1)In the present case the polynomials $\xi_k(x)$ have degree $k+1$ and a definite parity, equal to $(-1)^{k+1}$:

$$\xi_1={}^1/_2(x^2+1),\quad \xi_2=-{}^1/_{16}(2x^3+7x),$$
$$\xi_3={}^1/_{32}(2x^4+13x^2+20),\quad \xi_4=-{}^1/_{256}(10x^5+99x^3+305x),$$
$$\xi_5={}^1/_{512}(14x^6+191x^4+917x^2+1362),\ \text{etc}.$$

2)Here, the K_i are certain coefficients, depending on the parameters ν, b, and μ. We shall give details of the calculations, and also the explicit form of the coefficients K_i, in another publication.

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Resonance broadening of two-photon *S–S* transitions

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A theoretical investigation is made of the influence of resonance excitation-exchange collisions on the line profile of a two-photon S–S transition. It is shown that the profile differs from the form predicted by the adiabatic collision theory of the broadening. In particular, the ratio of the line width Γ to the shift $\Delta\omega_m$ does not obey the relationship $\Gamma/|\Delta\omega_m| = 1.4$. The characteristics of the broadening in the presence of hyperfine splitting of the levels are considered.

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§1. INTRODUCTION

The theoretically predicted[1] advantages of two-photon absorption as the most accurate method for investigating atomic and molecular transitions have recently been confirmed convincingly by several experimental investigations.[2-5] One of the most attractive features of this method is that it can be used to eliminate in principle and reduce considerably in practice the influence of the Doppler effect on the line width. Therefore, beginning from relatively low pressures of ~1 Torr, when the collisional line width becomes comparable with the radiative width, collisions play the dominant role in the formation of a line profile.

We shall consider the characteristics of the broadening of two-photon transitions in atoms. In a typical experimental situation the line profile of such a transition is formed as a result of collisions with neutral particles, which are atoms and molecules of the same substance in the form of a gas or of an impurity. The van der Waals interaction plays the main role in such collisions and its influence on the broadening of one-photon transition lines has been investigated quite thoroughly.

We shall also assume that the density of the perturbing particles is sufficiently low so that the collision (impact) theory of the broadening can be applied. The broadening of the two-photon transition lines by a foreign gas does not require any special treatment. We can use the standard formulas of the collision theory for the broadening of one-photon absorption spectra.[6]

The situation is different in the case of the broadening of two-photon transition lines by the gas of the same substance. In this case the broadening may be greatly affected by the resonance exchange of excitations in collisions between identical atoms, whose role is not taken into account in the standard adiabatic collision theory of the broadening. The analogous question of the influence of the resonance transfer of excitation on the profile of a one-photon line has been considered on many occa-

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sions.[7-9] It is worth noting the following points. It is known that such processes must be allowed for only in the calculation of the line profile of transitions from the ground atomic state. A considerable contribution to the broadening is made by these processes only at sufficiently high densities when the collisional line width becomes comparable with the radiative width. However, at these densities the optical thickness of a layer becomes very great which makes it extremely difficult to investigate the line profile experimentally. On the other hand, at least one of the states linked by a one-photon transition is degenerate. This degeneracy complicates the problem and its analytic investigation enormously. As a result, it is difficult to separate the effects of the resonance excitation exchange from the effects associated with the level degeneracy.

Both these difficulties disappear in most two-photon absorption spectra. We are speaking here of the S–S transitions from the ground state of an atom, which will be the only transitions considered in the present paper. In this case we can assume that there is no degeneracy, which eases greatly the theoretical study. Moreover, the optical thickness of a layer remains less than unity for practically any gas density.

