{\gamma_1\hbar\omega}, \quad a_2 = S_N\left[\frac{6\pi\lambda^2}{N\Delta\hbar\omega}\right]^2,$$

where I_0 is the intensity at the medium input at $x=0$. Allowance for arbitrary frequency offset from the line center is made in (16) by means of the redefinition

$$a_0 \to a_0 + 4(\Omega-\Omega_0)^2/\gamma_1^2.$$

The first term on the left-hand side of (16) corresponds to ordinary Bouguer attenuation in the absorption regime linear in intensity,[33] and the two other terms arise from nonlinear interference effects. The most interesting situation occurs for $\Delta\to 0$, when $a_2 \propto 1/\Delta^2$, and consequently we may neglect the first two items on the left-hand side of (16), leaving only the key interference term. In this case, intensity attenuation is governed by a square root law:

$$I/I_0 = \sqrt{1-x/L_{\rm coh}},$$
$$L_{\rm coh} = \frac{a_2I_0^2}{k_0} = \frac{3S_N}{2\pi N}\left(\frac{c\lambda n_{\rm phot}}{\Delta}\right)^2\frac{1}{n}, \quad n_{\rm phot} = \frac{I_0}{c\hbar\omega}. \tag{17}$$

Let us compare (17) with the law of intensity decline with length derived for the conditions of coherent population trapping.[23,25] According to Eq. (3.6) of Ref. 23, rewritten in our notation, we have for the line center

$$\frac{I}{I_0} = 1-\frac{x}{L_{\rm coh}}, \quad L_{\rm coh} = \frac{16c}{\gamma_1}\frac{n_{\rm phot}}{n}. \tag{18}$$

Comparison of (17) and (18) shows that medium bleaching under the action of nonlinear interference effects and coherent population trapping proceeds in significantly different ways. First, the power dependences on length are different (square root in the first case and linear in the second). Second, the length $L_{\rm coh}$, for which complete absorption of radiation occurs, is proportional to the original beam intensity for coherent population trapping, whereas for nonlinear interference effects it is proportional to the intensity squared. The ratio

$$L_{\rm coh}^{\rm NIE}/L_{\rm coh}^{\rm CPT} = \frac{3S_N}{64\pi^2N}\frac{\gamma_1\omega}{\Delta^2}\lambda^3n_{\rm phot} \tag{19}$$

is much greater than unity for $\Delta\sim\gamma_1\ll\omega$ and $\lambda^3n_{\rm phot}\gg 64\pi^2N/(3S_N)$, and grows linearly with increasing I_0 and quadratically with decreasing Δ. Thus, under the conditions specified, coherent medium bleaching due to nonlinear interference effects under consideration is much more significant that the analogous bleaching due to coherent radiation trapping. The latter starts dominating at the same intensity levels only for $\Delta > \sqrt{\gamma_1\omega}$. It should be remembered that the domains of manifestation of these two phenomena are separated by the organization of experiment in addition. One field is used in the level crossing scheme for the nonlinear interference effects under consideration, and two fields are used in the scheme of two-photon resonance in the case of coherent population trapping. A coincidence occurs when coherent radiation trapping among magnetic sublevels of the ground level is studied.[13,14]

Let us compare $L_{\rm coh}$ (17) with the linear absorption length $L_0=a_0/k_0$, for which I decreases by the factor e:

$$\frac{L_{\rm coh}}{L_0} = \frac{a_2I_0^2}{a_0} = \left(6\pi\frac{\omega}{N\Delta}\lambda^3n_{\rm phot}\right)^2\left[1+\frac{1}{3}\left(\frac{N\Delta}{\gamma_1}\right)^2\right]^{-1}. \tag{20}$$

Numerical estimates for atoms with $N=3$, $\lambda=0.7$ μm, $\gamma=10^8$ s^{-1}, $\Delta=10^6$ s^{-1}, $I_0=1$ mW/cm^2 give $L_{\rm coh}/L_0\simeq 7$. For $L_0=1$ W/cm^2, this ratio grows by six orders of magnitude. When passing to longer-wave radiation resonant with vibrational transitions of molecules, the expected increase in $L_{\rm coh}/L_0\propto\lambda^4$ does not take place because of the concurrent drastic decrease in the radiative decay constant of vibrational levels. For instance, at $\lambda=10.6$ μm, $I_0=1$ W/cm^2, $\gamma_1=10^3$ s^{-1} and $\Delta=10^6$ s^{-1}, the ratio $L_{\rm coh}/L_0$ grows only fourfold in comparison with the estimate for atoms. Note at the same time that according to these estimates, the effect under consideration is no less important for molecules than for atoms.

