RESONANCE INTERACTION OF UNIDIRECTIONAL WAVES IN GASES

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A theory of interaction between unidirectional waves in a gas is considered. The intensity of one of them is assumed to be weak. The absorption line shape of the weak wave in the presence of the strong wave is determined. The main features in the line shape in weak fields are associated with two-quantum transitions and with the effects of level splitting in a rapidly oscillating field in the case of strong fields. Applications of the theory to spectroscopy and stability problems are considered briefly.

THE interaction of the field of a standing wave with moving atoms and molecules leads to the formation of the well-known Lamb dip[1-4] in the center of the absorption line, with a width equal to the homogeneous transition width. In the interaction of two oppositely traveling waves of the same frequency, when the intensity of one of the waves is very small, new qualitative features appear in the absorption line of the weak wave.[5] At the center of the absorption line of a weak wave a dip appears, whose depth tends toward a constant value with increase in the field strength. This depth depends only on the relaxation constants of the operating levels. For small saturations, the width of the dip is equal to the homogeneous width as in the case of account only of the effects of population.[6] A different situation arises in the interaction of two unidirectional waves, the intensity of one of which is small. Here the specific features in the shape of the absorption line appear not only for large but also for small saturations. The aim of this research is the consideration of the absorption line shape of the weak wave in the presence of the strong. The results of the research are of interest for various applications in spectroscopy and in the solution of the problem of generation stability in gas lasers.

In essence, we consider the line shape of forced resonance scattering with account of level damping. As in [5], we consider the semiclassical theory of interaction of an atom with the field in which both fields are described classically, and the atom, quantum mechanically. By analogy with the scattering of moving classical oscillators, one must expect an essential difference in the forward and backward absorption line shape.[7] The motion of the oscillators adds essential qualitative features to the line shape. In forward scattering, there is complete phase compensation, due to the Doppler frequency shift. Therefore, all the atoms make a contribution independent of their velocity to the scattering at the field frequency. For back scattering, such a compensation does not occur and the atoms radiate the frequency $\omega - 2kv$, which depends on the velocity (ω is the frequency of the field, $k = \omega/c$, v is the projection of the velocity of the oscillator on the direction of propagation of the wave).

Similar phenomena should be expected in the absorption line shape of a weak signal in the presence of a strong one. As will be shown below, there is an essential difference in absorption line shape of unidirectional and oppositely traveling waves, which remains for strong fields. In weak fields, narrow dips appear against the background of the Bennett dip, at the frequency of the strong field, the widths of which are determined by the relaxation constants of the individual levels. This can be explained qualitatively in the following way. The interference of the two waves with neighboring frequencies leads to the modulation of the level population with a beat frequency $\Delta = \omega' - \omega$, where ω and ω' are the frequencies of the strong and weak fields, respectively. The modulation of the difference in populations and, consequently, of the coefficient of absorption, leads to amplitude modulation of the signal of the strong field and to the appearance of an additional signal at the frequency of the weak field. For Δ much less than the width of both levels, the phase of this signal is identical with the phase of the weak signal. The amplitude of the resulting signal at the frequency of the weak field is composed of the amplitude of the initial and the additional signals. Thus, some increase in the intensity of the weak field can be interpreted as a decrease in its absorption. For a frequency deviation Δ comparable with the width of any of the levels, a decrease takes place in the amplitude of the additional signal, and the shift of its phase is comparatively small. If Δ is much greater than the width of both levels, then the medium does not succeed in responding to the change in the instantaneous value of the amplitude of the field, and there is no amplitude modulation of the strong signal. Here the change is essentially only the average difference in populations. Two characteristic regions are already present in the absorption line shape in first order in the saturation and for different relaxation constants. The first is associated with the narrow dip in the frequency of the strong field. The second—the wide part of the line—is connected with the Bennett dip in the velocity distribution of the atoms.

