Polarization Phenomena and Nonlinear Interference Effects with Allowance for Collisions

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We consider the spectral singularities in the absorption (gain) of a weak wave in the presence of a strong wave with the same transition and in the presence of collisions that alter the velocity of the atom and the magnetic quantum number. We show that the weak-field absorption line shape depends strongly on the type of transition, on the polarization ratio of the strong and weak waves, and also on the structure of the collision integral. A simple interpretation of the noted effects is presented. The model of strong collisions is used.

1. INTRODUCTION

NONLINEAR interference effects (NIE) constitute an important branch of nonlinear spectroscopy, and are the subject of many papers[1-12]. Most of them consider simple schemes without allowance for level degeneracy. Variation of the field polarizations and the level degeneracy can lead, however, to significant changes in the results. This phenomenon was predicted theoretically by Dienes[6] for the spontaneous approximation. Its experimental observation was reported in[11].

Interesting phenomena occur also in the presence of collisions that alter the state of the atom. Some aspects of these phenomena were noted by Alekseev[10] for the particular case of the $J = 1 \to J = 0$ transition, linear polarizations of the strong and weak fields, and collisions with change of the magnetic quantum number M. Collision effects without allowance for degeneracy are investigated in detail in[12]. The purpose of the present paper is to analyze the case of arbitrary polarizations of strong and weak fields with allowance taken of the Zeeman structure of the levels. We consider a model of strong collisions that cause both mixing over the Zeeman sublevels and diffusion in velocity space.

2. BASIC EQUATIONS

Let the atom be located in an external electromagnetic field $\mathscr{E}$ represented by a strong and a weak traveling wave that are resonant to the same transition $mJ_m \to nJ_n$:

$$\mathscr{E} = \tfrac{1}{2}\{\mathbf{E}\exp[-i(\omega t - \mathbf{kr})] + \mathbf{E}_\mu \exp[-i(\omega_\mu t - \mathbf{k}_\mu \mathbf{r})]\} + \text{c.c.} \quad (2.1)$$

The subscript μ labels the weak field, which does not perturb the levels; $\omega = kc$ and $\omega_\mu = k_\mu c$ are the frequencies of the corresponding waves. In the resonant approximation, the system of equations for the density matrix elements is

$$\begin{gathered}(\partial/\partial t + \mathbf{v}\nabla + \Gamma_m)\hat{\rho}_m = i(\hat{V}\hat{\rho}^+ - \hat{\rho}\hat{V}^+) + \hat{q}_m + \hat{S}_m,\\ (\partial/\partial t + \mathbf{v}\nabla + \Gamma_n)\hat{\rho}_n = i(\hat{V}^+\hat{\rho} - \hat{\rho}^+\hat{V}) + \hat{q}_n + \hat{S}_n,\\ (\partial/\partial t + \mathbf{v}\nabla + \Gamma)\hat{\rho} = i(\hat{V}\hat{\rho}_n - \hat{\rho}_m\hat{V}),\\ V(mM, nM') = \mathscr{E}\mathbf{d}(mM, nM')\exp(i\omega_{mn}t)/\hbar.\end{gathered} \quad (2.2)$$

Here $\hat{V}$ is the matrix of the interaction between the atom and the field; ω_{mn} is the transition frequency; $\hat{\rho}$ consists of those total-density matrix elements which are not diagonal with respect to the levels and take the form $\rho(mM, nM')$; $\hat{\rho}_j (j = m, n)$ consists of the matrix elements $\rho(jM, nM')$ which are diagonal with respect to the levels; Γ_j are the natural widths of the levels $j = m, n$; Γ is the collision-broadened natural line width; $\hat{q}_j$ characterizes the excitation rate of the states $\{j, M, \mathbf{v}\}$:

$$\hat{q}_j = \frac{Q_j W(\mathbf{v})\hat{E}}{2J_j + 1}, \quad W(\mathbf{v}) = \frac{\exp\{-\mathbf{v}^2/\bar{v}^2\}}{\pi^{3/2}\bar{v}^3}, \quad (2.3)$$

where $\hat{E}$ is a unit matrix. We choose the collision integrals $\hat{S}_m$ and $\hat{S}_n$ in the form (the strong-collision model)

$$\begin{aligned}\hat{S}_j = &-\nu_j\hat{\rho}_j + \bar{\nu}_j' W(\mathbf{v})\langle\hat{\rho}_j\rangle_\mathbf{v} + \bar{\nu}_j''\frac{\hat{E}}{2J_j+1}\mathrm{Sp}_M\hat{\rho}_j\\ &+ \bar{\nu}_j''' W(\mathbf{v})\frac{\hat{E}}{2J_j+1}\mathrm{Sp}_M\langle\hat{\rho}_j\rangle_\mathbf{v},\end{aligned}$$

where $\langle\ldots\rangle_\mathbf{v}$ denotes averaging over the velocities. The first term in $\hat{S}_j$ describes the usual relaxation due to the departure from the state $\{j, M, \mathbf{v}\}$; ν_j is the frequency of collisions with quenching and with change of the velocity and of the magnetic quantum number M. The remaining terms describe the arrival at the state $\{j, M, \mathbf{v}\}$ for three types of collision, each of which is characterized by its own frequency. The second term in (2.4) describes strong collisions with change of velocity, but without change of the internal state of the atom (we choose equal arrival frequencies for the diagonal and off-diagonal elements $\rho(jM, jM)$ and $\rho(jM, jM')$); the third term describes strong collisions with reorientation of the atom, but without a change of velocity. Finally, the fourth term describes the last type of collision, with simultaneous change of both the velocity and the magnetic quantum number, and with establishment of an equilibrium distribution with respect to v and M. In the last two types of collision it is assumed that the phase relations are fully violated for the elements of the type $\rho(jM, jM')$ when $M \neq M'$, a fact ensured by the unit matrices $\hat{E}$.

