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SATURATION OF THE DOPPLER SPECTRUM

A. I. BURSHTEĬN

Institute of Chemical Kinetics and Combustion, Siberian Division, Academy of Sciences, USSR

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We calculate the Doppler line contour deformed by the absorption of intense radiation when there are independent mechanisms for thermal broadening and spectral diffusion caused by a change in velocity during collisions. We show that when the second mechanism is dominant the spectral decomposition of the transition probability narrows not only when the gas is compressed, but also when the light intensity is increased, and as a result the absorption spectrum saturates in an unusual manner.

INTRODUCTION

THE change in the contour of a Doppler line under the influence of collisions occurs partially due to a change in velocity during collisions, and partially due to its phase interruption. The two effects have been studied both separately and together but always for vanishingly weak radiation (the absorption of which is, essentially, also of interest). In Rautian and Sobel'man's latest survey [1] of the results referring to the theory of the Doppler effect when there is no saturation, they elucidated rather fully and from a unified point of view the literature devoted to the pressure narrowing of a Doppler line (Dicke effect) both for strong and for weak collisions. The only exception was the calculation made recently by the author [2] which could apparently, owing to a different formalism, not easily be compared with other calculations especially as in that paper the main interest was focused on the relaxation process rather than its spectrum. On the other hand, neither of these facts is important; the general framework of the theory of Markov processes [2,3] enables us not only to obtain more simply all known results referring to the form of Doppler spectra but also to advance further—into the realm of high light intensities saturating the line.

In the present paper we demonstrate this using as an example collisions that change greatly the velocity, although we could extend the calculations also to a more general case. The main and rather unexpected result is that the power of the light plays the same role as the collision frequency, so that the spectral decomposition of the transition probability and of the stationary population of states can be narrowed not only by a compression of the gas but also by an increase in the intensity of the monochromatic wave with which the spectrum is observed. The latter then undergoes a more complex change: the saturation, starting somewhat earlier, masks the effect of the narrowing of the transition probability and as a result of the combination of the two effects the spectrum broadens up to a certain limit which is independent of the power of the field. Only after the Doppler width narrowed by the radiation has become less than the thermal width does saturation again take on its normal character.

1. GENERAL FORMALISM

The Hamiltonian of an atom interacting with a plane monochromatic wave which is in close resonance only with two levels of the spectrum E_1 and E_2 has the form ($\hbar = 1$)

$$\hat{H} = \begin{Vmatrix} E_1 & -\frac{1}{2}\omega_1 \exp(-ikr + i\omega t) \\ -\frac{1}{2}\omega_1 \exp(ikr - i\omega t) & E_2 \end{Vmatrix}, \tag{1.1}$$

where $\omega_1 = D_{12}E_0$ is the frequency of the interaction of the atom with the light (D_{12} is the dipole moment of the transition) which is proportional to the wave amplitude E_0, $k = \omega_0/c$, ω_0 the frequency of the transition $E_2 - E_1$, c the velocity of light, and ω its frequency.

The velocity of the atom v is a Markov variable which remains constant along a mean free path and which changes instantaneously and uncorrelatedly at the moment of collision so that each subsequent value of the velocity is completely independent of the previous value and its probability is given by a one-dimensional Maxwell distribution:

$$\varphi(v) = \sqrt{\alpha/\pi}\exp(-\alpha v^2). \tag{1.2}$$

The Hamiltonian (1.2) depends, however, not directly on v but on the coordinate of the atom, $r = \int v dt$. It is a function of the process (Brownian motion) taking place in the interval (0, t) but not a unique function of the velocity realized at the given moment of time, independent of the prehistory. It is therefore impossible to apply directly to this problem the formalism developed in [2,3].

However, changing from the density matric of the atom ρ to the variable

$$\sigma_{12} = \rho_{12}\exp\left(i\frac{\omega_0}{c}\int v dt - i\omega t\right), \quad n = \rho_{11} - \rho_{22}, \tag{1.3}$$

we can write the Schrödinger equation $i\dot{\rho} = [\hat{H}\rho]$ in the form

$$\dot{n} = i(-\omega_1\sigma_{12} + \omega_1\sigma_{21}), \tag{1.4a}$$

$$\dot{\sigma}_{12} = i(y - \Delta\omega)\sigma_{12} - i\frac{\omega_1}{2}n, \tag{1.4b}$$

