Collisional averaging of the Stark and Zeeman spectrum structure

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The effect of depolarizing collisions on the shape of the spectrum in the quadratic Stark effect and the anomalous Zeeman effect is considered. It is shown that collisions may result in the merging (collapse) of allowed spectrum components. It is established that exchange between Stark and Zeeman components can be observed by analyzing the structure of the Lamb dip. An additional central dip is found. Methods are indicated for the experimental determination of the collisional relaxation constants and the Stark and Zeeman structure parameters.

INTRODUCTION

The structure of atomic multiplets, weakly split by electric or magnetic fields, undergoes a highly nontrivial transformation with increasing gas pressure. The particular feature of this process is that the individual spectrum components not only broaden but also exchange their positions on the frequency scale[1,2]. This phenomenon is a consequence of the fact that the collisions are nonadiabatic for atomic states which are energetically close to one another. Each of these states may become transformed as a result of collisions into a superposition of a number of other states, and the corresponding spectral line may become transformed into other lines.

At low pressures this spectral exchange appears in the spectrum as a matched broadening and shift of mutually coupled lines. However, when the exchange frequency is comparable with the splitting, one observes the collapse of the structure,[3] i.e., the merging of the lines and the reappearance of the spectrum corresponding to free atoms. The collapse is always accompanied by a partial contraction of the spectrum, which in extreme cases is found to continue as the pressure increases[1-3].

Extreme manifestations of spectral exchange (contraction due to motion, exchange contraction, and so on) are frequently encountered in magnetic spectroscopy,[4] but in optics there has been a tradition, deriving from fundamental work in statistical thermodynamics,[5] that on average collisions can only destroy the interference of states and never give rise to it[6]. Although in the general formalism of collision theory this prejudice has been overcome,[7-10] the spectroscopic consequences of the "phase memory" of the atom have not been completely recognized. The hydrogen atom whose spectrum is known to exhibit frequency exchange,[11,12] which leads to the collapse of the Stark structures of the Lyman lines,[3] is hardly the only system demonstrating the reality of the effect. Moreover, it is clear from general considerations that frequency exchange in both the linear Stark effect and in magnetic resonance[4] is only a special manifestation of a general spectroscopic phenomenon.

To prove this conclusion we shall consider the quadratic Stark effect and the anomalous Zeeman effect, which can be observed for practically any excited atom. It is, however, necessary to choose a transition in which the combining terms are both degenerate since otherwise one would require additional degeneracy (as in hydrogen) or a random term proximity (as in helium) in order to ensure that exchange becomes possible.[3] The presence of an external field (which is implied in the case of magnetic resonance) is a necessary condition for the appearance of spectral exchange in optics as well.

In absorption spectra which are linear in the incident light, exchange between the Stark and Zeeman components appears as a broadening and collapse of structures quite similar to those described earlier[1-3]. However, the detection of this phenomenon against the background of a broad Doppler profile requires the use of a high-intensity coherent beam of radiation capable of giving rise to the Lamb dip. It turns out that this produces a modification of the known phenomenon. In the presence of the saturation effect there is a weak additional line at the center of gravity of the spectrum, until the collapse of the structure brings together all the components to a single frequency. The production of the resolved structure is interesting because this gives more complete information on the relaxation invariants, including those which cannot be observed after the collapse and in the absence of the fields.

1. FORMULATION OF THE PROBLEM

The basic possibility of spectral exchange as a result of collisions can be seen in the equations of the theory of collisions[7-10] which describe the evolution of the dipole moment $\hat{d}$ or the density matrix $\hat{\rho}$:

$$\begin{aligned} \dot{d}_{if} &= i[\hat{H}, \hat{d}]_{if} - (P^{if}_{i'f'})^{+} d_{i'f'}, \\ \dot{\rho}_{if} &= -i[\hat{H}, \hat{\rho}]_{if} - P^{i'f'}_{if} \rho_{i'f'}, \end{aligned} \tag{1.1}$$

where $\hbar = 1$. Exchange takes place if the relaxation tensor is the matrix

$$P_{\alpha\beta} = P^{i'f'}_{if} = \langle \delta_{ii'}\delta_{ff'} - S_{ii'}S^{*}_{ff'} \rangle, \quad \alpha = \{i, f\}, \quad \beta = \{i', f'\}, \tag{1.2}$$

which is nondiagonal in the indices α, β of the spectral components. In atomic spectroscopy this is possible if the terms linked by the optical transition are degenerate (Fig. 1).