4. COLLISION EFFECTS

Let us consider first the case of high pressures of buffer gas, using again the model of equidistant lines of equal intensity for the sake of simplicity, and assuming

$$\Gamma_i=\Gamma_{ij}\equiv\Gamma\sim\nu\gg\gamma_1, \quad \delta_i=\delta_{ij}=0, \tag{21}$$

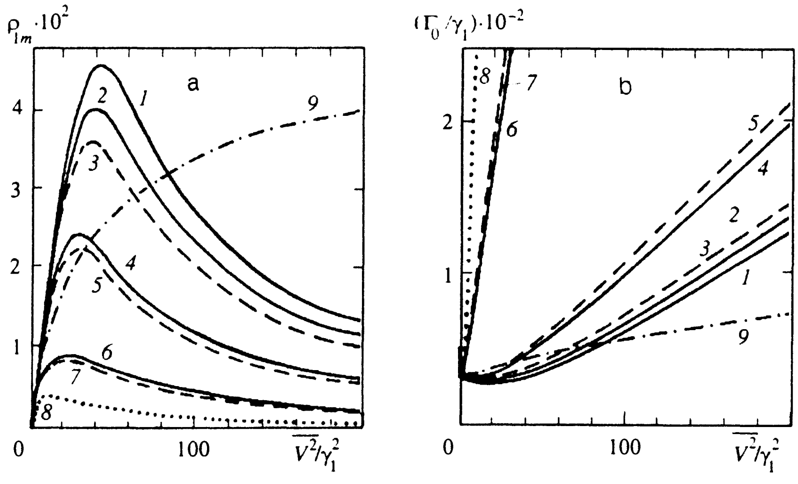

FIG. 1. Absorption at the line center (a) and profile half-width (b) as functions of average radiation intensity. 1) Calculation from Eq. (7); 2,4,6) calculation from (6) at $A_i = \text{const}_i$ for rectangular distributions of realization probabilities of $\Delta_{i+1,i}$ centered at $5\gamma_1$ and beginning at $3\gamma_1$, γ_1, and 0 respectively; 3,5,7) the same distributions for the $\Delta_{i+1,i}$ values with additional averaging over $V_i^2 = \mathscr{V} A_i$ with rectangular distribution of realization probability of V_i^2 from 0 to $2\overline{V^2}$; 8) exponential probability distributions for $\Delta_{i+1,i}$ and V_i^2 with widths respectively $5\gamma_1$ and $\overline{V^2}$ at a level $1/e$; 9) calculation in the absence of nonlinear interference effects with inclusion of population effects.

$$D'_{ij} = 1/\Gamma, \quad D''_{ij} = \Delta(i-j)/\Gamma^2, \quad \Gamma^2 \gg (\tilde{N}\Delta)^2.$$

It follows from (21) that in the limit $\Gamma \to 0$, the polarization-coupling matrix elements tend to zero, $D_{ij} \to 0$, and nonlinear interference effects are completely suppressed. If so, system (3) has the following solution:

$$\rho_1^{(0)} = \beta k_{\text{sat}}, \quad k_{\text{sat}} \varkappa \Gamma^2 / [(\Omega - \Omega_0)^2 + (1+\varkappa)\Gamma^2] < 1,$$

$$\varkappa = 2\tilde{N} V^2 / (\gamma_1 \Gamma), \quad \Omega_0 = -(\tilde{N}-1)\Delta/2, \tag{22}$$

i.e., an ordinary Lorentzian line profile with allowance for saturation described by the parameter $\varkappa$. Comparing (22) with the well-known Karplus–Schwinger formula,[30] we can see that the saturation intensity of the multilevel system considered is $\tilde{N}$ times less than the corresponding intensity for a two-level system.