§ 2. DENSITY MATRIX EQUATION

The profile of a two-photon absorption line representing a transition of an atom from state 1 to state 2 in a field

$$\mathbf{E}=\mathbf{E}_1 \exp\{i\mathbf{k}_1\mathbf{r}-i\omega_1 t\}+\mathbf{E}_2\exp\{-i\mathbf{k}_2\mathbf{r}-i\omega_2 t\}+\text{c.c.}$$

is governed by the density matrix element $\rho_{12}(\mathbf{v})$, where $\mathbf{v}$ is the velocity of the atom. We shall use $\sigma(\mathbf{v})$ to denote the time-independent part of the density matrix:

$$\sigma_{12}(\mathbf{v})=\rho_{12}(\mathbf{v})e^{i(\omega_1+\omega_2)t},\quad \sigma_{ii}(\mathbf{v})=\rho_{ii}(\mathbf{v}).$$

The equation for $\sigma_{12}(\mathbf{v})$ has the form

$$[i(\Delta\omega-\Delta\mathbf{k}\mathbf{v})+\gamma_r]\sigma_{12}(\mathbf{v})=\left(\frac{d\sigma_{12}}{dt}\right)^{\text{coll}}+G, \tag{1}$$

where $\Delta\omega=\omega_{12}-\omega_1-\omega_2$; ω_{12} is the frequency of the $1\to 2$ transition; γ_r is the radiative line width; $\Delta k=k_1-k_2$; G is the composite matrix element of the interaction of the atom with the field, including the sum over all the intermediate states; $(d\sigma_{12}/dt)^{\text{coll}}$ is the collision term describing the relaxation processes. Equation (1) is obtained on the assumption that $\sigma_{11}(\mathbf{v})=1$, $\sigma_{22}(\mathbf{v})=0$.

The expression for $(d\sigma_{12}/dt)^{\text{coll}}$ can be obtained by a method similar to that used in Ref. 7. We note that the increase in the two-particle density matrix $\sigma(\mathbf{v})\sigma(\mathbf{v}_1)$ [$\sigma(\mathbf{v}_1)$ is the density matrix of the perturbing atom] due to one collision is given by the standard expression

$$\Delta[\sigma(\mathbf{v})\sigma(\mathbf{v}_1)]=S^+\sigma(\mathbf{v})\sigma(\mathbf{v}_1)S-\sigma(\mathbf{v})\sigma(\mathbf{v}_1), \tag{2}$$

in which the S collision matrix is a function of the quantum numbers of both atoms. Taking the traces of the left- and right-hand sides of Eq. (2) for the quantum numbers of the perturbing atom and averaging over the collisions, we obtain

$$\left(\frac{d\sigma_{12}(\mathbf{v})}{dt}\right)^{\text{coll}}=\sum_{i,k,\alpha,\beta}\int[S^{*}_{i\alpha,1\beta}S_{k\gamma,2\beta}\sigma_{ik}(\mathbf{v})\sigma_{\alpha\gamma}(\mathbf{v}_1)-\sigma_{12}(\mathbf{v})\sigma_{\alpha\alpha}(\mathbf{v}_1)]F(g)dg, \tag{3}$$

where $F(g)dg$ is the number of collisions with the parameters between g and $g+dg$; the first and third index of S represent the quantum numbers of the investigated atom, whereas the second and fourth are the corresponding numbers of the perturbing atom. In spectral problems we can simplify the right-hand side by retaining only the matrix elements whose resonance frequencies are close (within the limits of the line width of the investigated transition) to the resonance frequency of the element $\sigma_{12}(\mathbf{v})$. Bearing in mind also that the population of the upper state of the atom is zero ($\sigma_{22}=0$), we need to retain on the right-hand side of Eq. (3) only the terms with $i=1, k=2, \alpha=\gamma=1$ and $\alpha=1, \gamma=2, i=k=1$. The sum over the index β still remains on the right-hand side of Eq. (3). However, we can easily see that if $\beta\neq 1$, the S-matrix elements describe nonresonance collisions when one of the atoms is in the initial state and the second is transferred to another energy level after the collision. We can quite accurately regard these elements as equal to zero, i.e., we need retain only the terms with $\beta=1$.