Collisions whose usual duration is $\tau_c \sim 10^{-13}$ sec cannot induce transitions separated by a quantum of optical frequency $\omega_\alpha = H_{ff} - H_{ii} \gg 1/\tau_c$. However, the same collisions may be nonadiabatic with respect to degenerate or weakly split components of each of these terms. Any

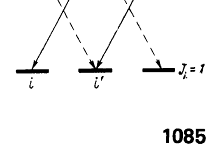

FIG. 1. The structure of transitions between degenerate states with total angular momenta $J_f = J_i = 1$. The solid and broken arrows correspond to radiation components with opposite circular polarizations.

such collision transforms a particular state of the degenerate term into a superposition of certain other states, and this is reflected in the fact that the S matrix is nondiagonal in the indices of the degenerate states, and $P_{\alpha\beta}$ is nondiagonal in the corresponding spectral lines.

As a result, the structure of the absorption spectrum, which is linear in the frequency ω of the incident radiation, [1-3,10]

$$K(\omega)=\operatorname{Re}\int_0^\infty d_{if}(\tau)\,d_{fi}(0)\,e^{-i\omega\tau}d\tau=\operatorname{Re}d_\alpha^*\frac{1}{i(\omega\delta_{\alpha\beta}-\omega_{\alpha\beta})-P_{\alpha\beta}},\quad (1.3)$$

can be elucidated only after inverting the matrix in the denominator of the last expression. In the energy representation $\omega_{\alpha\beta} = \omega_\alpha \delta_{\alpha\beta}$, but the fact that $P_{\alpha\beta}$ is nondiagonal requires the solution of the secular problem, the rank of which increases as the number of spectral components which undergo collisional exchange increases. It is precisely this feature of the nonadiabatic theory which is exhibited by the matched broadening and shift of lines, the final result of which is the collapse of the structure.

However, the nonadiabatic character of collisions is only the necessary condition for spectral exchange. As a rule, averaging over the impact parameter and velocity, shown by the angle brackets in Eq. (1.2), ensures that most of the nondiagonal elements of $P_{\alpha\beta}$ which are responsible for the transformation of the α component of the spectrum into the β component become equal to zero. Moreover, when one of the combining terms is nondegenerate and isolated, i.e. is unperturbed by collisions ($S_{ii'} \sim \delta_{ii'}$), this averaging generally diagonalizes the entire matrix $P_{\alpha\beta}$ in the representation characteristic for the total angular momentum J and its component J_z:

$$P_{if}^{if'}=P_{if}^{if}\,\delta_{ff'}.$$

This result is a general consequence of the isotropy of space at the instant of collision, [7] which is not violated even by the external field which is definitely weak in comparison with the interatomic interaction. It is only when both terms are degenerate that averaging over the directions of motion does not remove all the nondiagonal elements of $P_{\alpha\beta}$. Having carried this out in the general form we can show that the tensor $\hat{P}$ assumes the following structure:

$$P_{rs}^{r's'}=\sum_{\varkappa q}\gamma_\varkappa^{rs}(2\varkappa+1)\begin{pmatrix}J_r & \varkappa & J_s\\ -r & q & s\end{pmatrix}\begin{pmatrix}J_r & \varkappa & J_s\\ -r' & q & s'\end{pmatrix}(-1)^{2J_r-r-r'},\quad (1.4)$$

where J_r and J_s are the total angular momenta of states with components r and s; r, s = i, f and γ_κ^{rs} are the relaxation invariants [13,14]

$$\gamma_\varkappa^{rs}=8\pi^2 n\int\rho\,d\rho\int u^3 f(u)\,du\sum_J\begin{Bmatrix}J_r & \varkappa & J_r\\ J_s & J & J_s\end{Bmatrix}F(J)\qquad (1.4a)$$

The actual magnitude of these is determined by the result of the collision:

$$F(J)=\sum_{rr'ss'p}(2J+1)\begin{pmatrix}J_r & J_s & J\\ r' & s & p\end{pmatrix}\begin{pmatrix}J_r & J_s & J\\ r & s' & p\end{pmatrix}(\delta_{rr'}\delta_{ss'}-\tilde{S}_{rr'}\tilde{S}_{ss'}^*),\quad(1.4b)$$

i.e. by the $\tilde{S}$ matrix in the "collision system" where the x axis is collinear with the relative velocity **u**, and the y axis is collinear with the impact parameter $\boldsymbol{\rho}$ (n is the density of the collision partners and f(u) is the Maxwellian distribution).

Some of the γ_κ^{rs} have been computed [7,13-16] for particular values of the total angular momenta J_i and J_f and a number of interaction potentials, but in this paper we shall look upon them as phenomenological parameters. It is readily seen that the nondegeneracy of the i-th level, which is frequently encountered in absorption from the ground state (J_i = i = 0), leads to the diagonalization of $P_{if}^{i'f'}$. However, this is not the case in general: Eq. (1.4) enables us to conclude only that $i - f = i' - f'$.