where $\Delta\omega = \omega - \omega_0$ and $y = \omega_0 v/c$ is a Markov random variable. Thanks to our choice of representation (which varies randomly in time), the Hamiltonian in (1.4) turns out to be a function of the Markov variable y, and applying to (1.4) the formalism developed for uncorrelated perturbations [3] we can thus easily get for the partial density matrix

$$\dot{n}(t, y) = -i\omega_1\sigma_{12}(t, y) + i\omega_1\sigma_{21}(t, y) - [n(t, y) - \bar{n}(t)]/\tau_0,$$

$$\dot{\sigma}_{12}(t, y) = i(y - \Delta\omega)\sigma_{12}(t, y) - i\frac{\omega_1}{2}n(t, y) - [\sigma_{12}(t, y) - \bar{\sigma}_{12}(t)]/\tau_0,$$

$$\dot{\sigma}_{21}(t,y) = -i(y-\Delta\omega)\sigma_{21}(t,y) + i\frac{\omega_1}{2}n(t,y) - [\sigma_{21}(t,y) - \bar{\sigma}_{21}(t)]/\tau_0, \quad (1.5)$$

or

$$\dot{X}(t,y) = \hat{G}(y)X(t,y) + \frac{1}{\tau_0}\bar{X}(t), \quad (1.6)$$

where τ_0 is the time of free flight between collisions that change the velocity (but not its phase) and

$$\hat{G} = \begin{pmatrix} -1/\tau_0 & -i\omega_1 & +i\omega_1 \\ -i\omega_1/2 & i(y-\Delta\omega)-1/\tau_0 & 0 \\ +i\omega_1/2 & 0 & -i(y-\Delta\omega)-1/\tau_0 \end{pmatrix},$$

$$X = \begin{pmatrix} X_1 \\ X_2 \\ X_3 \end{pmatrix} = \begin{pmatrix} n \\ \sigma_{12} \\ \sigma_{21} \end{pmatrix}, \quad (1.7)$$

while

$$\bar{X}(t) = \int X(t,y)\varphi(y)\,dy = \int X\left(t, \frac{\omega_0}{c}v\right)\varphi(v)\,dv. \quad (1.8)$$

These integro-differential equations can with equal justification be used also in some other problems, in particular, for calculating the absorption of a spectral line widened due to the Doppler effect for non-moving atoms (in a crystal). The problem of the absorption of a strong microwave field by a non-uniformly broadened line in magnetic resonance when spin diffusion is taken into account reduces also to such a problem.

2. STATIONARY TRANSITION PROBABILITY

It has recently been shown[4] that one can obtain a statement about the level of stationary absorption of strong monochromatic radiation and about the non-equilibrium population of states arising under its influence from the usual formulae of the model for transitions

$$N = \frac{\omega_0 n_0 W_s}{1+2W_sT}, \quad n_s = \frac{n_0}{1+2W_sT}, \quad (2.1)$$

if we define the stationary probability W_S for transitions as follows

$$\frac{1}{1/T + 2W_s} = \int_0^\infty e^{-t/T}\bar{n}(t)\,dt \quad (2.2)$$

where $n(0) = 1$, $\rho_{12}(0) = \rho_{21}(0) = 0$. Here n_0 is the equilibrium population of states and T the time for the relaxation of phases and populations under the action of collisions which do not affect the velocity of the particle.

To obtain the information which is of interest to us from Eq. (1.6) we introduce the Laplace transform of $\bar{X}(t)$

$$\bar{L} = \int_0^\infty \bar{X}(t)e^{-t/T}\,dt = \begin{pmatrix} \bar{L}_1 \\ \bar{L}_2 \\ \bar{L}_3 \end{pmatrix}, \quad (2.3)$$

through which we can in a natural way express the probability (2.2):

$$2W_s = 1/\bar{L}_1 - 1/T \text{ when } X(0) = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}. \quad (2.4)$$

In exact correspondence with (2.3) we can define also a partial transform L(y) by means of which $\bar{L}$ can be expressed by a simple averaging over y:

$$L(y) = \int_0^\infty X(t,y)e^{-t/T}dt, \quad \bar{L} = \int L(y)\varphi(y)\,dy. \quad (2.5)$$