When the strong and weak fields act simultaneously on one transition, polarization is induced in the medium at the strong-field frequency ω, the weak-field frequency ω_μ, and the combination frequency $2\omega - \omega_\mu$. We confine ourselves henceforth to the first-order nonlinear corrections. The polarization at the com-

bination frequency can then be neglected, and we can seek the solution of the system (2.2) in the following convenient form:

$$\begin{gathered}\hat{V}=\hat{V}_0\exp\{-i(\Omega t-\mathbf{kr})\}+\hat{V}_\mu\exp\{-i(\Omega_\mu t-\mathbf{k}_\mu\mathbf{r})\},\\ \hat{\rho}=\hat{\rho}_0\exp\{-i(\Omega t-\mathbf{kr})\}+\hat{\rho}_\mu\exp\{-i(\Omega_\mu t-\mathbf{k}_\mu\mathbf{r})\},\end{gathered}\tag{2.5}$$

$$\begin{gathered}\hat{\rho}_j=\hat{\rho}_j^0+[\hat{\rho}_j^\mu\exp\{-i(\varepsilon t-\mathbf{qr})\}+\text{c.c.}],\\ \Omega=\omega-\omega_{mn},\quad \Omega_\mu=\omega_\mu-\omega_{mn},\quad \varepsilon=\omega_\mu-\omega,\quad \mathbf{q}=\mathbf{k}_\mu-\mathbf{k},\end{gathered}$$

where $\hat{\rho}_0$, $\hat{\rho}_\mu$, $\hat{\rho}_j^0$, and $\hat{\rho}_j^\mu$ do not depend on the coordinates and the time. The matrices $\hat{\rho}_0$ and $\hat{\rho}_j^0$ are due to the interaction with the strong wave only and satisfy the relations

$$\begin{gathered}\Gamma_m\hat{\rho}_m^0=i(\hat{V}_0\hat{\rho}_0^+-\hat{\rho}_0\hat{V}_0^+)+\hat{q}_m+\hat{S}_m^0,\\ \Gamma_n\hat{\rho}_n^0=i(\hat{V}_0^+\hat{\rho}_0-\hat{\rho}_0^+\hat{V}_0)+\hat{q}_n+\hat{S}_n^0,\\ (\Gamma-i\Omega')\hat{\rho}_0=i(\hat{V}_0\hat{\rho}_n^0-\hat{\rho}_m^0\hat{V}_0),\quad \Omega'=\Omega-\mathbf{kv},\end{gathered}\tag{2.6}$$

while the equations for $\hat{\rho}_\mu$ and $\hat{\rho}_j^\mu$ take the form

$$\begin{gathered}(\Gamma_m-i\varepsilon')\hat{\rho}_m^\mu=i(\hat{V}^\mu\hat{\rho}_0^+-\hat{\rho}_\mu\hat{V}_0^+)+\hat{S}_m^\mu,\\ (\Gamma_n-i\varepsilon')\hat{\rho}_n^\mu=i(\hat{V}_0^+\hat{\rho}_\mu-\hat{\rho}_0\hat{V}_\mu^+)+\hat{S}_n^\mu,\\ (\Gamma-i\Omega_\mu')\hat{\rho}_\mu=i(\hat{V}_\mu\hat{\rho}_n^0-\hat{\rho}_m^0\hat{V}_\mu)+i(\hat{V}_0\hat{\rho}_n^\mu-\hat{\rho}_m^\mu\hat{V}_0);\\ \Omega_\mu'=\Omega_\mu-\mathbf{k}_\mu\mathbf{v},\quad \varepsilon'=\varepsilon-\mathbf{qv}.\end{gathered}\tag{2.7}$$

The collision integrals $\hat{S}_j^0$ and $\hat{S}_j^\mu$ are obtained from (2.4) by making the substitutions $\rho_j\to\rho_j^0$ and $\rho_j\to\rho_j^\mu$, respectively.

We analyze the solution of Eqs. (2.6) and (2.7) in the weak-saturation approximation, using the usual condition $\Gamma\ll k\bar{v}$ for gas lasers. In addition, we confine ourselves to waves that propagate either in parallel ($\mathbf{k}_\mu\uparrow\uparrow\mathbf{k}$) or antiparallel ($\mathbf{k}_\mu\uparrow\downarrow\mathbf{k}$) directions.

3. OPERATION AT THE WEAK-FIELD FREQUENCY

We consider first the solution of Eqs. (2.6) and (2.7) for waves traveling in the same direction. The matrix $\hat{\rho}_\mu$ is given by

$$\begin{aligned}\hat{\rho}_\mu=\frac{-iNW(\mathbf{v})}{\Gamma-i\Omega_\mu'}\Big\{&\hat{V}_\mu-\frac{1}{\Gamma+i\Omega'}\Big[\hat{V}_\mu\hat{V}_0^+\hat{V}_0\Big(\tau_{1n}+\frac{\tau_{1m}}{1-i\varepsilon\tau_{1m}}\Big)\\ &+\hat{V}_0\hat{V}_0^+\hat{V}_\mu\Big(\tau_{1m}+\frac{\tau_{1n}}{1-i\varepsilon\tau_{1n}}\Big)\Big]-\frac{2\sqrt{\pi}}{k\bar{v}}\exp\Big\{-\Big(\frac{\Omega}{k\bar{v}}\Big)^2\Big\}\\ &\times\Big[\hat{V}_\mu\hat{V}_0^+\hat{V}_0\Big(\tau_{2n}'+\frac{\tau_{2m}'}{1-i\varepsilon\tau_m'}\frac{1}{1-i\varepsilon\tau_{1m}}\Big)\\ &+\hat{V}_0\hat{V}_0^+\hat{V}_\mu\Big(\tau_{2m}'+\frac{\tau_{2n}'}{1-i\varepsilon\tau_n'}\frac{1}{1-i\varepsilon\tau_{1n}}\Big)\Big]\\ &-\frac{1}{\Gamma+i\Omega'}\sum_j\Big[\frac{\tau_{2j}''}{2J_j+1}\Big(\hat{V}_\mu\,\mathrm{Sp}_M\,\hat{V}_0^+\hat{V}_0\\ &+\frac{1}{1-i\varepsilon\tau_j''}\frac{1}{1-i\varepsilon\tau_{1j}}\hat{V}_0\,\mathrm{Sp}_M\,\hat{V}_0^+\hat{V}_\mu\Big)\Big]-\frac{2\sqrt{\pi}}{k\bar{v}}\exp\Big\{-\Big(\frac{\Omega}{k\bar{v}}\Big)^2\Big\}\\ &\times\sum_j\frac{1}{2J_j+1}\Big[\tau_{2j}'''\hat{V}_\mu\,\mathrm{Sp}_M\,\hat{V}_0^+\hat{V}_0+\frac{1}{1-i\varepsilon\tau_{1j}}\Big(\frac{\tau_j-\tau_{1j}}{1-i\varepsilon\tau_j}-\\ &-\frac{\tau_{2j}'}{1-i\varepsilon\tau_j'}-\frac{\tau_{2j}''}{1-i\varepsilon\tau_j''}\Big)\hat{V}_0\,\mathrm{Sp}_M\,\hat{V}_0^+\hat{V}_\mu\Big]\Big\},\end{aligned}\tag{3.1}$$