We may therefore conclude that transitions between the sublevels of degenerate terms undergo exchange as a result of collisions. Nevertheless, no qualitative anomalies associated with this are to be expected in the spectra of free atoms. According to a theorem formulated by Anderson[17] in the first paper on the nonadiabatic theory of collisions, one Lorentz line corresponds to each pair of terms in the free-atom spectrum, independently of their degree of degeneracy. In the light of Eq. (1.4) this means that all the possible optical transitions at this frequency have the same collision width, specified by the quantity γ_κ^{if} (for electric dipole transitions $\kappa = 1$).

The situation changes radically only when an external field is introduced. When this field is represented by the atomic Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}$ the external field is given by

$$\hat{V}=-\hat{\mathbf{d}}\mathscr{E}-\hat{\boldsymbol{\mu}}\mathscr{H},\qquad (1.5)$$

where $\mathscr{E}$ and $\mathscr{H}$ are the electric and magnetic fields, and $\hat{\mathbf{d}}$ and $\hat{\boldsymbol{\mu}}$ are the corresponding dipole moments. In the κ representation which is characteristic for P, the operators $\hat{V}$ and $\hat{H}$ are nondiagonal and, therefore, the matrix $\omega_{\alpha\beta}$ is also nondiagonal in the indices of the spectral components. If, on the other hand, we consider the representation characteristic for $\hat{V}$ and diagonalize $\omega_{\alpha\beta}$, then $P_{\alpha\beta}$ becomes nondiagonal. In any case, the inversion of the denominator in Eq. (1.3) reduces to the solution of the same secular problem which accounts for the existence of spectral exchange.

If the relaxation tensor is diagonal in the indices of the Zeeman sublevels, then only transitions to states with different J can be exchange coupled. Since, as a rule, such transitions are very different in frequency, we have a secular simplification of the problem [4,18], i.e., we can neglect exchange effects. This is the situation for absorption from a nondegenerate state in practically all cases with the exception of the hydrogen atom in which owing to the additional L degeneracy these frequencies are randomly close [3,11,12]. If, on the other hand, both the combining terms are degenerate, and we neglect exchange between different multiplets, all that remains is the exchange between different M components of the same transition, which is present even in Eq. (1.4). The presence of a field which splits these components is necessary only to exhibit the effect.

It is important to remember that collapse occurs when the splitting of the spectrum is comparable with the width of its components. Since the latter has a collisional origin, the structure as a whole may be hidden under the broader Doppler profile. It can be isolated against this background only in laser spectroscopy in the form of the corresponding Lamb dips. It is precisely in this way that the normal Zeeman effect was recorded for the $2s_2 - 2p_1$ transition in neon[10]. Although spectral exchange was absent from this system, because the initial term was nondegenerate, there is no doubt that the anomalous Zeeman effect and the quadratic Stark effect, the structure of which should collapse with increasing pressure, can be observed in precisely the same way.

Nonlinear detection of these effects complicates their description. Equation (1.3) is then inconvenient, since in this form the correlation theory is in fact the theory of linear response. Instead, it is necessary to introduce the resonance interaction operator for circularly polarized light ($q = \pm 1$) explicitly into the atomic Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V} + \hat{F}(t)$:

$$\hat{F}=-\mathrm{Re}[\hat{d}^q E_q(z) e^{i\omega t}], \quad d_{if}{}^q=(-1)^{J_i-i}\begin{pmatrix} J_i & 1 & J_f \\ -i & q & f \end{pmatrix} d, \tag{1.6}$$

and calculate the work done by this field

$$\mathscr{P}=2N\omega\,\mathrm{Im}\int \rho_{if}(v,z,t)F_{fi}(z,t)\,dv \tag{1.7}$$

in terms of the density matrix $\hat{\rho}$, the equation for which must also be suitably generalized, taking into account the motion of the particles with velocity v along the z axis (direction of observation):

$$\dot{\hat{\rho}}+v\frac{\partial\hat{\rho}}{\partial z}=-i[\hat{H}-i\hat{\Gamma},\hat{\rho}]-\hat{P}\hat{\rho}+\hat{Q}\varphi(v). \tag{1.8}$$

In these expressions $\hat{\rho}$ determines the state of the atoms moving with velocity v, and the one-dimensional Maxwellian distribution $\varphi(v)$ specifies their fraction among the total number N of the "active" particles. The change in velocity on collision is neglected. To ensure stationary adsorption of light, we have introduced into Eq. (1.8) the pump matrix $\hat{Q}$ and the spontaneous decay matrix $\Gamma_{\alpha\beta}$ which must be taken into account for excited atoms.