If we bear, moreover, in mind that

$$\int_0^\infty \dot{X}(t,y)e^{-t/T}dt = -X(0) + \frac{1}{T}L(y), \quad (2.6)$$

we find, taking the Laplace transform of Eq. (1.6), that

$$L(y) = -\hat{P}(y)\left[X(0) + \frac{1}{\tau_0}\bar{L}\right], \quad \hat{P}(y) = \left[\hat{G}(y) - \frac{1}{T}\hat{E}\right]^{-1}, \quad (2.7)$$

$\hat{E}$ is the unit matrix. Averaging this result over y and solving for $\bar{L}$ we get

$$\bar{L} = \tau_0\left[\left(\hat{E} + \frac{1}{\tau_0}\hat{\bar{P}}\right)^{-1} - \hat{E}\right]X(0), \quad (2.8)$$

where

$$\hat{\bar{P}} = \int\left[\hat{G}(y) - \frac{1}{T}\hat{E}\right]^{-1}\varphi(y)\,dy. \quad (2.9)$$

Using these results in (2.4) we find finally

$$2W_s = \frac{1}{\tau_0[(\hat{E} + \hat{\bar{P}}/\tau_0)^{-1}_{11} - 1]} - \frac{1}{T}. \quad (2.10)$$

3. GENERAL SOLUTION

It is clear from (1.5) and (1.6) that the matrix

$$\hat{P}(y)^{-1} = \hat{G}(y) - \frac{1}{T}\hat{E} = \begin{Vmatrix} -1/T_0 & -i\omega_1 & i\omega_1 \\ -i\omega_1/2 & i(y-\Delta\omega)-1/T_0 & 0 \\ i\omega_1/2 & 0 & -i(y-\Delta\omega)-1/T_0 \end{Vmatrix} \quad (3.1)$$

depends only on the universal combination of the thermal relaxation 1/T and the frequency of change in the velocity $1/\tau_0$

$$\frac{1}{T_0} = \frac{1}{T} + \frac{1}{\tau_0}. \quad (3.2)$$

The matrix which is the inverse of (3.1) averaged over the Maxwell distribution can thus also be expressed in terms universally in terms of T_0:

$$\hat{\bar{P}} = T_0\begin{Vmatrix} 1 - w^2I_0 & -iw(I_0 - iI_1) & iw(I_0 + iI_1) \\ -\frac{iw}{2}[I_0 - iI_1] & \left(1+\frac{w^2}{2}\right)I_0 - iI_1 & \frac{w^2}{2}I_0 \\ i\frac{w}{2}[I_0 + iI_1] & \frac{w^2}{2}I_0 & \left(1+\frac{w^2}{2}\right)I_0 + iI_1 \end{Vmatrix} \quad (3.3)$$

Here

$$I_0(z) = \int_{-\infty}^{\infty}\frac{\varphi(x)\,dx}{1+w^2+(z-x)^2} = I_0(-z),$$

$$I_1(z) = \int_{-\infty}^{\infty}\frac{(z-x)\varphi(x)\,dx}{1+w^2+(z-x)^2} = -I_1(-z) \quad (3.4)$$

are integrals depending on three dimensionless parameters: z, w, and q, which is the width of the distribution (1.2) as a function of $x = \Delta\omega T_0$,

$$\varphi(x) = \frac{1}{q\sqrt{\pi}}\exp\left[-\frac{x^2}{q^2}\right] \quad (3.5)$$

where

$$q = \frac{\omega_0 T_0}{c\sqrt{\alpha}} = T_0\Delta\omega_D, \quad z = T_0\Delta\omega, \quad w = \omega_1 T_0, \quad (3.6)$$

where $\Delta\omega_D$ is the Doppler line width.

Equations (3.3) to (3.6) determine completely the matrix $\hat{\bar{P}}$ defined in (2.9). Since the latter is known, we need for the final result only the purely algebraic operations indicated in (2.10). Performing those we find

$$2W_s = \omega_1^2 T_0 \frac{I_0 - t_0(1+w^2)I_0^2 - t_0 I_1^2}{(1-t_0I_0)^2 - (1+t_0)w^2 I_0(1-t_0I_0) + w^4 I_0^2 + t_0(t_0+w^2)I_1^2} \quad (3.7)$$

where $t_0 = T_0/\tau_0$.