where

$$\begin{gathered}N=\frac{Q_m}{\Gamma_m+\nu_m-\bar{\nu}_m}-\frac{Q_n}{\Gamma_n+\nu_n-\bar{\nu}_n},\quad \bar{\nu}_j=\bar{\nu}_j'+\bar{\nu}_j''+\bar{\nu}_j''',\quad j=m,n;\\ \tau_{1j}=\frac{1}{\Gamma_j+\nu_j},\quad \tau_j'=\frac{1}{\Gamma_j+\nu_j-\bar{\nu}_j'},\quad \tau_j''=\frac{1}{\Gamma_j+\nu_j-\bar{\nu}_j''},\end{gathered}\tag{3.2}$$

$$\begin{gathered}\tau_j=\frac{1}{\Gamma_j+\nu_j-\bar{\nu}_j},\quad \tau_{2j}'=\frac{\bar{\nu}_j'}{\Gamma_j+\nu_j-\bar{\nu}_j'}\frac{1}{\Gamma_j+\nu_j}=\tau_j'-\tau_{1j},\\ \tau_{2j}''=\frac{\bar{\nu}_j''}{\Gamma_j+\nu_j-\bar{\nu}_j''}\frac{1}{\Gamma_j+\nu_j},\quad \tau_{2j}'''=\tau_j-\tau_{1j}-\tau_{2j}'-\tau_{2j}''.\end{gathered}$$

Terms that yield a contribution $\sim\Gamma/k\bar{v}$ after averaging over the velocities are not taken into account in (3.1). We begin the analysis of (3.1) with an explanation of the physical meaning of the times (3.2). Each of them is connected with a definite state of the atom. The time τ_{1j} is the average lifetime of the state $\{j, M, v\}$, i.e., the lifetime prior to collision of any type; τ_j' is the lifetime of the state $\{j, M\}$ (arbitrary velocity); τ_j'' is the lifetime of the state $\{j, v\}$ (arbitrary orientation of the atom); τ_j is the lifetime of the state $\{j\}$ (both the velocity and the orientation are arbitrary); τ_{2j}' is the lifetime of the state $\{j, M, v_p\}$, i.e., a state in which j and M are fixed and the velocity v is represented with a Maxwellian weight; τ_{2j}'' is the lifetime of the state $\{j, M_p, v\}$, i.e., a state in which j and v are fixed, and all the values of M are equally probable; τ_{2j}''' is the lifetime of the state $\{j, M_p, v_p\}$, in which the atom is at the level j and has an equilibrium distribution with respect to M and v.

The expression for $\hat{\rho}_\mu$ contains, as usual, a linear term and an increment due to the presence of the strong field. This increment breaks up into several terms that differ in their nature, and its structure reflects fully the specific features of the collision integral. In accordance with our model it is convenient to speak of four channels of interaction between the atom and the field (the terms of (3.1) corresponding to individual channels are grouped in square brackets). The interaction takes place in the state $\{j, M, v\}$ in the first channel, in the state $\{j, M, v_p\}$ in the second, $\{j, M_p, v\}$ in the third and $\{j, M_p, v_p\}$ in the fourth. It is quite natural that the terms representing each of the channels are proportional to the lifetimes in the states corresponding to these channels (in the last channel this holds true when $\epsilon = 0$). The total lifetime τ_j of the atom at the level j breaks up effectively into four non-overlapping intervals, in each of which the interaction with the field is via one of the four channels.

We note also the following detail. An increase of the frequencies $\tilde{\nu}_j''$ or $\tilde{\nu}_j'''$ leads to a decrease of the time τ_{2j}', and an increase of $\tilde{\nu}_j'$ or $\tilde{\nu}'''$ leads to a decrease of τ_{2j}''. To the contrary, an increase of $\tilde{\nu}_j'$ or $\tilde{\nu}_j''$ leads to an increase of the time τ. This is natural, since collisions in which M is changed quench the state $\{j, M, v_p\}$, and collisions with change of v quench the state $\{j, M_p, v\}$. On the other hand, there is no elastic collision capable of taking the atom out of the state $\{j, M_p, v_p\}$, i.e., the time alotted to the fourth channel can perhaps only increase with increasing frequency of such collisions, because two successive collisions (one with a change of M only, and the other with a change of v only) also lead to the state $\{j, M_p, v_p\}$.

We consider now the absorption of a weak field. We direct the quantization axis along the strong-field wave vector, and resolve the electric vectors of the fields into circular components:

$$E_{\pm1}=(E_x\pm iE_y)/\sqrt{2},\quad E_{\pm1}^\mu=(E_x^\mu\pm iE_y^\mu)/\sqrt{2}.\tag{3.3}$$

The interaction matrix elements are then given by

$$V^0(mM, nM') = \frac{1}{2\hbar}\sum_{\alpha=\pm 1} E_\alpha d^\alpha(mM, nM'),$$
$$V^\mu(mM, nM') = \frac{1}{2\hbar}\sum_{\alpha=\pm 1} E_\alpha^\mu d^\alpha(mM, nM'), \quad (3.4)$$

where $d^\alpha(mM, nM')$ are the circular components of the dipole moment[13]. We write out the expression for the work of the weak field:

$$\mathscr{P}_\mu \infty \exp\left[-\left(\frac{\Omega_\mu}{k\bar v}\right)^2\right] \mathrm{Re}\left\{\sum_\alpha |G_\alpha^\mu|^2 - \right.$$
$$- \frac{1}{2\Gamma - i\varepsilon}\left[C_m\left(\tau_{1n} + \frac{\tau_{1m}}{1 - i\varepsilon\tau_{1m}}\right) + C_n\left(\tau_{1m} + \frac{\tau_{1n}}{1 - i\varepsilon\tau_{1n}}\right)\right]$$
$$- \frac{\sqrt{\pi}}{k\bar v}\exp\left[-\left(\frac{\Omega}{k\bar v}\right)^2\right]\left[C_m\left(\tau_{2n}' + \frac{\tau_{2m}'}{1 - i\varepsilon\tau_m'}\frac{1}{1 - i\varepsilon\tau_{1m}}\right)\right.$$
$$\left. + C_n\left(\tau_{2m}' + \frac{\tau_{2n}'}{1 - i\varepsilon\tau_n'}\frac{1}{1 - i\varepsilon\tau_{1n}}\right)\right] -$$
$$- \frac{^2/_3}{2\Gamma - i\varepsilon}\sum_j \frac{\tau_{2j}''}{2J_j + 1}\left[C_1 + C_2\frac{1}{1 - i\varepsilon\tau_j''}\frac{1}{1 - i\varepsilon\tau_{1j}}\right] - \quad (3.5)$$
$$- \frac{2}{3}\frac{\sqrt{\pi}}{k\bar v}\exp\left[-\left(\frac{\Omega}{k\bar v}\right)^2\right]\sum_j \frac{1}{2J_j + 1}\left[C_1\tau_{2j}''' + \right.$$
$$\left.\left. + C_2\left(\frac{\tau_j - \tau_{1j}}{1 - i\varepsilon\tau_j} - \frac{\tau_{2j}'}{1 - i\varepsilon\tau_j'} - \frac{\tau_{2j}''}{1 - i\varepsilon\tau_j''}\right)\frac{1}{1 - i\varepsilon\tau_{1j}}\right]\right\};$$

$$C_m = A_0\sum_\alpha (|G_\alpha G_\alpha^\mu|^2 + A_1|G_\alpha G_{-\alpha}^\mu|^2 + A_2 G_\alpha G_{-\alpha}{}^* G_{-\alpha}^\mu G_\alpha^{\mu *}),$$
$$G_\alpha = E_\alpha d_{mn}/2\hbar;$$
$$C_n = A_0\sum_\alpha (|G_\alpha G_\alpha^\mu|^2 + A_2|G_\alpha G_{-\alpha}^\mu|^2 + A_1 G_\alpha G_{-\alpha}{}^* G_{-\alpha}^\mu G_\alpha^{\mu *}),$$
$$G_\alpha^\mu = E_\alpha^\mu d_{mn}/2\hbar;$$
$$C_1 = \sum_\alpha (|G_\alpha G_\alpha^\mu|^2 + |G_{-\alpha}|^2|G_\alpha^\mu|^2),\quad C_2 = \sum_\alpha (|G_\alpha G_\alpha^\mu|^2 + G_\alpha G_{-\alpha}{}^* G_{-\alpha}^\mu G_\alpha^{\mu *})$$

Here d_{mn} is the reduced matrix element of the dipole moment. The coefficients A_0, A_1, and A_2 depend on the type of transition; their values are:

	$J \to J$	$J-1 \to J$	$J \to J-1$
A_0:	$\frac{2}{5}\frac{2J^2+2J+1}{J(J+1)(2J+1)}$	$\frac{2}{5}\frac{6J^2-1}{J(2J-1)(2J+1)}$	$\frac{2}{5}\frac{6J^2-1}{J(2J-1)(2J+1)}$
A_1:	$\frac{(2J-1)(2J+3)}{2(2J^2+2J+1)}$	$\frac{(J-1)(2J-3)}{2(6J^2-1)}$	$\frac{(J+1)(2J+3)}{2(6J^2-1)}$
A_2:	$\frac{(2J-1)(2J+3)}{2(2J^2+2J+1)}$	$\frac{(J+1)(2J+3)}{2(6J^2-1)}$	$\frac{(J-1)(2J-3)}{2(6J^2-1)}$

C_1 and C_2 differ from C_m and C_n, obviously, because of the depolarizing collisions.

We point out the general regularities characterizing all the channels of the interaction with the field. These include the presence of two different terms, a "population" term due to the change of the level populations under the influence of the strong field, and an interference term due to the interference of the atomic states. The "population" term gives either a relatively broad contour of width 2Γ), or a Doppler contour resulting from homogeneous saturation. The interference terms take the form of narrower "dips" against the background of the Doppler contour. Each channel introduces its own interference "dip". It is typical that the widths of these "dips" do not depend on the times to which their amplitudes are proportional. Thus, for the second, third, and fourth channels of the interaction, the widths of the narrowest "dips" are determined by the lifetimes of the states $\{j, M\}$, $\{j, v\}$, and $\{j\}$, respectively, whereas the amplitudes of these "dips" are proportional to the shorter times τ_{2j}', τ_{2j}'', and τ'''. Only in the first channel does a single time τ_{1j} characterize both the amplitude and the width of the "dip"; consequently, each relaxation process gives a spectral structure with a width determined by the characteristic time for the given process. The "dip" produced in the fourth channel may turn out to be much narrower than the others, since its width includes only the natural level width and the frequency of the quenching collisions, which is usually much lower than the frequency of the elastic collisions.

Thus, the presence of diffusion in velocity space and of collisions with change of M greatly enriches the spectral composition and leads to more complicated interference effects. The appearance of collision effects is connected with the ratio of the amplitudes of the corresponding terms. Thus, collisions in which only M changes (third channel) can manifest themselves if

$$\tau_{2j}''/(2J_j + 1) \sim 1/(\Gamma_j + \nu_j). \quad (3.6)$$

At not too large J_j this means that the frequency of such collisions should be of the order of the width of the level j. The second and fourth channels make a noticeable contribution under a stronger condition, namely if, respectively,

$$\tau_{2j}'/k\bar v \sim 1/\Gamma(\Gamma_j + \nu_j),\quad \tau_{2j}'''/k\bar v(2J_j + 1) \sim 1/\Gamma(\Gamma_j + \nu_j). \quad (3.7)$$

Owing to the large Doppler width, the conditions (3.7) can be satisfied only when the collision frequencies greatly exceed the level widths. In the case of dragging of the resonant radiation, such a situation is perfectly feasible (cf., e.g.,[14]). As to elastic collisions, relations (3.7) are feasible under realistic constants ($p \sim 1$ Torr) either for metastable states of atoms or for molecular systems.