To ensure continuity with the result of correlation theory,[1-3] we shall consider in the new formulation of the problem a purely linear effect in the field of the progressive wave, and neglect the Doppler shift. We shall then take into account the nonlinear modification which leads to the formation of the Lamb dip with resolved structure. These effects will be demonstrated for transitions between levels with total angular momenta $J_i = J_f = 1$.

2. LINEAR EFFECT

Let us therefore suppose that the field splitting of the components and their collisional width are greater than the Doppler width. In this situation, even the usual absorption spectra exhibit spectral exchange which is not complicated either by saturation or by the Doppler effect. Neglecting the latter $[\varphi(v) = \delta(v)]$, and assuming that $E_q(z) = \frac{1}{2}E_0\exp(-ikz)$, it is convenient to consider only one pair of lines, for example, that with right-handed polarization ($q = +1$), since the other is broadened and split in precisely the same way and quite independently. This follows from the fact that the relaxation matrix splits into unconnected blocks for these pairs, each of which is of the form

$$P_{\alpha\beta}=\frac{1}{2}\left\|\begin{matrix}\gamma_1+\gamma_2 & \gamma_1-\gamma_2\\ \gamma_1-\gamma_2 & \gamma_1+\gamma_2\end{matrix}\right\|, \quad \gamma_\varkappa=\gamma_\varkappa{}^{if}, \quad \varkappa=1,2. \tag{2.1}$$

The diagonal elements of this matrix specify the individual widths of the components, and the nondiagonal elements specify the frequency with which they exchange[1)].

The final form of the spectrum can be determined only with allowance for both factors and for the form of the matrix

$$\omega_{\alpha\beta}=\left\|\begin{matrix}\omega_\alpha & 0\\ 0 & \omega_\beta\end{matrix}\right\|, \quad \omega_\alpha=H_{00}-H_{-1,-1}, \quad \omega_\beta=H_{11}-H_{00}, \tag{2.2}$$

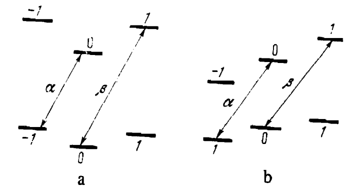

FIG. 2. The effect of circularly polarized radiation on split sublevels of the transition $J_f = J_i = 1$: a – quadratic Stark effect, b – anomalous Zeeman effect.

which specifies the splitting of the components in the electric or magnetic field[19] (Fig. 2):

$$\begin{aligned}&\text{a) Stark effect}: \ \omega_\alpha=\omega_0+\bar{\omega}+{}^1\!/_2 b_i\mathscr{E}^2, \quad \omega_\beta=\omega_0+\bar{\omega}-{}^1\!/_2 b_f\mathscr{E}^2,\\ &\text{b) Zeeman effect}: \ \omega_\alpha=\omega_0+\mu_0 g_i\mathscr{H}, \quad \omega_\beta=\omega_0+\mu_0 g_f\mathscr{H}.\end{aligned} \tag{2.3}$$

In these expressions ω_0 is the transition frequency in the absence of the fields, $\bar{\omega} = \frac{1}{2}(a_i - a_f)\mathscr{E}^2$ is the shift in the external electric field $\mathscr{E}$, b_i and b_f are constants characterizing the individual shift of sublevels in this field, and g_i and g_f are the Landé g factors which play the same role in the magnetic field $\mathscr{H}$ (μ_0 is the Bohr magneton). For the sake of simplicity, the constant fields are assumed to be parallel to the z axis.

The above data are sufficient to enable us to use Eq. (1.3) to obtain an idea of the Stark and Zeeman spectrum structure. However, Eqs. (1.7) and (1.8) also require the definition of the quantities

$$\Gamma_{\alpha\beta}=\frac{1}{2}\left\|\begin{matrix}\Gamma_i+\Gamma_f & 0\\ 0 & \Gamma_i+\Gamma_f\end{matrix}\right\|, \quad Q_{jj}^{j'j'}=Q_j\delta_{jj'}\,(j=i,f). \tag{2.4}$$