When there is no saturation, i.e., in a vanishingly weak field when we can put w = 0 both in Eq. (3.7) itself as well as in the integrals I_0 and I_1 we get easily Rautian and Sobel'man's result[1] which refers to this case

$$2W_s = \omega_1^2 \tau_0 \frac{t_0 I_0(1-t_0I_0) - (t_0I_1)^2}{(1-t_0I_0)^2 + (t_0I_1)^2}. \quad (3.8)$$

Depending on whether the frequency of change in the velocity or the frequency of the broadening collisions is higher we obtain from this even more particular formulae

$$2W_s = \omega_1^2 \tau_0 \frac{I_0(1-I_0) - I_1^2}{(1-I_0)^2 + I_1^2}, \quad t_0 \to 1 \quad (3.8a)$$

$$2W_s = \omega_1^2 T_0 I_0, \quad t_0 \to 0. \quad (3.8b)$$

The first of these describes the Doppler contour which can be steeply narrowed when the pressure ($1/\tau_0$) is increased (Dicke effect). The second describes a trivial transformation through collisions that interrupt not the frequency of the vibrations but the phase, as a result of which the Doppler line takes on the collision form with a monotonically increasing width when the pressure is increased.

Saturation has in these different limiting cases also a qualitatively different character. In the first case (when $T \to \infty$) we get from (3.7) the following generalization of (3.8a):

$$2W_s = \omega_1^2 \tau_0 \frac{I_0[1-\gamma^2 I_0] - I_1^2}{[1-\gamma^2 I_0]^2 + \gamma^2 I_1^2}, \quad \gamma^2 = 1 + \omega_1^2 \tau_0^2, \quad (3.9)$$

where I_0 and I_1 also depend on γ^2. However, in the case when the collisions disrupting the phase dominate (when $\tau_0 \to \infty$) the correct generalization of (3.8b) has the form

$$2W_s = \frac{\omega_1^2 T I_0}{1 - \omega_1^2 T^2 I_0}. \quad (3.10)$$

The influence of the power of the field (ω_1^2) on these two formulae and through them upon the stationary population of the states and the magnitude of the absorption in (2.1) merits a separate discussion.

4. SATURATION IN THE DICKE EFFECT

To establish what kind of new effect occurs in Eq. (3.9) when $\gamma \neq 1$ it is useful to describe the structure of its solution in all regions of values of the parameters γ, q, and z. Specifying the range of values in terms of the coordinates $z/q = \Delta\omega/\Delta\omega_D$ and $\ln(\gamma/q)$ (Fig. 1) we discover easily that a cut parallel to the axis of the abscissa gives a frequency dependence of the transition probability in units of the Doppler width $\Delta\omega_D$ and the abscissa axis itself is the natural boundary between two opposite situations, differing in the inequality signs in the inequality

$$\gamma / q = \sqrt{1/q^2 + w^2/q^2} \lessgtr 1. \quad (4.1)$$

When $\gamma \gg q$, i.e., in the upper half-plane, we have a Lorentz frequency dependence which is steeply narrowed compared with the Doppler contour width through either pressure or radiation. When $\gamma \ll q$, on the other hand, the spectrum of the transition probability is close to a quasi-static one, at least where it reproduces exactly a Maxwellian (i.e., a Gaussian) distribution.

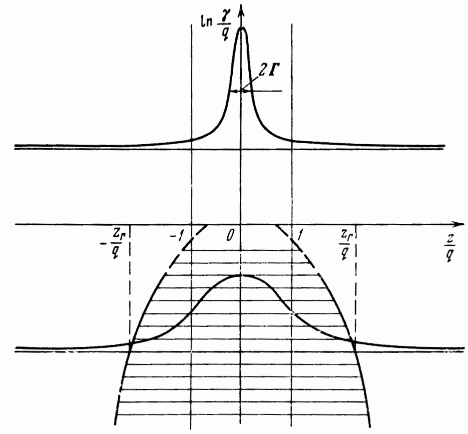

FIG. 1. Separation of physical situations: the region of the occurrence of a Gaussian spectrum is indicated by horizontal shading and its width by the vertical lines |z|/q = 1. The two line contours drawn here illustrate the structure of the spectrum $W_S(\Delta\omega)$ in different sections: quasistatic in the lower half-plane and narrowed by pressure or radiation in the upper half-plane.