4. MODEL OF THREE RELAXATION CONSTANTS

This model presupposes the absence of diffusion in velocity state and mixing over the sublevels, and the role of the collisions reduces to quenching and to phase randomization. It is therefore necessary to neglect in the collision integral (2.4) the arrival over all the interaction channels. Then

$$\mathscr{P}_\mu \infty \exp\left\{-\left(\frac{\Omega_\mu}{k\bar v}\right)^2\right\}\left[\sum_\alpha |G_\alpha^\mu|^2 - \frac{C_m}{2\Gamma - \Gamma_m - \nu_m}\frac{\Gamma_m + \nu_m}{(\Gamma_m + \nu_m)^2 + \varepsilon^2}\right.$$
$$- \frac{C_n}{2\Gamma - \Gamma_n - \nu_n}\frac{\Gamma_n + \nu_n}{(\Gamma_n + \nu_n)^2 + \varepsilon^2} - (2\Gamma - \Gamma_m - \nu_m - \Gamma_n - \nu_n)\frac{2\Gamma}{(2\Gamma)^2 + \varepsilon^2}$$
$$\left. \cdot\left(\frac{C_m}{(\Gamma_n + \nu_n)(2\Gamma - \Gamma_m - \nu_m)} + \frac{C_n}{(\Gamma_m + \nu_m)(2\Gamma - \Gamma_n - \nu_n)}\right)\right]. \quad (4.1)$$

We see from (4.1) that the weak-field absorption line contour contains, against the Doppler background, three "dips" with center at the frequency $\epsilon = \Omega_\mu - \Omega = 0$ and with different widths (2Γ, $\Gamma_m + \nu_m$, $\Gamma_n + \nu_n$) and amplitudes. It is easily seen that upon satisfaction of the condition

$$2\Gamma - \Gamma_m - \nu_m - \Gamma_n - \nu_n = 0, \quad (4.2)$$

which takes place, in particular, for spontaneous approach ($2\Gamma = \Gamma_m + \Gamma_n$, $\nu_m = \nu_n = 0$), the "dip" with width 2Γ drops out. If the Weisskopf broadening mechanism becomes appreciable, then $2\Gamma > \Gamma_m + \nu_m + \Gamma_n + \nu_n$, and the "dip" with width 2Γ being the broadest, can be separated against the background of the remaining "dips" $\Gamma_m + \nu_m$ and $\Gamma_n + \nu_n$. This peculiarity is also characteristic of the simple two-level model (without allowance for degeneracy).

One of the results of the model of two nondegenerate states is the satisfaction of the condition $C_m = C_n$, which leads in practice to equality of the amplitudes of the "dips" $\Gamma_m + \nu_m$ and $\Gamma_n + \nu_n$. When degeneracy is taken into account, the ratio C_m/C_n depends strongly on the state of the polarizations of the fields. If, for example, both fields are equally polarized, then

$$C_m = C_n = A_0 \sum_{\alpha} [|G_\alpha|^2 + (A_1 + A_2)|G_{-\alpha}|^2]|G_\alpha{}^\mu|^2. \qquad (4.3)$$

To the contrary, the maximum difference between C_m and C_n occurs for orthogonal circular polarizations of the fields (see the table). The ratio C_m/C_n can differ greatly here from unity. This makes amplitude discrimination of the "dips" $\Gamma_m + \nu_m$ and $\Gamma_n + \nu_n$ possible, a rather important fact from the experimental point of view. We note also that an increase of the total angular momentum J leads to equalization of C_m and C_n for all polarization states, so that in the limiting case $J \gg 1$ we obtain in this sense the equivalent of the problem of two nondegenerate states. We point out, however, that at $J \lesssim 10$ (molecular systems) the ratio C_m/C_n is still noticeable.

It is convenient to interpret the polarization effects from the point of view of the concept of the splitting of the energy levels by a strong field[5]. We assume for concreteness that the waves have orthogonal circular polarizations and act on the transition m, $J_m = 1 \rightarrow n$, $J_n = 2$. If the quantization axis is chosen to be parallel to the wave vectors, the transition scheme takes the form shown in Fig. 1a. The straight lines correspond to the strong field and the wavy lines to the weak one. The perturbing action of the strong field is manifest by splitting, in this case, of the Zeeman sublevels with formation of energy quasilevels (Fig. 1b)[5]. These quasilevels correspond to the characteristic roots

$$\alpha_{1m} = -\gamma_n - i[E_m/\hbar + \Omega - \mathbf{kv}), \quad \alpha_{2m} = -\gamma_m - iE_m/\hbar,$$
$$\alpha_{1n} = -\gamma_n - iE_n/\hbar, \quad \alpha_{2n} = -\gamma_m - i[E_n/\hbar - \Omega + \mathbf{kv}]. \qquad (4.4)$$

in the approximation employed, one of the components

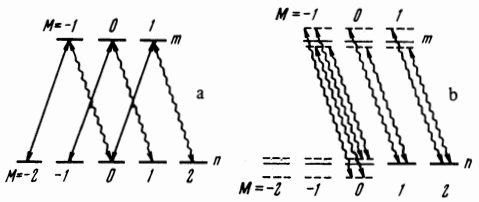


FIG. 1. a) Scheme of transitions between the Zeeman sublevels m, $J_m = 1$ and n, $J_n = 2$. The strong and weak fields are waves with orthogonal circular polarizations. b) The same scheme when the field is replaced by the result of its action—the splitting of the Zeeman sublevels. In both cases the quantization axis is directed along the strong-field wave vector.

is assumed to be unshifted (its shift is less than hG in energy units). In terms of the density matrix, $\gamma_m = (\Gamma_m + \nu_m)/2$ and $\gamma_n = (\Gamma_n + \nu_n)/2$. The spectral characteristics of the absorption (emission) of the weak field are thus determined completely with the aid of the level-splitting scheme (Fig. 1b).