The form of $\hat{Q}$ corresponds to the assumption of isotropic pumping and collisional relaxation of populations which must be taken into account in calculations of the nonlinear effect. It is described by the matrices

$$P_{jj}^{j'j'}=\left\|\begin{matrix}{}^1\!/_2\gamma_{1j}+{}^1\!/_6\gamma_{2j} & -{}^1\!/_3\gamma_{2j} & -{}^1\!/_2\gamma_{1j}+{}^1\!/_6\gamma_{2j}\\ -{}^1\!/_3\gamma_{2j} & {}^2\!/_3\gamma_{2j} & -{}^1\!/_3\gamma_{2j}\\ -{}^1\!/_2\gamma_{1j}+{}^1\!/_6\gamma_{2j} & -{}^1\!/_3\gamma_{2j} & -{}^1\!/_2\gamma_{1j}+{}^1\!/_6\gamma_{2j}\end{matrix}\right\|; \tag{2.5}$$

$$\gamma_{\varkappa j}=\gamma_\varkappa{}^{jj}; \ \varkappa=1,2.$$

Direct calculations show that

$$\mathscr{P}\sim|G|^2\left(\frac{Q_i}{\Gamma_i}-\frac{Q_f}{\Gamma_f}\right)\mathrm{Re}\left[\frac{1+i(\gamma_2-\gamma_1)/\Delta}{\Gamma-i(\Omega-\Delta/2)}+\frac{1-i(\gamma_2-\gamma_1)/\Delta}{\Gamma-i(\Omega+\Delta/2)}\right], \tag{2.6}$$

$$G=\frac{E_0 d}{2}, \quad \Gamma=\frac{\Gamma_i+\Gamma_f}{2}+\frac{\gamma_1+\gamma_2}{2}, \quad \Omega=\omega-\frac{\omega_\alpha+\omega_\beta}{2}, \tag{2.7}$$

$$\Delta=[(\omega_\alpha-\omega_\beta)^2-(\gamma_1-\gamma_2)^2]^{1/2}. \tag{2.8}$$

This result is formally similar to that obtained earlier[1-3]. The coupling between the two lines is due to the "phase memory" which is preserved because of the difference between γ_1 and γ_2. The spectrum structure depends on its actual size in comparison with the splitting $\omega_\alpha - \omega_\beta$.

The quantities γ_1 and γ_2 are of the same order of magnitude as the collision frequency among the particles producing the broadening. When the density n of these particles is low, the splitting of the doublet is large in comparison with the broadening and vice versa. Assuming that the $\gamma_\varkappa$ are real,[14] we have

$$\Delta=\omega_\alpha-\omega_\beta-2\delta, \quad \delta\approx(\gamma_2-\gamma_1)^2/4(\omega_\alpha-\omega_\beta), \tag{2.9a}$$

$$\Delta=i(\gamma_2-\gamma_1+2\gamma), \quad \gamma\approx(\omega_\alpha-\omega_\beta)^2/4(\gamma_2-\gamma_1) \tag{2.9b}$$

at low and high densities respectively.

Using this expansion in Eq. (2.6), we have for low partial pressure

$$\mathscr{P}\sim|G|^2\left[\frac{\Gamma}{\Gamma^2+(\Omega_\alpha+\delta)^2}\left(1+\frac{\gamma_2-\gamma_1}{\Gamma}\frac{\Omega_\alpha+\delta}{\omega_\alpha-\omega_\beta}\right)\right.$$

$$+\frac{\Gamma}{\Gamma^2+(\Omega_\beta-\delta)^2}\left(1-\frac{\gamma_2-\gamma_1}{\Gamma}\frac{\Omega_\beta-\delta}{\omega_\alpha-\omega_\beta}\right)\Bigg], \qquad (2.10a)$$

where $\Omega_\alpha = \omega - \omega_\alpha$, $\Omega_\beta = \omega - \omega_\beta$, and at high partial pressure

$$\mathscr{P}\sim|G|^2\left[\frac{\Gamma_1+\gamma}{(\Gamma_1+\gamma)^2+\Omega^2}\left(2+\frac{\gamma}{\gamma_2-\gamma_1}\right)-\frac{\Gamma_2-\gamma}{(\Gamma_2-\gamma)^2+\Omega^2}\frac{\gamma}{\gamma_2-\gamma_1}\right], \qquad (2.10b)$$

$$\Gamma_{1,2}={}^1/_2(\Gamma_i+\Gamma_f)+\gamma_{1,2}.$$

According to Eq. (2.10a), both components of the resolved structure distort asymmetrically in proportion to $(\gamma_2 - \gamma_1)/(\omega_\alpha - \omega_\beta)$ and shift toward one another by the amount

$$\begin{aligned}\delta&=(\gamma_2-\gamma_1)^2/2(b_i+b_f)\mathscr{E}^2 \quad \text{Stark effect,}\\ \delta&=(\gamma_2-\gamma_1)^2/4\mu_0(g_i-g_f)\mathscr{H} \quad \text{Zeeman effect,}\end{aligned} \qquad (2.11)$$

in inverse proportion to the field splitting. This shift is readily distinguished from the usual shift by the further result that it is quadratic in the partial pressure. As the pressure increases the components progressively approach one another until the collapse of the structure occurs for $|\omega_\alpha - \omega_\beta| = |\gamma_1 - \gamma_2|$. Although the spectrum formally consists of two lines (adsorption and emission) with different width and intensity even after this happens, it exhibits only one adsorption maximum which lies at its center of gravity, and the emission-line component vanishes with increasing pressure.