To verify this we bear in mind that when $\gamma/q \ll 1$, i.e., in the upper half-plane, the following expansions in a small parameter are valid for the integrals I_0 and I_1 [1)]

$$I_0 = \frac{1}{\gamma^2(1+\alpha^2)}\left[1 - \frac{q^2}{2\gamma^2}\frac{1-3\alpha^2}{(1+\alpha^2)^2} + \frac{3q^4}{4\gamma^4}\frac{1-10\alpha^2+5\alpha^4}{(1+\alpha^2)^4} + \ldots\right], \quad (4.2a)$$

$$I_1 = \frac{\alpha}{\gamma(1+\alpha^2)}\left[1 - \frac{q^3}{2\gamma^2}\frac{3-\alpha^2}{(1+\alpha^2)^2} + \frac{3q^4}{4\gamma^4}\frac{5-10\alpha^2+\alpha^4}{(1+\alpha^2)^4} + \ldots\right], \quad (4.2b)$$

where $\alpha = z/\gamma$. Using them in (3.9) we can obtain after the appropriate calculations

$$2W_s = \omega_1^2 \tau_0 \frac{q^2(z^2+\gamma^2)}{2[z^2(z^2+\gamma^2)^2 + \gamma^2 q^4/4]}. \quad (4.3)$$

This line has a complicated form: at its periphery, i.e., when $z \gg \gamma$ it decreases as $\Delta\omega^{-4}$:

$$2W_s = \omega_1^2 \tau_0 \frac{q^2}{2z^4} = \frac{\omega_1^2 \tau_0}{2}\frac{\Delta\omega_D^2}{\Delta\omega^4} \text{ when } \Delta\omega^2 \gg \frac{1}{\tau_0^2} + \omega_1^2. \quad (4.3a)$$

On the other hand, in the center—when $z \ll \gamma$—it has the usual collision contour

$$2W_s = \omega_1^2 \tau_0 \frac{q^2}{2\gamma^2[z^2 + (q^2/2\gamma)^2]} = \frac{\omega_1^2}{\sqrt{1+\omega_1^2\tau_0^2}}\frac{\Gamma}{\Delta\omega^2 + \Gamma^2}, \quad (4.3b)$$

but with a width

$$\Gamma = \frac{q^2}{2\gamma} = \frac{\Delta\omega_D^2}{2\sqrt{1/\tau_0^2 + \omega_1^2}} \quad (4.4)$$

which decreases not only when the pressure ($1/\tau_0$) is increased, but also when the power in the field (ω_1^2) is increased. When the radiation is weak the narrowing of this contour is due to collisions (Dicke effect) while for relative strong radiation it is due to the radiation.

However, the narrowing of the frequency dependence of the transition probability does not yet mean that the

1) These series are obtained by expanding the Lorentz contours in (3.4) in the vicinity of x = 0. Since $\varphi(x)$ is nearly a δ-function, they converge rapidly.

absorption curve is in fact narrowed. Only the frequency dependence of n_S reproduces a similar dependence of $W_S(\Delta\omega)$, as can be seen from (2.1). As far as the absorbed power of the field is concerned, it has according to (2.1) and (4.3b) the form

$$N=\frac{\omega_0 n_0 \omega_1^2 \Gamma}{2\sqrt{1+\omega_1^2\tau_0^2}\,[\Delta\omega^2+\Gamma^2+\omega_1^2\Gamma T/\sqrt{1+\omega_1^2\tau_0^2}]} \qquad (4.5)$$

when $\Delta\omega^2 \ll 1/\tau_0^2 + \omega_1^2$. This formula is correct for all values of ω_1^2 if $\Delta\omega_D\tau_0 \ll 1$, i.e., provided even for a vanishingly weak field the line would already be narrowed because of the Dicke effect. One sees easily that in that case saturation starts before the inequality $\omega_1\tau_0 < 1$ changes sign.

Indeed, the increase in absorption in the line center stops when the two last terms in the denominator become of the same order of magnitude, i.e., when

$$\Gamma=\frac{\omega_1^2 T}{\sqrt{1+\omega_1^2\tau_0^2}} \quad \text{or} \quad \omega_1^2=\frac{\Delta\omega_D^2}{2}\frac{\tau_0}{T} \ll \frac{\Delta\omega_D^2}{2} \ll \frac{1}{\tau_0^2}. \qquad (4.6)$$