In the strong field, a change takes place in the line shape both in the first region and in the second, and the change in the second region is determined by the parameter γ/Γ, where $\gamma = 2\gamma_1\gamma_2/(\gamma_1 + \gamma_2)$, γ_1 and γ_2 are the widths of the upper and lower levels, Γ the half-width of the line. As in [5], we connect the behavior of the line width in this region with the effects of splitting in a strong and rapidly oscillating field. The absorption line shape of a weak signal was studied previously by Rautian with the field limitation $\gamma\chi/\Gamma \ll 1$ and the relaxation constants $\gamma_1 \gg \gamma_2$, where χ is the saturation parameter.[2] The condition $\gamma\chi/\Gamma \ll 1$ means that the probability of finding the atom at any level does not oscillate, and the effects of splitting do not play an important role. Polarization effects were considered in [8] in the interaction of two fields in the presence of collisions and attention was directed to the features of the absorption line shape. We consider here a gas of two-level atoms in the presence of collisions that suppress and shift the phase, without limitation on the field and the relaxation constants. The new features found qualitatively in the scattering line shape permit us to use them for the measurement of the ratio of the lifetimes of the levels, the widths of the lines and the individual levels, and also for the measurement of the absolute value of the matrix element of the dipole moment.

1. LINE SHAPE

We write down the equations of motion for the elements of the density matrix ρ in the field of two unidirectional waves propagating along the z axis:

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}+\gamma_2\right)n_2 = id[(Ee^{-i\omega t}+E'e^{-i\omega' t})e^{ikz}+\text{c.c.}]\times(\rho_{21}^*-\rho_{21})+\gamma_2 N_2(v),$$

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}+\gamma_1\right)n_1 = -id[(Ee^{-i\omega t}+E'e^{-i\omega' t})e^{ikz}+\text{c.c.}]\times(\rho_{21}^*-\rho_{21})+\gamma_1 N_1(v),$$

$$\left(\frac{\partial}{\partial t}+v\frac{\partial}{\partial z}+i\omega_0+\Gamma\right)\rho_{21} = id(Ee^{-i\omega t}+E'e^{-i\omega' t})e^{ikz}(n_1-n_2). \quad (1)$$

Here E and E′ are the amplitudes of waves having the frequencies ω and ω', respectively; ω_0 is the transition frequency, v the projection of the velocity of the atom on the z axis, $N_1(v)$ and $N_2(v)$ the Maxwell velocity distributions of the atoms at the levels 1 and 2; hd is the dipole matrix element of the transition. Without loss of generality, we assume E to be a real quantity, since this can always be achieved by the choice of the initial instant of time. We set the wave vector $k' = \omega'/c$ equal to $k = \omega/c$, since this leads here only to an error $(\omega - \omega')v/c\gamma \ll 1$ in the exponential. Using the smallness of E′, we find ρ_{21} from perturbation theory and then determine the polarization of the medium. Analysis of the polarization shows that, owing to the weak field at the frequency $\omega' = \omega + \Delta$, an additional polarization arises at the frequencies ω' and the "mirror" frequency $\omega - \Delta$,[1, 8, 9] where Δ is the difference in the frequencies of the weak and strong fields.[1)] For $\Delta \sim \gamma_1, \gamma_2$, the contribution of this polarization also determines the fundamental features in the absorption of the weak signal.

We have for the weak field absorption coefficient α

$$\frac{\alpha}{\alpha_0} = \frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\Gamma(y^2+\Gamma^2)\mathscr{E}(y+\Omega)}{[(y-\Delta)^2+\Gamma^2](y^2+\Gamma_0^2)}dy + \frac{\Gamma}{\pi}\operatorname{Re}\left\{\frac{\chi(\Delta+2i\Gamma)f}{2}\int_{-\infty}^{\infty}\frac{(y-i\Gamma)(y+\Delta+i\Gamma)\mathscr{E}(y+\Omega)}{(y-\Delta-i\Gamma)(y^2+\Gamma_0^2)(y^2+\beta^2)}dy\right\}, \quad (2)$$

where

$$\mathscr{E}(x) = \exp\{-x^2/\omega_d^2\},\quad \Gamma_0 = \Gamma\sqrt{1+\chi},\quad \chi = 4(dE)^2/\gamma\Gamma,$$
$$\beta^2 = -(\Delta+i\Gamma)\cdot[\Delta+i\Gamma(1+\chi f)],\quad f = \gamma(\gamma_{12}-i\Delta)/(\gamma_1-i\Delta)(\gamma_2-i\Delta),$$
$$\gamma_{12} = (\gamma_1+\gamma_2)/2,\quad \Omega = \omega-\omega_0,$$

ω_d is the Doppler line width and α_0 the unsaturated absorption coefficient. The result obtained determines the coefficient of absorption of the weak wave in the presence of a strong one propagating in the same direction. The first term is the absorption coefficient due only to population effects. The additional term takes into account the effect of the strong field on the polarization of the medium.