When the frequency of the weak field is scanned, the following situation arises. There are two resonances at the transitions $M = 0 \rightarrow 1$ and $1 \rightarrow 2$:

$$[(\gamma_m + \gamma_n)^2 + (\Omega_\mu - \mathbf{k}_\mu \mathbf{v})^2]^{-1}, \quad [(2\gamma_n)^2 + (\varepsilon - \mathbf{qv})^2]^{-1}. \qquad (4.5)$$

The first spectral line is inhomogeneously broadened by the Doppler effect and gives a Doppler contour. The second line, in the case of waves having the same direction ($\mathbf{q} = 0$), does not experience broadening upon averaging over the velocities, and enters the final result in the form of an individual spectral structure of width equal to the level relaxation constant n. We see that these transitions do not give the "dip" $\Gamma_m + \nu_m$. On the other hand, for the transition $M = -1 \rightarrow 0$, scanning of Ω_μ yields, generally speaking, four resonances:

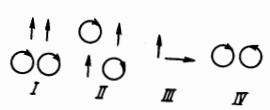
$$[(\gamma_m + \gamma_n)^2 + (\Omega_\mu')^2]^{-2}, \quad [(2\gamma_n)^2 + (\varepsilon')^2]^{-1}, \quad [(2\gamma_m)^2 + (\varepsilon')^2]^{-1},$$
$$[(\gamma_m + \gamma_n)^2 + (\Omega' - \varepsilon')^2]^{-1}, \quad \Omega' = \Omega - \mathbf{kv}, \quad \Omega_\mu' = \Omega_\mu - \mathbf{k}_\mu \mathbf{v}, \quad \varepsilon' = \varepsilon - \mathbf{qv}. \qquad (4.6)$$

From the definite symmetry of the scheme we can conclude that the result for the transition between the given sublevels of the line with widths $2\gamma_m$ and $2\gamma_n$ will enter with equal weights. The last resonance occurs at the combination frequency ($2\Omega - \Omega_\mu - 2\mathbf{k}\cdot\mathbf{v} + \mathbf{k}_\mu \cdot \mathbf{v} = 0$). This line drops out in the approximation under consideration.

Thus, in the transition $M = -1 \rightarrow 0$, both "dips," $\Gamma_n + \nu_n$ and $\Gamma_m + \nu_m$, appear in the weak-field absorp-

Values of C_m/C_n for particular cases of the polarization states of the strong and weak wave

Polarization	C_m/C_n	$J-1 \rightarrow J$	$0 \rightarrow 1$	$1/2 \rightarrow 3/2$	$1 \rightarrow 2$	$3/2 \rightarrow 5/2$	$2 \rightarrow 3$	$10 \rightarrow 11$
I	1	1	1	1	1	1	1	1
II	$\frac{1+A_1}{1+A_2}$	$\frac{14J^2-5J+1}{14J^2+5J+1}$	$\frac{1}{2}$	$\frac{5}{8}$	$\frac{47}{67}$	$\frac{76}{101}$	$\frac{56}{71}$	$\frac{1607}{1717}$
III	$\frac{1+A_1-A_2}{1-A_1+A_2}$	$\frac{(J-1)(6J+1)}{(J+1)(6J-1)}$	0	$\frac{1}{4}$	$\frac{13}{33}$	$\frac{24}{49}$	$\frac{19}{34}$	$\frac{115}{126}$
IV	$\frac{A_1}{A_2}$	$\frac{(J-1)(2J-3)}{(J+1)(2J+3)}$	0	0	$\frac{1}{21}$	$\frac{3}{28}$	$\frac{1}{6}$	$\frac{19}{30}$



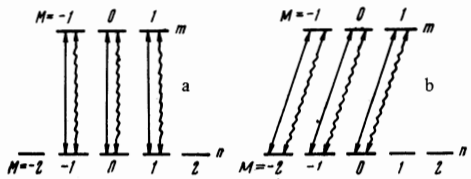


FIG. 2. Scheme of transitions between the sublevels of the levels m, $J_m = 1$ and n, $J_n = 2$ for waves with like polarizations: a) linear polarizations (the quantization axis is along the electric vectors) b) circular polarization (the quantization axis is along the wave vectors).

tion line, and their amplitudes are equal. Since each transition makes an additive contribution to the work of the weak field, it is seen in accordance with the foregoing that in the analyzed scheme the amplitude of the "dip" $\Gamma_n + \nu_n$ should exceed that of $\Gamma_m + \nu_m$. The concrete amplitude ratio depends on the ratio of the oscillator strengths in the transitions between the Zeeman sublevels, i.e., on J, ΔJ, M, and ΔM. In the scheme of Fig. 1 we have $C_m/C_n = 1/21$, i.e., the "dip" $\Gamma_n + \nu_n$ prevails so much that the contribution of the "dip" $\Gamma_m + \nu_m$ to the line contour can be neglected.

In the general case of arbitrary total angular momenta J, in accordance with our interpretation, the prevailing amplitude is that of the "dip" having the width of the level whose J is larger. For transitions of the $J \to J$ type, consequently, we have $C_m = C_n$ regardless of the field polarization. Another natural fact within the framework of the present analysis is that in the transitions $J = 0 \leftrightarrow J = 1$ and $J = \frac{1}{2} \leftrightarrow J = \frac{3}{2}$ there is in general no "dip" with the width of the level having $J = 0$ or $\frac{1}{2}$. Indeed, in such a situation, only one Zeeman level is perturbed in any one transition on which the weak field acts. If the field polarizations are identical (say, the same circular polarization or polarization in one plane), then, as shown in Fig. 2, both fields act on the same transitions between the Zeeman sublevels. Reasoning as before, we arrive at the natural result that $C_m = C_n$.

It is clear from the foregoing that there exists an analogy with the nondegenerate two-level and three-level systems. Although we are considering an essentially "many-level" system, it can be broken up into an aggregate of two- and three-level subsystems. We turn again to Fig. 1a. The subsystems connected with the Zeeman sublevels M = 0 and 1 of the level m have a clearly pronounced three-level character (the presence of a weak field in the transition $M = -1 \leftrightarrow 0$ is immaterial in this case, since it does not perturb the atomic states). To the contrary, the subsystem connected with the sublevel M = −1 of the level m, is in fact equivalent to a two-level system. Indeed, the strong field splits the lower Zeeman levels in the transitions $M = -1 \to -2$ and $1 \to 0$ in the same fashion. The wavy arrow from the transition $M = -1 \to 0$ can therefore formally be transferred to the transition $M = -2$. As a result, the entire system breaks up into one two-level and two three-level sybsystems which are not connected with one another.