Due to the fact that $\omega_1\tau_0$ is still less than unity the line width Γ is constant as before ($\approx \Delta\omega_D^2\tau_0^2/2$) and the last term in (4.5) increases in proportion to ω_1^2. Indeed, saturation occurs in the usual way: it spreads from the line center outwards. In the saturation region the absorption then approaches its natural limit $N_{max} = \omega_0 n_0/2T$, which is independent of W_S since the speed of absorption of energy is limited by the frequency of the thermal transitions. When ω_1 is comparable to $1/\tau_0$, the width of the saturation region, though it ceases to increase, turns out to be already appreciably larger than the initial one, which moreover begins to narrow ($\Gamma \to \Delta\omega_D^2/2\omega_1$), so that we find, retaining only the last term in the denominator of (4.5)

$$N_0=\frac{\omega_0 n_0 \Delta\omega_D^2}{4\tau_0[\Delta\omega^2+\Delta\omega_D^2 T/2\tau_0]}. \qquad (4.7)$$

Within the width of this line, absorption has its maximum possible value N_{max} when $\Delta\omega^2 \gg \Delta\omega_D^2 T/2\tau_0$, while for $\Delta\omega^2 \gg \Delta\omega_D^2 T/2\tau_0$ it is determined by the expression $\omega_0 n_0 \Delta\omega_D^2/4\tau_0\Delta\omega^2$, which is appreciably less than the maximum value (Fig. 2). Equation (4.7) gives thus the limiting absorption of energy which would, if there were no other relaxation mechanisms (for $T \to \infty$), remain unchanged when the light power increases without limit.

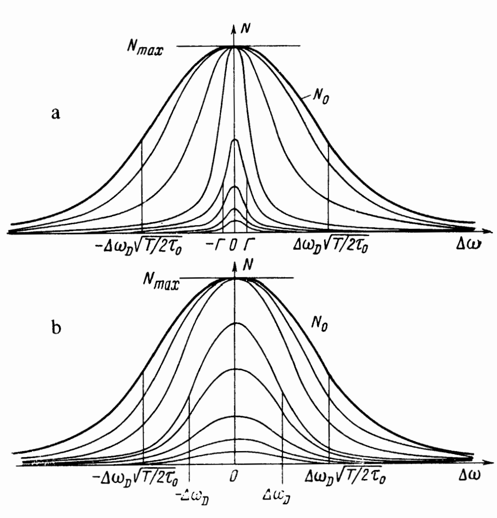

FIG. 2. Form of the saturation of a contour narrowed by pressure (a) and a static contour (b) for $T \gg \tau_0$.

The existence of this limit is the result of the superposition of the effect of the narrowing of $W_S(\Delta\omega)$ through radiation and the usual increase of the probability W_S with ω_1^2 in accordance with the correspondence principle. When the width of the Doppler contour, narrowed by radiation, $\Gamma = \Delta\omega_D^2/2\omega_1$ becomes less than the thermal width ($\Gamma \ll 1/T$) the first cause disappears and only the second one remains: the probability again begins to increase $\sim\omega_1^2$ and saturation takes on its normal character as will be shown in the next section.

Let us now turn to an analysis of the situation which is the opposite of the one considered, and which is realized in the lower half-plane of Fig. 1 when $q/\gamma \gg 1$. Instead of the expansion of (4.2) we have in that region the following approximate estimates for the integrals:

$$I_0=\frac{\pi}{\gamma}\varphi(z)=\frac{\sqrt{\pi}}{\gamma q}\exp\left(-\frac{z^2}{q^2}\right), \qquad (4.8a)$$

$$I_1=0, \qquad (4.8b)$$

valid when $|z| \ll z_\Gamma$ where z_Γ is defined by the following equation[6]:

$$\ln\frac{\gamma}{q}=-\left(\frac{z_\Gamma}{q}\right)^2+2\ln\frac{z_\Gamma}{q}+\ln\sqrt{\pi}, \qquad (4.9)$$

while for $|z| \gg z_\Gamma$ the expansion (4.2) becomes valid as before.[2)] Hence, the periphery of the line ($\Delta\omega \gg \Delta\omega_\Gamma$) is saturated in the way already described above and we are only interested in the Gaussian center of the line when the power increases.