It is of interest to note the following property. In view of the fact that the additional term, as a function of Δ, is analytic in the upper halfplane,

$$\int_{-\infty}^{\infty}[\text{additional term}]\,d\Delta = 0.$$

For $\omega_d \gg \Gamma_0$, we can take the exponential outside the integral sign at the point y = 0. Then the integrals are computed. For the second integral, it is convenient to close the contour integral in the lower half-plane. This gives

$$\frac{\alpha}{\alpha_0} = \mathscr{E}(\Omega+\Delta) - b\frac{(\Gamma_0+\Gamma)^2}{\Delta^2+(\Gamma_0+\Gamma)^2}\mathscr{E}(\Omega)+\chi\operatorname{Re}\left\{\frac{i\Gamma(\Delta+2i\Gamma)f}{2(\Gamma_0^2-\beta^2)}\times\left[\frac{\beta+\Gamma}{\beta}\frac{\Delta-i(\beta-\Gamma)}{\Delta+i(\beta+\Gamma)}-\frac{\Gamma_0+\Gamma}{\Gamma_0}\frac{\Delta-i(\Gamma_0-\Gamma)}{\Delta+i(\Gamma_0+\Gamma)}\right]\right\}\mathscr{E}(\Omega), \quad (3)$$

where $b = \chi/(1+\chi+\sqrt{1+\chi})$. For $\Delta = 0$, we have

$$\frac{\alpha}{\alpha_0} = \left[\frac{1}{\sqrt{1+\chi}} - \frac{\chi}{2(1+\chi)^{3/2}}\right]\mathscr{E}(\Omega). \quad (4)$$

The expression (4) for α/α_0 at $\Delta = 0$ is identical with that obtained in [2] by another method for $\gamma_2\chi/\gamma_1 \ll 1$ and $\gamma_2/\gamma_1 \ll 1$. Since we made no limitations in the derivation of the formula relative to the relaxation constants and the saturation parameter, (4) is valid for any value of γ/Γ and χ. It is interesting that α/α_0 for $\Delta = 0$ does not depend explicitly on the relaxation constants and is determined only by the saturation parameter.

2. DISCUSSION OF THE RESULTS

As we have already noted, the principal features in the line shape appear near the frequency of the strong field. In the case of weak fields $\chi \ll 1$, Eq. (3) takes the simple form:

$$\frac{\alpha}{\alpha_0} = \mathscr{E}(\Delta+\Omega) - \frac{\chi}{2}\frac{(2\Gamma)^2}{\Delta^2+(2\Gamma)^2}\mathscr{E}(\Omega) - \frac{\chi}{2}\frac{(2\Gamma)^2}{\Delta^2+(2\Gamma)^2}\times\left(\frac{\gamma_1}{\Delta^2+\gamma_1^2}+\frac{\gamma_2}{\Delta^2+\gamma_2^2}\right)\frac{\gamma_1\gamma_2}{\gamma_1+\gamma_2}\mathscr{E}(\Omega) + \chi\frac{\gamma}{8\Gamma}\frac{(2\Gamma)^2}{\Delta^2+(2\Gamma)^2}\left(\frac{\Delta^2}{\Delta^2+\gamma_1^2}+\frac{\Delta^2}{\Delta^2+\gamma_2^2}\right)\mathscr{E}(\Omega). \quad (5)$$

Formula (5) describes several dips in the dispersion shape, the widths of which are determined by the relaxation constants γ_1, γ_2, and Γ.

In the presence of collisions leading to a phase shift, the shape of the sharp peaks does not change. Changes take place only in the wide part of the line shape. When

1) A detailed solution of (1) was given in our preprint No. 12, Institute of the Physics of Semiconductors, Siberian Branch, USSR Academy of Sciences, 1970.

$\Gamma \gg \gamma_1$ and $\Gamma \gg \gamma_2$, the absorption line shape is the sum of three dips in the dispersion shape with half-widths 2Γ, γ_1, and γ_2 and with respective depths $[\chi/2]/2(\gamma_1+\gamma_2)$ and $\chi\gamma_1/2(\gamma_1+\gamma_2)$.