We shall now allow for the influence of a hyperfine structure on Eqs. (1) and (3). We shall consider only the case when none of the field frequencies ω_1 and ω_2 is in resonance with any of the components of the fine structure of virtual states. It is important to note that the orbital momentum of the states 1 and 2 is zero. Then, the composite matrix element G, which is the matrix element of the electrostatic operator, is diagonal in respect of the spin and the total momentum of the transition. Therefore, it is sufficient to use one index F for the matrix elements σ_{12} and this index gives the total momentum of the transition: σ_{12}^F. The S-matrix elements are also matrix elements of the electrostatic operator. Therefore, they differ from zero only if the total momentum is conserved by each atom. On the other hand, the resonance defect in collisions accompanied by, for example, the $1F\to 2F$ transition of the first atom and $2F'\to 1F'$ transition of the second atom is very small for $F\neq F'$ so that we can assume that the S collision matrix is independent of F and F'. We then obtain

$$\left(\frac{d\sigma_{12}{}^F(\mathbf{v})}{dt}\right)^{\text{coll}}=\sigma_{12}{}^F(\mathbf{v})N\int|\mathbf{v}-\mathbf{v}_1|W(\mathbf{v}_1)[S^{*}_{11,11}S_{21,21}-1]2\pi\rho\,d\rho\,d\mathbf{v}_1$$
$$+\sum_{F'}N_{F'}\int|\mathbf{v}-\mathbf{v}_1|W(\mathbf{v}_1)\sigma_{12}{}^{F'}(\mathbf{v}_1)S^{*}_{11,11}S_{12,21}\,2\pi\rho\,d\rho\,d\mathbf{v}_1, \tag{4}$$

where N_F is the total number of atoms at the sublevel F; $\Sigma_F N_F=N$; ρ is the impact parameter; $W(\mathbf{v})$ is the Maxwellian distribution function of the atomic velocities.

To avoid unjustifiable complication of the formulas, we shall use the nonnormalized intensity of two-photon absorption $I(\omega)$:

$$I(\omega)=\operatorname{Re}\sum_F G^{*}N_F\int\sigma_{12}{}^F(\mathbf{v})W(\mathbf{v})d\mathbf{v}. \tag{5}$$

§ 3. S COLLISION MATRIX

We shall consider a collision between two identical atoms, one of which is in the ground state n_1S and the other is in the excited state n_2S. The main contribution to the interaction between these potentials is made by the van der Waals potential. After the collision the atoms may remain in the initial states or they may exchange the excitations. We shall use U_I for the amplitude of the wave function of two atoms corresponding to the case when the first atom is excited and the second is not, and we shall employ U_{II} for the case when the first atom is not excited and the second is excited. The Schrödinger equation for the state amplitudes is

$$i\dot{U}_{\rm I}=V_{\rm I}U_{\rm I}+V_{\rm II}U_{\rm II},\quad i\dot{U}_{\rm II}=V_{\rm I}U_{\rm II}+V_{\rm II}U_{\rm I}, \tag{6}$$

where $V_i(t)=C_i(\rho^2+v^2t^2)^{-3}$, v is the relative velocity of the atoms,

$$C_{\rm I}=-\frac{2}{3\hbar}\sum_{n,n'}\frac{\langle n_1S\|d\|nP\rangle^2\langle n_2S\|d\|n'P\rangle^2}{E_{nP}+E_{n'P}-E_{n_1S}-E_{n_2S}}, \tag{7}$$

$$C_{\rm II}=-\frac{2}{3\hbar}\sum_{n,n'}\frac{\langle n_1S\|d\|nP\rangle\langle nP\|d\|n_2S\rangle\langle n_2S\|d\|n'P\rangle\langle n'P\|d\|n_1S\rangle}{E_{nP}+E_{n'P}-E_{n_1S}-E_{n_2S}}, \tag{8}$$

$\langle nS\|d\|n'P\rangle$ is the reduced matrix element of the dipole moment, and E_{np} is the energy of the atomic states. Solution of the system (6) corresponding to the initial state $U_1(-\infty)=1, U_{II}(-\infty)=0$, is

$$\left.\begin{aligned}U_{\rm I}(+\infty)&=S_{21,\,21}={}^1/_2[\exp\{-i(\eta_{\rm I}+\eta_{\rm II})\}+\exp\{-i(\eta_{\rm I}-\eta_{\rm II})\}],\\ U_{\rm II}(+\infty)&=S_{21,\,12}=S_{12,\,21}={}^1/_2[\exp\{-i(\eta_{\rm I}+\eta_{\rm II})\}-\exp\{-i(\eta_{\rm I}-\eta_{\rm II})\}],\end{aligned}\right\} \tag{9}$$

where $\eta_i=\frac{3}{8}\pi C_i/\rho^5v$.