We easily get from (4.8) and (4.9) the following result:

$$2W_s=\frac{\pi\omega_1^2\tau_0\varphi(z)}{\gamma[1-\pi\gamma\varphi(z)]}\approx\frac{\pi\omega_1^2\tau_0\varphi(z)}{\gamma} =\frac{\omega_1^2\sqrt{\pi}\exp\{-\Delta\omega^2/\Delta\omega_D^2\}}{\Delta\omega_D\sqrt{1+\omega_1^2\tau_0^2}}, \qquad (4.10)$$

which is valid everywhere when $\Delta\omega \ll \Delta\omega_\Gamma$ and until $q \gg \gamma = (1+\omega_1^2\tau_0^2)^{1/2}$. In order that this be realized it is in any case necessary that $q = \tau_0\Delta\omega_D \gg 1$, i.e., when there is no saturation ($\gamma = 1$) there can not be an averaged Doppler line contour $\varphi(z)$. The change in the probability with increasing ω_1^2 proceeds then in the following sequence: initially ($\omega_1 \ll 1/\tau_0 \ll \Delta\omega_D$) the probability has its usual form

$$2W_s=\omega_1^2\frac{\sqrt{\pi}}{\Delta\omega_D}\exp\left[-\frac{\Delta\omega^2}{\Delta\omega_D^2}\right], \qquad (4.10a)$$

i.e., it is proportional to the power of the field. Later ($1/\tau_0 \ll \omega_1 \ll \Delta\omega_D$) it changes to the following expression:

$$2W_s=\frac{\omega_1\sqrt{\pi}}{\tau_0\Delta\omega_D}\exp\left[-\frac{\Delta\omega^2}{\Delta\omega_D^2}\right], \qquad (4.10b)$$

which is proportional to the field amplitude, i.e., to the square root of the power and finally when $1/\tau_0 \ll \Delta\omega_D$

[2)]This is connected with the fact that the Lorentz contours in the integrals (3.4) can play the role of almost δ-functions as far as the Gaussian curve (3.5) is concerned only in the limit $|z| \ll z_\Gamma$, while outside this limit the situation is again changed to the opposite one since the Gaussian curve decreases appreciably faster on the periphery.

$\ll \omega_1$ when the sign of the inequality $q/\gamma \gg 1$ is changed, Eq. (4.10) must yield precedence to the previously introduced (4.3), since an increase in the field leads to a shift from the lower half-plane of Fig. 1 to the upper half-plane, where the narrowing of the dependence of $W_S(\Delta\omega)$ on the increase of ω_1 begins.

Saturation starts before this last stage, i.e., already when the center of the line has a Gaussian form and in fact—when $W_S(0)T = 1$, when

$$\frac{\omega_1^2}{\Delta\omega_D^2} = \frac{\sqrt{1+\omega_1^2\tau_0^2}}{\Delta\omega_D T},$$

i.e., when

$$\frac{\omega_1^2}{\Delta\omega_D^2} = \begin{cases} \dfrac{1}{\Delta\omega_D T} \ll \dfrac{1}{\Delta\omega_D \tau_0} \ll 1, & \text{if} \quad \omega_1\tau_0 \ll 1, \\ \dfrac{\omega_1\tau_0}{\Delta\omega_D T} \ll \dfrac{\omega_1}{\Delta\omega_D} \ll 1, & \text{if} \quad \omega_1\tau_0 \gg 1. \end{cases}$$

Hence, the saturation of the central Gaussian part of the spectrum is completely described by Eq. (4.10), which when substituted into (2.1) gives

$$N = \frac{\omega_0 n_0 \omega_1^2 \sqrt{\pi}}{\Delta\omega_D \sqrt{1+\omega_1^2\tau_0^2}\exp(\Delta\omega^2/\Delta\omega_D^2) + \omega_1^2\sqrt{\pi}\,T} \qquad (4.11)$$

but with further increase of the power of the field, when the periphery of the spectrum ($z \gg z_\Gamma$) becomes involved in the saturation, only Eq. (4.5) gives a correct estimate of the absorbed energy and this equation again confirms the existence of a limiting absorption (4.7) to which N tends when ω_1^2 increases without limit, as can be seen from Fig. 2b.

5. NORMAL SATURATION

Qualitatively different results are obtained from Eq. (3.10), which describes the absorption of light by a non-uniformly broadened line when there is no spectral diffusion ($\tau_0 = \infty$). In a vanishingly weak field the spectrum is a group of resonance curves, each with width $1/T$, which is normally distributed. If the width is larger than the width of the distribution $\Delta\omega_D$, we are essentially dealing with the simplest uniformly collision-broadened lines, Lorentzian in form; if, on the other hand, $\Delta\omega_D \gg 1$ the spectrum is mainly Gaussian, it has the form of an enveloping distribution and each of its components saturates independently from the others. The picture of the saturation differs correspondingly.