This fact is of interest in spectroscopy and therefore we shall analyze (5) in more detail. The second term is connected with the saturation of the population differences. It describes a dip whose width is twice as great as the Bennett dip and is identical with the width of the absorption line with account only of population effects. The subsequent terms are connected with the appearance of an additional polarization at the frequency of the weak field, due to the strong one. An important difference from the case of oppositely traveling waves is the appearance of additional terms in the absorption coefficient of the weak wave even in first order in the saturation. For $\Delta = 0$, this difference is determined by the second term in (4). It might seem strange that signals with the same frequency and propagating in the same direction have different absorption coefficients (we note that the first term in (4) determines the absorption coefficient of the strong wave[6]). This result must be understood in the following way. In the consideration of the interaction of two fields with the medium, the initial phases of the fields are inconsequential, since we have already assumed that the time of measurement $t_{meas} \gg 1/\Delta$. As $\Delta \to 0$, we assume that the time of measurement should increase without limit, so that $t_{meas}\Delta \gg 1$. (The same result for $\Delta = 0$ can be obtained by averaging over the phase difference of the weak and strong fields.)

For practical applications in spectroscopy, it is of interest to analyze the line shape as a function of the ratio of the relaxation constants. Let collisions be absent; i.e., $\Gamma = (\gamma_1+\gamma_2)/2$. For equal constants, we have $\gamma_1 = \gamma_2$,

$$\frac{\alpha}{\alpha_0} = \mathscr{E}(\Delta+\Omega) - \frac{\chi}{2}\frac{(2\gamma_1)^2}{\Delta^2+(2\gamma_1)^2}\mathscr{E}(\Omega) - \chi\frac{(2\gamma_1)^2}{\Delta^2+(2\gamma_1)^2}\frac{2\gamma_1^2-\Delta^2}{4(\Delta^2+\gamma_1^2)}\mathscr{E}(\Omega). \quad (6)$$

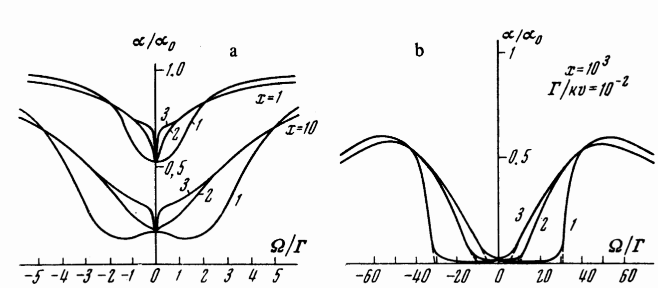
For essentially different constants $\gamma_1 \gg \gamma_2$, we have

$$\frac{\alpha}{\alpha_0} = \mathscr{E}(\Delta+\Omega) - \frac{\chi}{2}\frac{\gamma_1^2}{\gamma_1^2+\Delta^2}\mathscr{E}(\Omega) - \frac{\chi}{2}\frac{\gamma_2^2}{\Delta^2+\gamma_2^2}\mathscr{E}(\Omega). \quad (7)$$

It is seen from a comparison of (6) and (7) that the sharpest changes in the line shape take place for different relaxation constants: a narrow peak appears against the background of the wide part of the dip, with depth $\chi/2$ and half-width γ_2. The last term gives an additional contribution and is significant only for identical γ_1 and γ_2. It can be omitted for very different γ_1 and γ_2 (see (5)).

In the analysis of the absorption line shape in a strong field, we shall distinguish between the behaviors of the broad and narrow parts of the line. Inasmuch as the physical reasons for their generation are different, their characteristic dimensions are also different. The broad region has a size of the order $\Gamma\sqrt{1+\chi}$ and the size of the narrow region is independent of the field, and is determined by the constants γ_1 and γ_2. This makes it possible to consider the behavior of the narrow and broad parts in a strong field separately.