In the final anslysis, an increase in the collision frequency transforms the spectrum so that it takes the form of a single Lorentz line of twice the intensity, which is broadened in precisely the same way as in the absence of the field:

$$\begin{aligned}&\mathscr{P}\sim 2|G|^2\frac{\Gamma_1}{\Gamma_1^2+\Omega^2}, \qquad \Gamma_1=\frac{\Gamma_i+\Gamma_f}{2}+\gamma_1,\\ &\Omega=\omega-\omega_0-\frac{1}{2}\begin{cases}[a_i-a_f+{}^1/_2(b_i-b_f)]\mathscr{E}^2\\ \mu_0(g_i+g_f)\mathscr{H}.\end{cases}\end{aligned} \qquad (2.12)$$

The width of this line can be expressed in a universal fashion in terms of γ_1, there is no field splitting, and there is only a shift which depends on the field and shows that the field is not zero. Therefore, the rapid frequency exchange removes differences between the components of equal polarization, and combines them at the center of gravity of the multiplet.

It is clear from a comparison between Eqs. (2.12) and (2.6) that observations of the Stark and Zeeman effects give more complete information about collisions which produce line broadening than the atomic spectrum in the absence of the fields. The latter is characterized by the single parameter γ_1 whereas the shifts of the components of the field-resolved structure provides information on both γ_1 and γ_2. The only exception is the normal Zeeman effect ($g_i = g_f$) in which the exchanged components are not split.

When $\gamma_1 < \gamma_2$ the resolved structure lines are somewhat narrower than the spectrum in the absence of the field. In the opposite case, on the other hand, the single line given by Eq. (2.12) is narrower. This means that, when $\gamma_1 < \gamma_2$, the spectrum contracts as a result of collapse (from Γ, the width of the resolved components in a large field, to $\Gamma + \gamma_1$ when the field is reduced). When $\gamma_1 = 0$, the reduction in the collisional width may continue without limit with increasing pressure, since in this case $\gamma = (\omega_\alpha - \omega_\beta)^2/4\gamma^2 \to 0$. This extreme manifestation of collapse ("collisional contraction") is possible if the broadening and the phase memory [diagonal and nondiagonal elements of the matrix given by Eq. (2.1)] are equal in absolute magnitude. It is clear that this occurs when $\gamma_1 = 0$; however, it is obvious that this condition cannot be satisfied in the system under consideration and, therefore, the contraction has an unrestricted character (up to γ_1).

3. NONLINEAR EFFECT

Let us now consider the absorption of the standing wave $E_q(z) = 2E_0 \sin kz$ in the presence of a substantial Doppler broadening ($k\bar{v} \gg \Gamma$, where v is the mean thermal velocity). The work done by the field when the saturation of resonance transitions is included, but terms of the order of $\Gamma/k\bar{v}$ are neglected at low pressures ($|\gamma_1 - \gamma_2| < |\omega_\alpha - \omega_\beta|$), is of the form

$$\begin{aligned}\mathscr{P}\sim|G|^2\Bigg[&\exp\left\{-\frac{\Omega_+^2}{(k\bar{v})^2}\right\}+\exp\left\{-\frac{\Omega_-^2}{(k\bar{v})^2}\right\}\\ &-\frac{|G|^2}{6}\left(I_0+I\frac{\Gamma}{\Gamma^2+\Omega^2}+I_+\frac{\Gamma}{\Gamma^2+\Omega_+^2}+I_-\frac{\Gamma}{\Gamma^2+\Omega_-^2}\right)\Bigg],\end{aligned} \qquad (3.1)$$

where $\Omega_\pm = \Omega \pm \Delta/2$.