When both atoms are in the ground state n_1S before a collision, there is only one scattering channel (state amplitude U_0):

$$\left.\begin{aligned}i\dot{U}_0&=V_0U_0,\\ V_0&=C_0(\rho^2+v^2t^2)^{-3},\\ C_0&=-\frac{2}{3\hbar}\sum_{n,n'}\frac{\langle n_1S\|d\|nP\rangle^2\langle n_1S\|d\|n'P\rangle^2}{E_{nP}+E_{n'P}-2E_{n_1S}}.\end{aligned}\right\} \tag{10}$$

Then,

$$U_0(+\infty)=S_{11,11}U_0(-\infty)=e^{-i\eta_0}U_0(-\infty),\quad \eta_0=\frac{3\pi}{8}\frac{C_0}{\rho^5v}. \tag{11}$$

Substituting Eqs. (9) and (11) into Eq. (4), and integrating with respect to the impact parameter ρ, we obtain

$$\left(\frac{d\sigma_{12}^F(\mathbf{v})}{dt}\right)^{\rm coll}=-\sigma_{12}^F(\mathbf{v})\Gamma_+N\int\left|\frac{\mathbf{v}-\mathbf{v}_1}{v_0}\right|^{3/5}W(\mathbf{v}_1)d\mathbf{v}_1 +\sum_{F'}\Gamma_-N_{F'}\int\left|\frac{\mathbf{v}-\mathbf{v}_1}{v}\right|^{3/5}W(\mathbf{v}_1)\sigma_{12}^{F'}(\mathbf{v}_1)d\mathbf{v}_1, \tag{12}$$

where

$$\left.\begin{aligned}\Gamma_\pm&=\frac{\pi}{5}\left(\frac{3\pi}{8}\right)^{2/5}v_0^{3/5}\left[A_\pm\int_0^\infty(1-\cos x)x^{-7/5}dx -iB_\pm\int_0^\infty x^{-7/5}\sin x\,dx\right]\approx 2v_0^{3/5}(A_\pm-i0{,}75B_\pm),\\ A_\pm&=a^{2/5}\pm b^{2/5},\quad B_\pm=\frac{b}{|b|}b^{2/5}\pm\frac{a}{|a|}a^{2/5}, a=C_0-C_{\rm I}+C_{\rm II}, b=C_0-C_{\rm I}-C_{\rm II};\end{aligned}\right\} \tag{13}$$

v_0 is the mean thermal velocity of the atoms.

It follows from Eqs. (12)–(13) that the specific features of the collision integral depend on the relative values of the constants a and b. The following comment should be made. The constant C_0 is negative and is usually much smaller than the absolute value of the constant $C_{\rm I}$, representing the interaction in the excited state. If the constants $C_{\rm I}$ and $C_{\rm II}$ are dominated by the contribution of the same intermediate state, it follows from Eqs. (7) and (8) that $C_{\rm I}=C_{\rm II}$. In the other case, when the constant $C_{\rm I}$ includes a considerable contribution from several nearest intermediate states, we usually find that $|C_{\rm II}|<|C_{\rm I}|$.

By way of example, we shall give these constants for the $3S-4S$ transition in sodium. Calculations were carried out using the tables of Bates and Damgaard given in Sobel'man's book.[6] In the calculation of $C_0=C(3S)$ it was possible to consider only the contribution of the state $3P$: $C_0=1.3\times10^{-30}$ cm^6/sec. In the calculation of $C_{\rm I}$ and $C_{\rm II}$ an allowance was made for the contributions of the $3P$ and $4P$ states: $C_{\rm I}=13.1\times10^{-30}$ cm^6/sec and $C_{\rm II}=3.8\times10^{-30}$ cm^6/sec.