We can see from (2.7) that when two fields act on one transition, the nonlinear interference effects reduce to beats of the populations at the difference frequency, as a result of which "dips" with widths equal to the level widths appear in the weak-field spectrum. On the other hand in three-level systems, the nonlinear interference effects are beats of the polarization induced in the forbidden transition, with frequency $\Omega_\mu - \Omega$ equal to the difference between the deviations from the frequency of their transition for the weak and strong fields. The result is a single "dip" with the width of the forbidden-transition line (for waves having the same direction and for $|\mathbf{k}_\mu - \mathbf{k}| < |\mathbf{k}|$). For our three-level subsystems we have $\Omega_\mu - \Omega = \omega_\mu - \omega = \epsilon$, and the width of the forbidden transition is $\Gamma_n + \nu_n$. Thus, the three-level subsystems give only the "dip" $\Gamma_n + \nu_n$, while the two-level ones yield both "dips," and we arrive at the earlier conclusion. In the case of like polarizations we obtain only two-level subsystems, so that the result here is obvious.

As is well known, nonlinear interference effects in opposing strong- and weak-field waves are suppressed by Doppler broadening. The work of the weak field, as a function of Ω_μ, has only one "dip" at the frequency $\Omega_\mu = -\Omega$, with a width 2Γ:

$$\mathcal{P}_\mu \infty \exp\left\{-\left(\frac{\Omega_\mu}{k\bar{v}}\right)^2\right\}\left[\sum_\alpha |G_\alpha^\mu|^2 - \left(\frac{C_m}{\Gamma_n+\nu_n}+\frac{C_n}{\Gamma_m+\nu_m}\right)\frac{2\Gamma}{(2\Gamma)^2+(\Omega_\mu+\Omega)^2}\right]. \tag{4.7}$$

It is most remarkable that the times $1/(\Gamma_m + \nu_m)$ and $1/(\Gamma_n + \nu_n)$ combine in (4.7) with unequal weights, which vary with the field polarization. This can be used successfully to obtain information on the level relaxation constants and on the character of their variation with pressure. Indeed, comparative experiments performed for identical and different (say, opposing circular) field polarizations to determine the "dip" amplitude make it possible to determine the ratio $(\Gamma_m + \nu_m)/(\Gamma_n + \nu_n)$. Previously employed procedures to determine the level widths reduced to investigations of the spectral line shapes. This always entails greater difficulties and larger errors than amplitude measurements. The observed fact therefore deserves close attention from the experimental point of view.

5. POLARIZATION PHENOMENA AND COLLISIONS

Let us examine the influence of a change in the polarization states of the strong and weak fields on the weak-field absorption line shape, including in our consideration collisions that change M and v. In the model of the nondegenerate states[12] the amplitudes of the "population" and interference terms are equal. In our case, as seen from (3.5), this equality does not hold in the general case ($C_1 \neq C_2$, $C_m \neq C_n$). The amplitude ratio varies when the field polarizations are changed. It turns out that each channel (each collision model) reacts differently to the change of polarization. If both fields are equally polarized, then $C_1 = C_2$, $C_m = C_n$, and the amplitudes of the interference terms, just as in the model of nondegenerate states, are equal to the "population" amplitudes. In the case of opposing circular or perpendicular linear polarizations we have

$$\mathcal{P}_\mu \infty \exp\left\{-\left(\frac{\Omega_\mu}{k\bar{v}}\right)^2\right\} |G^\mu|^2 \operatorname{Re}\left\{1 - \frac{|G|^2}{2\Gamma - i\varepsilon}\left[C_m'\left(\tau_{1n} + \frac{\tau_{1m}}{1 - i\varepsilon\tau_{1m}}\right)\right.\right.$$

$$+C_n'\left(\tau_{1m}+\frac{\tau_{1n}}{1-i\varepsilon\tau_{1n}}\right)\Big]-|G|^2\frac{\sqrt{\pi}}{k\bar{v}}\exp\left\{-\left(\frac{\Omega}{k\bar{v}}\right)^2\right\}\cdot$$
$$\times\Big[C_m'\left(\tau_{2n}'+\frac{\tau_{2m}'}{1-i\varepsilon\tau_{m}'}\frac{1}{1-i\varepsilon\tau_{1m}}\right)+C_n'\left(\tau_{2m}'+\frac{\tau_{2n}'}{1-i\varepsilon\tau_{n}'}\frac{1}{1-i\varepsilon\tau_{1n}}\right)\Big]$$
$$-|G|^2\sum_j\frac{2}{3}\frac{1}{2J_j+1}\Big[\frac{\tau_{2j}''}{2\Gamma-i\varepsilon}+\frac{\sqrt{\pi}}{k\bar{v}}\exp\left\{-\left(\frac{\Omega}{k\bar{v}}\right)^2\right\}\tau_{2j}'''\Big]\Big\}, \tag{5.1}$$

where

1) $C_m'=A_0(1+A_1-A_2)$, $C_n'=A_0(1-A_1+A_2)$;
2) $C_m'=A_0A_1$, $C_n'=A_0A_2$.

The values 1) and 2) have been written out for linear-perpendicular and opposing-circular polarizations, respectively. Formula (5.1) describes the following interesting fact: the coefficient C_2 turns out to equal zero, so that the interference terms drop out in the last two channels. The interference term remains in the second channel, as before. To interpret this effect, it is useful to write out the system (2.7) in expanded form. For concreteness, we take the case of linear perpendicular polarizations (the quantization axis is chosen along the strong-field electric vector, q = 0):

$$(\Gamma_m-i\varepsilon)\rho^\mu(mM,mM')=-i\Big[\rho^\mu(mM,nM')G^*c^0(M'M')$$
$$-\rho^0(mM',nM')\sum_\alpha G_\alpha^\mu c^\alpha(MM')\Big]+S^\mu(mM,mM'),$$
$$(\Gamma_n-i\varepsilon)\rho^\mu(nM,nM')=i\Big[\rho^\mu(mM,nM')G^*c^0(MM)$$
$$-\rho^0(mM,nM)\sum_\alpha G_\alpha^\mu c^\alpha(MM')\Big]+S^\mu(nM,nM'), \tag{5.2}$$
$$(\Gamma-i\Omega_\mu')\rho^\mu(mM,nM')=iG[\rho^\mu(nM,nM')c^0(MM)-$$
$$-\rho^\mu(mM,mM')c^0(M'M')]+i\sum_\alpha G_\alpha^\mu c^\alpha(MM')\cdot$$
$$\times[\rho^0(nM',nM')-\rho^0(mM,mM')];$$
$$c^\alpha(MM')=d^\alpha(mM,nM')/d_{mn},\quad \alpha=-1,0,1;\quad G_0^\mu=0.$$

The interference terms are obviously given by the first term of the last equation of (5.2). The elements $\rho^\mu(jM, jM')$ are excited only when the elements $\rho^0(jM, jM')$ are present. Since $G_0^\mu = 0$, only the elements that are not diagonal in M, $\rho^\mu(jM, jM')$, are excited, while the elements of the type $\rho^\mu(jM, jM)$ remain equal to zero. The structure of the collision integral (2.4) is therefore of importance. Namely, the role of collisions with change of M reduces to an increase of the relaxation constant of the elements $\rho^\mu(jM, jM')$ by the frequency of these collisions, and there is no arrival term. The interference term therefore contains no "dips" corresponding to the third and fourth channels.