Terms which are linear in absorption reproduce the Doppler shape of the doublet components, whilst in nonlinear absorption against the constant background produced by the first component

$$I_0=\frac{2R_1}{\Gamma}\left[1+\frac{2(\gamma_2-\gamma_1)^2}{4\Gamma^2+\Delta^2}\right]-\frac{R_2}{\Gamma}\frac{(\omega_\alpha-\omega_\beta)^2}{4\Gamma^2+\Delta^2}, \qquad (3.1a)$$

we can distinguish three Lorentz lines of equal width, but different intensity and asymmetry:

$$I=R_2\left(\frac{\omega_\alpha-\omega_\beta}{\Delta}\right)^2+2R_3\frac{\Omega}{\Gamma}\frac{(\gamma_1-\gamma_2)(\omega_\alpha-\omega_\beta)}{\Delta^2}, \qquad (3.1b)$$

$$I_\pm=\left(R_1\pm\frac{\omega_\alpha-\omega_\beta}{\Delta}R_3\right)\left(1\mp\frac{\Omega_\pm}{\Gamma}\frac{\gamma_1-\gamma_2}{\Delta}\right)-R_2\left(\frac{\omega_\alpha-\omega_\beta}{\Delta}\right)^2. \qquad (3.1c)$$

These appear as Lamb dips on the resultant line profile, and their magnitude is entirely dependent on the coefficients

$$\begin{aligned}&R_1=\sum_j\frac{1}{\Gamma_j}\left[1-\frac{\gamma_{2j}}{12(\Gamma_j+\gamma_{2j})}-\frac{\gamma_{1j}}{4(\Gamma_j+\gamma_{1j})}\right], \qquad R_2=\sum_j\frac{2}{3}\frac{\gamma_{2j}}{\Gamma_j(\Gamma_j+\gamma_{2j})},\\ &R_3=\frac{1}{4}\left[\frac{\gamma_{2f}}{\Gamma_f(\Gamma_f+\gamma_{2f})}-\frac{\gamma_{1f}}{\Gamma_f(\Gamma_f+\gamma_{1f})}-\frac{\gamma_{2i}}{\Gamma_i(\Gamma_i+\gamma_{2i})}+\frac{\gamma_{1i}}{\Gamma_i(\Gamma_i+\gamma_{1i})}\right],\end{aligned} \qquad (3.2)$$

which are universally expressed in terms of the probabilities of spontaneous and stimulated transitions which compete with one another.

The extreme components of the triplet in Eq. (3.1) can be readily identified according to width and position with a doublet of the Stark or Zeeman structure, and are investigated in detail in the next Section. Depending on the shift of these components as a function of the particle density and the intensity of the corresponding fields, it can readily be identified with Eq. (2.11) and can be used as a source of information about $\gamma_1 - \gamma_2$. There is also considerable interest in the central component of the triplet, which appears exclusively as a result of transitions between sublevels of the combining terms and gives information about their probabilities.

The appearance of the central component in the structure of the Lamb dip can readily be interpreted with the aid of Fig. 3. At any frequency of the standing wave there are, in general, four Bennet "holes" in the populations: two on the α transition and two on the β transition. Their position on the velocity scale is given by the intersection of the horizontal ω = const and the straight lines $\omega^\pm_{\alpha,\beta} = \omega_{\alpha,\beta} \pm kv$. As the frequency is varied, the dips

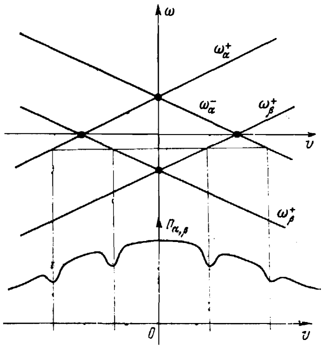

FIG. 3. Interaction of Bennet "holes" in population inversions on α and β transitions.

show a degeneracy three times: twice for $v = 0$ ($\omega = \omega_{\alpha,\beta}$) and once for $v = \pm(\omega_\alpha - \omega_\beta)/2k$, $\omega = (\omega_\alpha + \omega_\beta)/2$. In the first case, the appearance of the Lamb dips is unavoidable, and in the second, it is possible only for rapid relaxation between the sublevels, since opposing waves under these conditions cut the holes in the populations associated with different optical transitions. In point of fact, in the absence of collisional transitions ($\gamma_{2j} \ll \Gamma_j$) the central dip vanishes as $\frac{2}{3}\gamma_{2j}/\Gamma_j$, and for $\gamma_{2j} \gg \Gamma_j$ it becomes symmetric and its intensity is

$$I = \frac{2}{3}\left(\frac{1}{\Gamma_f} + \frac{1}{\Gamma_i}\right)\left(\frac{\omega_\alpha - \omega_\beta}{\Delta}\right)^2, \tag{3.3}$$

which is comparable with the side components so long as $|\omega_\alpha - \omega_\beta| > |\Delta| > |\gamma_1 - \gamma_2|$.