§4. BROADENING OF AN ISOLATED SPECTRAL LINE

We shall consider the broadening of an isolated spectral line when there is no hyperfine structure. Equation (1) with the collision integral (12) then becomes

$$\left.\begin{aligned}&[i(\Delta\omega-\Delta\mathbf{k}\mathbf{v})+\gamma_r]\sigma_{12}(\mathbf{v})=-\sigma_{12}(\mathbf{v})\gamma_+f(v) +\gamma_-\int\left|\frac{\mathbf{v}-\mathbf{v}_1}{v_0}\right|^{3/5}W(\mathbf{v}_1)\sigma_{12}(\mathbf{v}_1)d\mathbf{v}_1+G,\\ &\gamma_\pm=\Gamma_\pm N=\gamma_\pm'+i\gamma_\pm'',\quad f(v)=\int\left|\frac{\mathbf{v}-\mathbf{v}_1}{v_0}\right|^{3/5}W(\mathbf{v}_1)d\mathbf{v}_1.\end{aligned}\right\} \tag{14}$$

From now onward we shall ignore the radiative broadening because it can be included in all the subsequent formulas by the simple substitution $i\Delta\omega\to i\Delta\omega+\gamma_r$.

The integral Eq. (14) cannot be solved exactly because of the presence of the factor $\xi=|(\mathbf{v}-\mathbf{v}_1)/v_0|^{3/5}$ in its kernel. Since the function ξ varies slowly with $\mathbf{v}$ and $\mathbf{v}_1$ in the effective integration domain, we can represent this function in the form $\xi=1+g(\mathbf{v},\mathbf{v}_1)$ and then solve the integral Eq. (14) by the method of successive approximations assuming formally that the function $g(\mathbf{v},\mathbf{v}_1)$ is a small parameter. The zeroth approximation is then taken to be the solution of the integral Eq. (14) in which the function ξ is replaced by unity.

In the first approximation with respect to $g(\mathbf{v},\mathbf{v}_1)$, we find that the absorbed power is

$$\left.\begin{aligned}I(\omega)&=N\,{\rm Re}|G|^2\frac{J}{1-\gamma_-J}\left(1-\frac{1}{1-\gamma_-J}\frac{\Delta J}{J}\right),\\ J&=\int\frac{W(\mathbf{v})d\mathbf{v}}{i(\Delta\omega-\Delta\mathbf{k}\mathbf{v})+\gamma_+},\\ \Delta J&=\int d\mathbf{v}\,d\mathbf{v}_1W(\mathbf{v})W(\mathbf{v}_1)g(\mathbf{v},\mathbf{v}_1)\\ &\left[\frac{\gamma_+}{[i(\Delta\omega-\Delta\mathbf{k}\mathbf{v})+\gamma_+]^2}-\frac{\gamma_-}{[i(\Delta\omega-\Delta\mathbf{k}\mathbf{v})+\gamma_+][i(\Delta\omega-\Delta\mathbf{k}\mathbf{v}_1)+\gamma_+]}\right].\end{aligned}\right\} \tag{15}$$

Continuation of the successive approximation procedure gives a series for $I(\omega)$, which converges only asymptotically when the function g approaches zero: $g\to0$. However, we may expect a satisfactory result if only the first term of the series is included. For $\gamma_-=0$, the result given by Eq. (15) can be compared with the exact line profile obtained directly from Eq. (14):

$$I(\omega)={\rm Re}\,N|G|^2\int\frac{W(\mathbf{v})d\mathbf{v}}{i(\Delta\omega-\Delta\mathbf{k}\mathbf{v})+\gamma_+f(v)}, \tag{16}$$

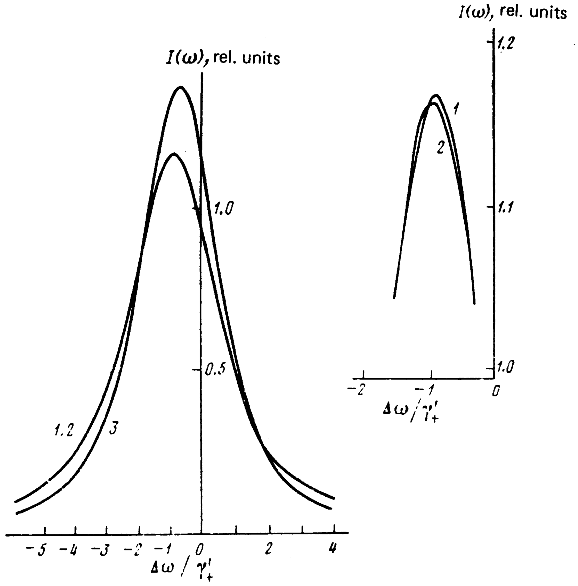

FIG. 1.