The inclusion of polarization interference with nonzero D_{ij} (21) to a first approximation in the small parameter $(\tilde{N}\Delta/\Gamma)^2$ gives

$$\rho_1 = \rho_1^{(0)} + \frac{\gamma_1}{\Gamma} \rho_1^{(0)} K_{\text{sat}} \left\{ 1 + \frac{1}{4} \left(\frac{\tilde{N}\Delta}{\Gamma} \right)^2 \times \left[1 - 8k^2 - \frac{4\Gamma}{3\tilde{N}\nu} k(1-2k) \right] \right\}, \tag{23}$$

$$k = \Gamma^2 [(\Omega - \Omega_0)^2 + \Gamma^2]^{-1} \leqslant 1.$$

As can be seen from (23), the magnitude of the relative contribution of nonlinear interference effects at high pressures is determined first of all by the small ratio of the radiative and collisional relaxation constants γ_1/Γ. This corresponds to the foregoing estimate (5) of this contribution made on the basis of $\varkappa_{\text{coh}}$. The expression with alternating signs in curly brackets (23) has an additional small parameter $\sim(\tilde{N}\Delta/\Gamma)^2$, and the contribution of collisional redistribution of populations among splitting sublevels associated with the parameter ν is still smaller by a factor of about $\tilde{N}$. The relative increment of the population ρ_1 in the line center due to interference is $(\gamma_1/\Gamma)\varkappa/(1+\varkappa)$, correct to $O((\tilde{N}\Delta/\Gamma)^2)$, whereas the relative addition to ρ_1 at half maximum, $\rho_1^{(0)}$, is one-half this quantity. Consequently, at high pressures, nonlinear interference effects always result in a small line profile narrowing.

Analytic expressions for the corrections to Eqs. (6) and (7) in the case of low pressures can be obtained in a similar way by expanding $D_{ij}(\Gamma_{ij})$ (4) in powers of $\Gamma_{ij}/|\Delta_{ij}| \ll 1$, but these expressions will be more tedious than (23). The reason for the complication is the differentiated action of nonlinear interference effects in the region of small and medium pressures. These effects may result in profile $\rho_1(\Omega)$ narrowing or splitting with increasing gas pressure, depending on various relations between the Einstein coefficients and the Rabi frequencies of optical

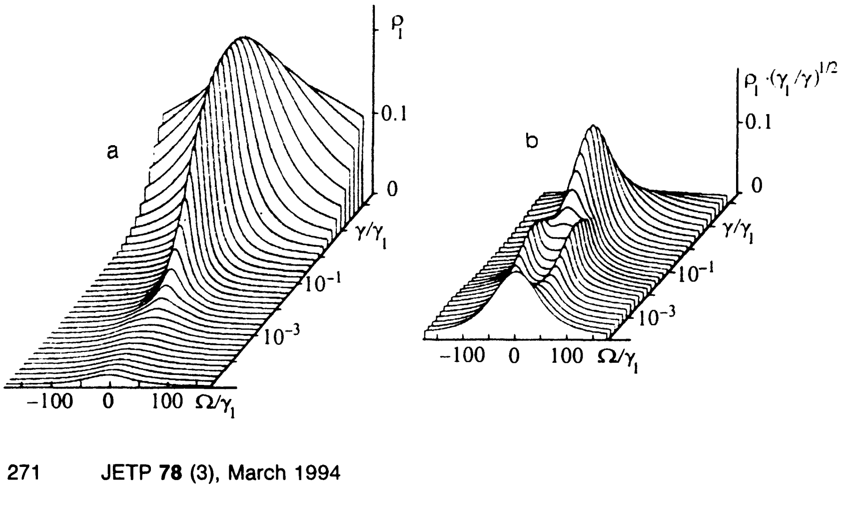

FIG. 2. Line shape as a function of gas pressure: a) $A_1:A_2:A_3 = 1:100:1$; b) $A_1 = A_2 = A_3 = \gamma_1/3$. Here, $A_1 + A_2 + A_3 = \gamma_1$, $V_1^2:V_2^2:V_3^2 = 1:100:1$, $V_1 = \gamma_1$, $\Delta = 2.34\gamma_1$, $\Gamma_i = \Gamma_{ij} = 1.5\gamma$, $i,j = 1,2,3$, $i \neq j$. Frequency Ω offset here and in Fig. 3 is taken relative to the frequency-average optical transition.