Collapse sets in when the last inequality is reversed, and thereafter the individual components of the Lamb dip are indistinguishable because they are all shifted into the center of gravity of the spectrum, assuming different widths:

$$\mathcal{P} \sim |G|^2\left[\exp\left[\frac{-\Omega^2}{(k\bar{v})^2}\right] - \frac{|G|^2}{24}\left(\zeta_0 + \frac{\zeta\Gamma}{\Gamma^2+\Omega^2} + \frac{\zeta_+\Gamma_+}{\Gamma_+^2+\Omega^2} + \frac{\zeta_-\Gamma_-}{\Gamma_-^2+\Omega^2}\right)\right], \tag{3.4}$$

where $\Gamma_\pm = \Gamma \pm i\Delta/2$ and

$$\zeta_0 = 2R_1\left(\frac{1}{\Gamma_+} + \frac{1}{\Gamma_-} - i\frac{\gamma_1-\gamma_2}{\Delta}\right) - \left(\frac{\omega_\alpha-\omega_\beta}{\Delta}\right)^2\left(\frac{1}{\Gamma_+} + \frac{1}{\Gamma_-} - \frac{2}{\Gamma}\right),$$

$$\zeta = 2\left(\frac{\omega_\alpha-\omega_\beta}{\Delta}\right)^2\left(R_2 + 2R_3\frac{\Omega}{\Gamma}\frac{\gamma_1-\gamma_2}{\omega_\alpha-\omega_\beta}\right), \tag{3.4a}$$

$$\zeta_\pm = 2\left(1 \mp i\frac{\gamma_1-\gamma_2}{\Delta}\right)\left(R_1 \mp R_3\frac{i\Omega}{\Gamma_\pm}\frac{\omega_\alpha-\omega_\beta}{\Delta}\right) - R_2\left(\frac{\omega_\alpha-\omega_\beta}{\Delta}\right)^2.$$

In the special case of equal g factors the formulas in Eq. (3.4) become identical with those obtained by Wang et al.[14] for the normal Zeeman effect:

$$\mathcal{P} \sim |G|^2\left[\exp\left[\frac{\Omega_1^2}{(k\bar{v})^2}\right] - R_1\frac{|G|^2}{6\Gamma_1}\left(1 + \frac{\Gamma_1^2}{\Gamma_1^2+\Omega_1^2}\right)\right],$$
$$\Omega_1 = \omega - \omega_0 - \mu_0 g\mathcal{H}. \tag{3.5}$$

The analogous result is valid in the case of the collapse of the structure when the g factors are not equal.

CONCLUSION

It is clear from the foregoing that analysis of the Stark and Zeeman effects can be used not only to demonstrate the existence of spectral exchange in the optical range, but also to use it to determine important parameters of collisional relaxation which cannot be seen in the absence of the field and in the normal Zeeman effect. There is also the opposite possibility: if the necessary collision invariants have been determined with, say, the anomalous Zeeman effect, then by investigating the quadratic Stark effect for the same atom one can find from the observed shifts the Stark structure constants a_j and b_j which are difficult to calculate theoretically. Equally, it is possible to determine the scale of the transition probabilities between the components of weakly split terms, using the intensity of the additional line in the Lamb dip, provided the radiative widths of these terms are known.

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APPENDIX

Let us suppose that the constants γ_x are complex:

$$\gamma_x = \gamma_x' + i_x\gamma'', \quad \mathrm{Im}\,\gamma_x = \gamma_x'' \neq 0. \tag{A.1}$$

Equation (2.6) then retains its form, and the parameter Δ is expressed in terms of γ_x' and γ_x'' as follows:

$$\Delta = \left(\frac{r+a}{2}\right)^{1/2} + i\left(\frac{r-a}{2}\right)^{1/2},$$

$$r = |[a^2 + 4(\gamma_1' - \gamma_2')^2(\gamma_1'' - \gamma_2'')^2]^{1/2}|, \tag{A.2}$$
$$a = (\omega_\alpha - \omega_\beta)^2 + (\gamma_1'' - \gamma_2'')^2 - (\gamma_1' - \gamma_2')^2.$$

It is clear that $|a| < r$ when $\gamma_1'' \neq \gamma_2''$. This is why in the present case the line is represented by two separated Lorentz shapes with different widths. When $a = 0$ the frequency splitting is equal to the width difference:

$$\mathrm{Re}\,\Delta = \mathrm{Im}\,\Delta = [(\gamma_1' - \gamma_2')(\gamma_1'' - \gamma_2'')]^{1/2} \tag{A.3}$$

and is independent of the field strength. Since in the absence of the field the shifts are usually small in comparison with the collision corrections to the width, it may be supposed that $|\gamma_1'' - \gamma_2''| < |\gamma_1' - \gamma_2'|$. When this inequality is satisfied, the Lorentz profiles approach one another without limit as the pressure increases, and the amplitude of one of them falls to zero.

[1)]The exchange frequency is zero only in the limiting case of strong collisions $\gamma_1 = \gamma_2$ [7, 20]

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