In the first case, i.e., when $\Delta\omega_D T \ll 1$ we can use the estimate (3.2a) for I_0, retaining only the first term and bearing in mind that $T_0 = T$, $\gamma^2 = 1 + \omega_1^2 T^2$, $z = \Delta\omega T$ we get

$$I_0 = \frac{1}{z^2+\gamma^2} = \frac{1}{z^2+1+\omega_1^2T^2}. \qquad (5.1)$$

Hence we get from (3.10)

$$2W_s = \frac{\omega_1^2 T}{1+\Delta\omega^2T^2}$$

$$N = \frac{\omega_0 n_0 \omega_1^2 T}{2[1+\Delta\omega^2T^2+\omega_1^2T^2]}, \qquad (5.2)$$

as is always the case when homogeneously broadened lines are saturated.

The situation is somewhat more complicated if $\Delta\omega_D T \gg 1$, when the original spectrum is inhomogeneously broadened. It is clear from (4.8) and (4.10) that

$$2W_s = \frac{\omega_1^2\sqrt{\pi}}{\Delta\omega_D\sqrt{1+\omega_1^2T^2}}\exp\left[-\frac{\Delta\omega^2}{\Delta\omega_D^2}\right], \qquad (5.3)$$

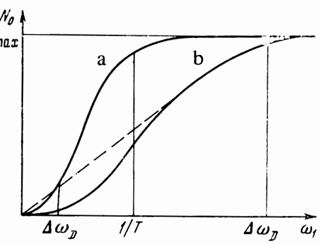

FIG. 3. Saturation under resonance conditions for homogeneous (a) and inhomogeneous (b) broadening of the spectrum.

for all $\Delta\omega \ll \Delta\omega_\Gamma$ while for very weak radiation

$$2W_s = \frac{\omega_1^2\sqrt{\pi}}{\Delta\omega_D}\exp\left(-\frac{\Delta\omega^2}{\Delta\omega_D^2}\right), \qquad (5.3a)$$

in accordance with the correspondence principle, while for $\omega_1 T \gg 1$

$$2W_s = \frac{\omega_1\sqrt{\pi}}{T\Delta\omega_D}\exp\left(-\frac{\Delta\omega^2}{\Delta\omega_D^2}\right) \qquad (5.3b)$$

in spite of that this already appeared in (4.10b). However, in the present case in contrast to the previous one saturation does not start until ω_1 is no longer comparable with $\Delta\omega_D$, i.e., always in the limits where (5.3) is applicable the absorption of light is determined by the general formula

$$N = \frac{\omega_0 n_0 \omega_1^2\sqrt{\pi}}{2[\Delta\omega_D\sqrt{1+\omega_1^2T^2}\exp(\Delta\omega^2/\Delta\omega_D^2)+\omega_1^2\sqrt{\pi}\,T]}$$

$$= \frac{\omega_0 n_0 \omega_1^2\sqrt{\pi}}{2\Delta\omega_D\sqrt{1+\omega_1^2T^2}}\exp\left\{-\frac{\Delta\omega^2}{\Delta\omega_D^2}\right\}, \qquad (5.4)$$

which must be replaced by (5.2) as soon as ω_1 becomes larger than $\Delta\omega_D$. Since, however, at that moment already $\omega_1 T \gg 1$ the limit $\omega_1 = \Delta\omega_D$ is the boundary of saturation proceeding further exactly in the same way as in the previous case (Fig. 3):

$$N = \frac{\omega_0 n_0 \omega_1^2 T}{2[\Delta\omega^2+\omega_1^2T^2]}. \qquad (5.5)$$

This is natural as when $\omega_1 \gg \Delta\omega_D$ the position of all lines in a non-uniform spectrum is identical.

Since broadening by collisions always takes place this result is obtained when the light power increases without limit and when $\tau_0 \neq \infty$ after the width (4.4) narrowed by radiation becomes less than $1/T$ and neglecting the latter, as was assumed in the preceding section, ceases to be valid. Hence, when $\Gamma = 1/T$ and further, i.e., when $\omega_1 > \Delta\omega_D^2 T/2$, the limiting expression (4.7) must yield place to (5.5), and the width of the spectrum starts again to increase appreciably and all resonance sections that are further away will be saturated up to the limiting magnitude $N_{max} = \omega_0 n_0/2T$.

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Translated by D. ter Haar
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