and thus determine the sufficiency of the approximation (15). A study of the profile (16) is also of intrinsic interest because the presence of the function $f(v)$ makes the line profile $I(\omega)$ different from the convolution of the Doppler and dispersion profiles given by the usual adiabatic collision theory (see, for example, Ref. 6). As pointed out many times,[10,11] this difference is due to the dependence of the collisional line width on the velocity of the investigated particle and it disappears only in the case $f(v)=1$.

The results of numerical integration of Eqs. (16) and (15) in the $\gamma_-=0$ case show that these expressions give practically identical results for the line profile $I(\omega)$ with any ratio of the Doppler $\Delta k v_0$ and collisional γ'_+ widths. By way of example, Fig. 1 gives the results for the line profile $I(\omega)$ obtained for $\Delta k v_0=\gamma'_+$ from Eqs. (16) (curve 1) and (15) with $\gamma_-=0$ (curve 2), together with the result of convolution $I(\omega)=\operatorname{Re} J$ of the Doppler and dispersion profiles calculated from Eq. (16) for $f(v)=1$ (curve 3). It is clear from Fig. 1 that the convolution $\operatorname{Re} J(\omega)$, which is the zeroth-in respect of the parameter $\Delta J/J$-term of the expansion (15), differs quite considerably from the exact result (16). Inclusion in Eq. (15) of even the first correction in respect of $\Delta J/J$ results in practically complete coincidence of the approximate and exact results. Therefore, we may expect that Eq. (15) is satisfactory also for $\gamma_-\neq 0$.

We shall now consider the profile of the S–S transition line in the presence of resonance collisions such that $\gamma_-\neq 0$. At low pressures characterized by $\gamma'_+/\Delta k v_0 \ll 1$, the profile (15) has the usual Doppler form. At intermediate pressures the line profile differs from the result of convolution of the Doppler and dispersion profiles. We can see from Eq. (15) that this difference is due to two factors: the presence of the integral term in Eq. (14) for the nondiagonal element of the density matrix and the dependence of the line width on the velocity of the investigated particle.

Equations similar to Eq. (14) appear also if allowance is made for the change in the atomic velocity due to collisions. For example, the equation for the classical distribution function in the model of strong collisions discussed in Ref. 12 is completely identical[1)] with Eq. (14) for $g(\mathrm{v},\mathrm{v}_1)=0$. However, this identity is purely apparent. It follows from the derivations of this equation in Ref. 12 that in the model of strong collisions the quantity γ_- represents the gas-kinetic collision frequency and, therefore, its real part is positive. In the case under discussion here the sign of the real part of γ_- is arbitrary and depends on the relative values of the constants C_0, C_{I}, and C_{II}. Moreover, as pointed out above, the most realistic situation is one with negative γ'_-. It is known that in the model of strong collisions[12] at pressures characterized by $\gamma'_-/\Delta k v_0 \sim 1$ an equation of the (14) type describes collisional narrowing of a Doppler line profile. In our case there is no such narrowing because of the negative real part of the constant γ_-. Physically, the absence of narrowing is due to the fact that collisions not only exchange excitations, which is equivalent to a change in the atomic velocity, but also shift the phase of the atomic oscillators, which results in further line broadening and prevents the narrowing.