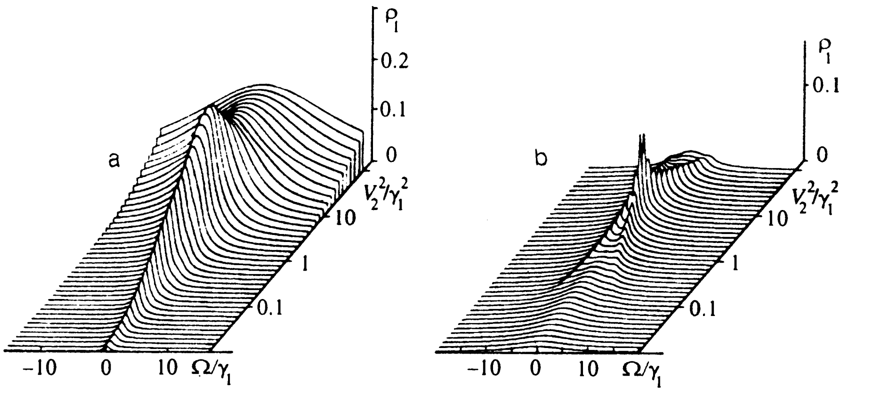

FIG. 3. Line shape as a function of radiative intensity: a) $A_1:A_2:A_3=1:100:1$; b) $A_1=A_2=A_3=\gamma_1/3$. Here, $A_1+A_2+A_3=\gamma_1, V_1^2:V_2^2:V_3^2=1:100:1, \Delta=2.34\gamma_1$, $\Gamma_i=\Gamma_{ij}=1.5\gamma$, $i,j=1,2,3$, $i\neq j$, $\gamma=0.05\gamma_1$.

transitions.[28,29] Therefore, we shall perform the subsequent analysis of the case of arbitrary pressures invoking numerical calculations taking a four-level system with three equidistant sublevels of the ground state as an example. The transitions from the sublevels to the excited state are allowed. The transitions from the sublevels to the excited state are allowed. With this goal, we obtained an exact analytic solution of system (3) with $\tilde{N}=N=3$, $\bar{N}=0$, $\gamma_p=\gamma$, $\nu_p=2\gamma$, $\delta_i=\delta_{ij}=0$. The analytical programming language REDUCE was used. The numerical values of the solution $\rho_1(\Omega)$ are presented in Figs. 2 and 3.

It can be seen from Fig. 2a that in the case $A_1:A_2:A_3=V_1^2:V_2^2:V_3^2$, the growth of pressure and, correspondingly, the collisional constant γ proportional to it, results in the profile $\rho_1(\Omega)$ narrowing in the medium pressure range, $0.01\gamma_1<\gamma<10\gamma_1$, as with a three-level system.[28] The maximum amplitude ρ_{1m} of the profile and its half-width Γ_0 as functions of γ are presented together in Fig. 4 for clarity. The location of the profile center $\Omega_0=0$ in this case, in contrast to Ref. 28, is unchanged because of symmetry of the system considered. For $A_1=A_2=A_3$ in the range $5\cdot10^{-3}\gamma_1<\gamma<2\cdot10^{-2}\gamma_1$, the profile splits into two symmetrically located components due to collisions. Over a narrower range, $2\cdot10^{-2}\gamma_1<\gamma<5\cdot10^{-2}\gamma_1$, the profile splits into three components, which corresponds to the number of allowed transitions in the system. However, in this case, the magnitude of the largest offset of the side components from the center, $\simeq70\gamma_1$, considerably exceeds the difference between the optical transition frequencies, $\approx2\gamma_1$, which must be accounted for by the combined action of collisions and the field.