The result is the consequence of the collision model chosen by us, wherein the collisions with change of M lead to quenching of the density-matrix elements that are not diagonal in M. Unlike the diagonal part of the integral (2.4), collisions with change of velocity only conserve in our model the phase relations in the jM → jM′ transition, and consequently the corresponding interference "dip" is retained. If we choose opposing circular polarizations, then we can likewise show that only the nondiagonal elements $\rho^\mu(jM, jM')$ are excited, and the conclusions remain the same. For fields with like polarization, it can be seen, with Fig. 2 as an example, that the problem breaks up effectively into an aggregate of two-level problems, i.e., the elements $\rho^\mu(jM, jM')$ which are not diagonal in M are not excited, and the collision integral (2.4) is fully effective.

In addition to the discussed effects, variation of the field polarizations changes the amplitude of the total "dip" on the Doppler contour, which is proportional to

$$(C_m+C_n)\frac{\tau_{1n}+\tau_{1m}}{2\Gamma}+\frac{\sqrt{\pi}}{k\bar{v}}\exp\Big[-\left(\frac{\Omega}{k\bar{v}}\right)^2\Big](C_m\tau_{2m}'+C_n\tau_{2n}')$$
$$+(C_1+C_2)\frac{1}{2\Gamma}\frac{2}{3}\sum_j\frac{\tau_{2j}''}{2J_j+1}+\frac{\pi^{1/2}}{k\bar{v}}\exp\Big[-\left(\frac{\Omega}{k\bar{v}}\right)^2\Big]\frac{2}{3}C_2\sum_j\frac{\tau_{2j}'''}{2J_j+1}. \tag{5.3}$$

It is easily noted that in the model of three relaxation constant the ratio of the "dip" amplitudes measured at different polarization states is a specified number that depends neither on the atomic constants Γ_j and Γ nor on the pressure. Elastic collisions of any type (the concrete model obviously does not matter here) change this ratio, and from its deviation from the calculated model one can assess the validity of the model of three relaxation constants.

No less important information can be extracted from measurements of the amplitude of the total nonlinear increment to (3.5), which is proportional to

$$(C_m+C_n)\Big[\frac{\tau_{1n}+\tau_{1m}}{2\Gamma}+\frac{\sqrt{\pi}}{k\bar{v}}\exp\left\{-\left(\frac{\Omega}{k\bar{v}}\right)^2\right\}(\tau_{2m}'+\tau_{2n}')\Big]$$
$$+\frac{2}{3}(C_1+C_2)\sum_j\frac{1}{2J_j+1}\Big[\frac{\tau_{2j}''}{2\Gamma}+\frac{\sqrt{\pi}}{k\bar{v}}\exp\left\{-\left(\frac{\Omega}{k\bar{v}}\right)^2\right\}\tau_{2j}'''\Big]. \tag{5.4}$$

If there are no depolarizing collisions (i.e., collisions with change of M), then the amplitude of the nonlinear increment should vary in accordance with a strictly specified law with changing field polarization. Violation of this law should consequently be evidence of the presence of depolarizing collisions. From this point of view, the model to which these collisions corresponds is of no importance whatever.

When opposing waves are considered, we have the following expression for the work of the weak field:

$$\mathcal{P}_\mu\infty\exp\left\{-\left(\frac{\Omega_\mu}{k\bar{v}}\right)^2\right\}\Big[\sum_\alpha|G_\alpha^\mu|^2$$
$$-\frac{2\Gamma}{(2\Gamma)^2+(\Omega_\mu+\Omega)^2}\Big(C_m\tau_{1n}+C_n\tau_{1n}+\frac{2}{3}\sum_j\frac{\tau_{2j}''}{2J_j+1}C_1\Big) \tag{5.5}$$
$$-\frac{\sqrt{\pi}}{k\bar{v}}\exp\left\{-\left(\frac{\Omega}{k\bar{v}}\right)^2\right\}\Big(C_m\tau_{2n}'+C_n\tau_{2m}'+\frac{2}{3}C_1\sum_j\frac{\tau_{2j}'''}{2J_j+1}\Big)\Big].$$

The interference terms drop out in all channels. The remaining "population" terms produce a "dip" of width 2Γ on the Doppler contour, and in the second and fourth channels there is only a broad Doppler "pedastal." The depolarizing collisions without change of the velocity influence the "dip" amplitude and can therefore introduce a certain error when $(\Gamma_m + \nu_m)/(\Gamma_n + \nu_n)$ is determined by the method described above.

6. CONCLUSION

Let us summarize our results briefly. When the level degeneracy is taken into account, variation of the field polarizations results in an appreciable change in the weak-field absorption line shape. By using this

fact, we can obtain more accurate information on the atomic constants and on the character of the collisions. Besides the analysis of the line shape, information can be gained also from amplitude measurements, which can also offer a check on the validity of certain models. The fact that polarization effects are very sensitive to the structure of the collision integral makes it possible to determine experimentally the role of collisions that disturb the phase relations in the transitions jm $\rightarrow$ jM′. Finally, the proposed analysis indicates the applicability limits of models in which the degeneracy of the levels is not taken into account. The results obtained with such models are applicable to the following particular cases: for like field polarization: and in the model of three relaxation constants for the transitions J $\rightarrow$ J or for sufficiently large J (the polarizations can be arbitrary in both cases). In the study of elastic collisions, the polarization effects come into play regardless of the type of transition and of the value of J, so that in this case it may be essential to take the level degeneracy into account.

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