If the pressure is sufficiently high so that $\gamma'_+/\Delta k v_0 \gg 1$ and the Doppler broadening can be ignored, Eq. (15) becomes

$$\left.\begin{aligned} I(\omega)&=N\operatorname{Re}|G|^2\frac{1}{i\Delta\omega+\gamma_+-\gamma_-}\left[1-q\frac{\gamma_+-\gamma_-}{\gamma_+-\gamma_-+i\Delta\omega}\right]\\ q&=\int d\mathrm{v}\,d\mathrm{v}_1 W(\mathrm{v})W(\mathrm{v}_1)\left(\left|\frac{\mathrm{v}-\mathrm{v}_1}{v_0}\right|^{3/5}-1\right)=\frac{2^{13/10}\Gamma(9/5)}{\sqrt{\pi}}-1\approx 0.29.\end{aligned}\right\}\quad(17)$$

The profile (17) is identical, to within terms of the order of q, with the dispersion profile

$$I(\omega)=N|G|^2\operatorname{Re}\frac{1}{i\Delta\omega+(1+q)(\gamma_+-\gamma_-)}, \quad (18)$$

which can be checked by expanding Eq. (18) as a series in terms of the small parameter q. We recall that in Eqs. (15) and (17) only the terms of the first order in q are retained. Therefore, Eq. (18) describes a line profile with the same precision as Eq. (17). Thus, at high pressures the line profile of a two-photon S–S transition can be represented quite accurately by the dispersion profile of width Γ and shift $\Delta\omega_m$ described by

$$\left.\begin{aligned} \Gamma&=1.3(\gamma_+'-\gamma_-')\approx 5.2 b^{2/5} v_0^{3/5} N,\\ \Delta\omega_m&=1.3(\gamma_-''-\gamma_+'')\approx 3.9\frac{a}{|a|}a^{2/5}v_0^{3/5}N.\end{aligned}\right\}\quad(19)$$

In the classical adiabatic collision theory of the broadening, in which the resonance exchange of excitations in collisions between identical atoms is ignored, there is a very definite ratio of the line width Γ to the absolute shift of its maximum $\Delta\omega_m$. In the van der Waals interaction case this ratio is $\Gamma/|\Delta\omega_m|=1.4$. However, in one-photon linear absorption spectra, when at least one of the levels coupled by a one-photon transition is degenerate, this ratio does not apply because of the considerable role of collisions accompanied by momentum reorientation. One can expect this ratio to apply only to the S–S transitions, for which there is no degeneracy. We can see from Eq. (19) that in the case of braodening by a gas of the same substance, the ratio of the line width to the shift is

$$\Gamma/|\Delta\omega_m| = 1.4(b/a)^{2/5},$$

where $\Gamma/|\Delta\omega_m| \geqslant 1.4$, because $|a| \leqslant |b|$. Consequently, even in the case of the S–S transitions we can expect the ratio $\Gamma/|\Delta\omega_m| = 1.4$ to be valid only for the broadening by a foreign gas when $a = b$.

Thus, the measurements of the ratio of the line width to the shift can be used to deduce-from the difference between this ratio and 1.4-the importance of the influence of collisions with resonance excitation exchange on the broadening of two-photon transition lines.

§5. BROADENING OF HYPERFINE STRUCTURE COMPONENTS

In the presence of a hyperfine structure the equation for the nondiagonal density matrix element, similar to Eq. (14), becomes

$$i(\Delta\omega_F - \Delta\mathbf{k}\mathbf{v})\sigma_{12}^F(\mathbf{v}) = -\sigma_{12}^F(\mathbf{v})\gamma_+ f(v) + \frac{\gamma_-}{N}\sum_{F'} N_{F'} \int \left|\frac{\mathbf{v}-\mathbf{v}_1}{v_0}\right|^{3/5} \sigma_{12}^{F'}(\mathbf{v}_1) W(\mathbf{v}_1)\, d\mathbf{v}_1 + G, \tag{20}$$

where $\Delta\omega_F$ is the frequency detuning of the field from the frequency of the transition of the F component of the hyperfine structure. Applying the same procedure as in the preceding section, we find that in the first approximation with respect to $g(\mathbf{v}, \mathbf{v}_1)$, the absorbed power is