A more complete picture of the field intensity dependence of the line profile at nonzero pressure is presented in Figs. 3a and 3b for two relations between the radiative constants A_i employed to calculate the curves in Figs. 2a and 2b. For different Einstein coefficients, the line profile is not split or shifted (Fig. 3a) as in the case of the pressure dependence of $\rho_1(\Omega)$ (Fig. 2a). In the moderate intensity range, its amplitude ρ_{1m} reaches a maximum, falls, and increases again to the limit determined by absorption saturation. The power narrowing of the line is not observed by virtue of the choice $N=3$ (see Eqs. (8) and (9) and their discussion), but the rate of power broadening is low up to the point at which ρ_{1m} reaches its maximum $\rho_{1\max}$. The dependences $\rho_{1m}(V^2)$ and $\Gamma_0(V^2)$ presented in Fig. 5, obtained on the basis of data from Fig. 2a, complement Fig. 1 with results for the case of nonzero pressure of the broadening gas. The profile behavior for $A_1=A_2=A_3$ is more interesting (Fig. 2b). As the intensity increases, the profile successively splits into three and two components, and in the range $5\lesssim V_2^2/\gamma_1^2\lesssim8$, the components merge into a single narrow profile with a large amplitude. Further intensity growth creates an inverted splitting pattern when the limit of large V^2 is achieved. In this pattern there is no profile splitting, and the profile amplitude and width grow.

The basic qualitative observation following from the analysis of Figs. 4 and 5 is that the presence of collisions results in nonzero absorption in the limit of large intensi-

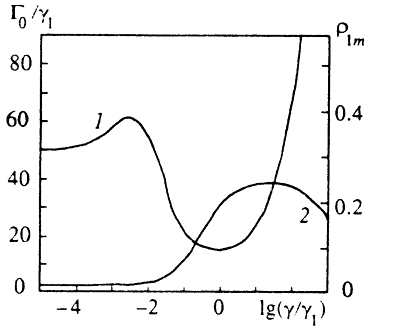

FIG. 4. Half-width Γ_0 (curve *1*) and maximum amplitude ρ_{1m} (*2*) of profiles presented in Fig. 2a as functions of pressure.

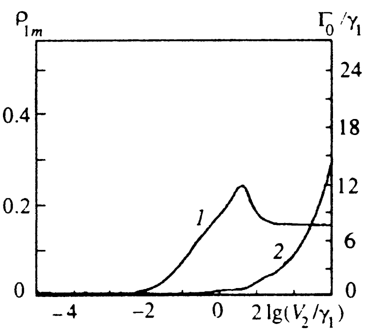

FIG. 5. Maximum amplitude ρ_{1m} (curve *1*) and half-width Γ_0 (*2*) of the profiles presented in Fig. 3a versus field intensity.

ties even for small γ values. Thus, the effect of interference suppression of absorption under study is bounded in principle by collisions. Nevertheless, it can be quite appreciable at moderate intensities.

Another important qualitative feature of nonlinear interference effect manifestation in the presence of collisions is in the line profile splitting over certain ranges of the collisional relaxation constants and the field intensities in the case of equal radiative decay constants.

5. CONCLUSION

The considered phenomenon of interference suppression of absorption and the accompanying effects of power narrowing line splitting are most pronounced for the ground state splittings lying within the radiative line width in the absence of collisions or at low pressures of the buffer gas. The radiative intensities needed for its realization are much lower than the radiative intensities saturating the atomic and molecular transitions. Thus, the conditions for the manifestation of interference suppression of absorption are close to the conditions for cell measurements of low-pressure gas spectra, radiation propagation in the upper atmospheric layers, laser radiation action on atomic beams, etc. This necessitates the analysis of the conditions and the results of such measurements for the presence of interference suppression of absorption, whereas they are often interpreted invoking a simple two-level model of the medium. Without allowance for this phenomenon, highly distorted quantitative data can be obtained.

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