$$\left.\begin{aligned} I(\omega) &= |G|^2 \operatorname{Re}\left[\sum_F N_F(J_F - \Delta J_F)\right]\left[1 - \frac{\gamma_-}{N}\sum_F N_F(J_F - \Delta J_F)\right]^{-1}, \\ J_F &= \int \frac{W(\mathbf{v})\, d\mathbf{v}}{\gamma_+ + i(\Delta\omega_F - \Delta\mathbf{k}\mathbf{v})}, \\ \Delta J_F &= \gamma_+ \int \frac{g(\mathbf{v}, \mathbf{v}_1) W(\mathbf{v}) W(\mathbf{v}_1)}{[\gamma_+ + i(\Delta\omega_F - \Delta\mathbf{k}\mathbf{v})]^2}\, d\mathbf{v}\, d\mathbf{v}_1 \\ &- \frac{\gamma_-}{N}\sum_{F'} N_{F'} \int \frac{g(\mathbf{v}, \mathbf{v}_1) W(\mathbf{v}) W(\mathbf{v}_1)\, d\mathbf{v}\, d\mathbf{v}_1}{[\gamma_+ + i(\Delta\omega_F - \Delta\mathbf{k}\mathbf{v})][\gamma_+ + i(\Delta\omega_{F'} - \Delta\mathbf{k}\mathbf{v}_1)]}. \end{aligned}\right\} \tag{21}$$

We shall first consider the case of low pressures, when the width of the components is much less than the frequency splitting between them. Then, we can investigate the line profile of each of the components retaining only one term in all the sums over F in Eq. (21). We then find that the profile of each of the components is given by Eq. (15) where the substitutions $N \to N_F$, and $\gamma_- \to \gamma_- N_F/N$ have to be made. At higher pressures the Doppler line width may become much smaller than the collisional width of the component well before these components begin to overlap. Under these conditions the line profile of each of the components has the dispersion form of width Γ_F and with a shift of the maximum $\Delta\omega_m^F$ given by

$$\left.\begin{aligned} \Gamma_F &= 2.6 N v_0^{3/5}\left[b^{2/5}\left(1 + \frac{N_F}{N}\right) + a^{2/5}\left(1 - \frac{N_F}{N}\right)\right], \\ \Delta\omega_m^F &= 2 N v_0^{3/5}\left[\frac{b}{|b|} b^{2/5}\left(1 - \frac{N_F}{N}\right) + \frac{a}{|a|} a^{2/5}\left(1 + \frac{N_F}{N}\right)\right]. \end{aligned}\right\} \tag{22}$$

The width and line shift given by Eq. (22) are generally of the same order but their ratio is not $\Gamma_F/|\Delta\omega_m^F| = 1.4$. Finally, at sufficiently high pressures when all the components of the hyperfine structure overlap, the resultant line profile is again given by Eq. (19).

Quantitative characteristics of the broadening of the hyperfine structure components appear most clearly in the limiting case when $|b| \gg |a|$ $(C_{\mathrm{I}} \approx C_{\mathrm{II}}$, $|C_0| \ll |C_{\mathrm{I}}|, |C_{\mathrm{II}}|$. The broadening of an isolated hyperfine structure component can in this case be described by substituting $a = 0$ in Eq. (22). An increase in the density may result first in an overlap of any two components, for example, F_1 and F_2. The profile of the overlapping components has the width and shift given by

$$\left.\begin{aligned} \Gamma &= 2.6 N v_0^{3/5} b^{2/5}\left(1 + \frac{N_{F_1} + N_{F_2}}{N}\right), \\ \Delta\omega_m &= 2 N v_0^{3/5} \frac{b}{|b|} b^{2/5}\left(1 - \frac{N_{F_1} + N_{F_2}}{N}\right). \end{aligned}\right\} \tag{23}$$

The slope of the line $\Gamma(N)$ is then greater and the shift $\Delta\omega_m(N)$ is then less than the slope of the corresponding straight lines for each of the components, although all the isolated components shift in the same direction. Finally, in the case of complete overlap of all the components the width and shift of the resultant dispersion profile are

$$\Gamma = 5.2 N v_0^{3/5} b^{2/5}, \qquad \Delta\omega_m = 3.9 N v_0^{3/5} \frac{a}{|a|} a^{2/5}. \tag{24}$$

It is interesting to note that the line shift is then not only much smaller but may differ in respect to the sign from the shift of isolated hyperfine structure components.

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[1)] Complete identity of the equations is obtained by introducing a new function $\varphi = \sigma_{12}(\mathbf{v}) W(\mathbf{v})$.

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