Let us consider the behavior of the absorption coefficient in the narrow region for strong fields ($\chi \gg 1$). Let the relaxation constants be identical initially ($\gamma_1 = \gamma_2 = \gamma = \Gamma$). In this region, we have from (3)



a—Absorption line shape of weak wave for $\chi = 1$ and $\chi = 10$. Curve 1 corresponds to $\gamma_1/\gamma_2 = 1$, 2—$\gamma_1/\gamma_2 = 10$, 3—$\gamma_1/\gamma_2 = 100$;
b—Absorption line shape of weak wave for $\chi = 100$ and $\chi = 1000$. Curve 1—$\gamma_1/\gamma_2 = 1$, 2—$\gamma_1/\gamma_2 = 10$, 3—$\gamma_1/\gamma_2 = 100$. The dashed curve is a plot of (10).

$$\frac{\alpha}{\alpha_0} = \left[\frac{1}{\sqrt{\chi}} - \frac{1}{\sqrt{\chi}}\frac{2\gamma^2+3\Delta^2}{4(\Delta^2+\gamma^2)}\right]\mathscr{E}(\Omega). \quad (8)$$

In the case of different relaxation constants $\gamma_1 \gg \gamma_2$, for $\gamma\chi/\Gamma >> 1$,

$$\frac{\alpha}{\alpha_0} = \left[\frac{1}{\sqrt{\chi}} - \frac{1}{\sqrt{\chi}}\operatorname{Re}\left\{\frac{\sqrt{f}}{\sqrt{f}+1}\right\}\right]\mathscr{E}(\Omega). \quad (9)$$

For strong fields, the depth of the sharp dip is equal to $1/2\sqrt{\chi}$ and the shape of the sharp peak does not depend on the field and is determined by the relaxation constants. We note an important fact: in spite of the condition $\gamma\chi/\Gamma \gg 1$, which corresponds to the equalization of the effective lifetimes of the levels of resonantly interacting atoms ($kv \sim dE$, $\Omega = 0$), the line shape nevertheless depends on γ_2 near $\Delta = 0$. This is connected with the contribution to the absorption of atoms whose velocity gives $kv \gg dE$. In the broad region, with accuracy $1/\sqrt{\chi}$, we have from (3):

$$\alpha/\alpha_0 = 0, \quad |\Delta| < 2dE,$$
$$\frac{\alpha}{\alpha_0} = \frac{|\Delta|\sqrt{\Delta^2-(2dE)^2}}{\Delta^2+\Gamma^2\chi(1-\gamma/\Gamma)}\mathscr{E}(\Delta+\Omega), \quad |\Delta| > 2dE. \quad (10)$$

It is seen from (10) that there are two characteristic regions in the case of a strong field, which are determined by the quantity dE (see Fig. b).

3. PHYSICAL INTERPRETATION

A comparison of the absorption line shape of the weak signal for oppositely traveling waves[5] and unidirectional waves indicates their essential difference in strong and in weak fields.[2)] For weak fields, we associate the difference in the line shape of forward and back scattering with two-photon processes. In the quantum theory of scattering, the two-photon process arises in second order perturbation theory and corresponds to absorption of the original photon with the simultaneous emission of the other photon.[10] In our case, the initial and final states of the atom are identical, which corre-

[2)]The difference in the forward and back scattering line shape was discovered theoretically for a three-level system in [11] and experimentally in [12].

sponds to the case of undisplaced resonance scattering. In the case of the scattering of an atom in the ground state when irradiated by monochromatic light, the scattering line will also be monochromatic. In our case, the initial state of the atom has a finite width, which also leads to some peculiarities of scattering near resonance. Account of the damping of both levels leads to a finite width of the line of two-photon scattering and to the necessity of account of gradual transitions. In the general case, the scattering line is determined by interference of these processes. In the classical interpretation, this corresponds to account of free vibrations of the oscillator for finite times of action of the stimulating force. In a gas, the effect of two-photon processes is clearly evident in the forward scattering (see (5)).

For different relaxation constants of the levels, line structure is clearly evident in the forward scattering line; this consists of several curves of dispersion shape with half-widths equal to the widths of the individual levels. The second term, as we have shown, is connected with population effects and is due to single-quantum stepwise transitions. The third term is associated by us with two-quantum processes.[3)] It describes a dip whose width corresponds to the width of the line of the two-quantum transition. Finally, the last term can be connected with the interference of the gradual and two-quantum processes. For equal relaxation constants, the contribution of this term is largest and the line shape cannot be interpreted as the result only of single-quantum and two-quantum transitions, and is determined by the interference of these two processes.

It is important to note that the scattering line shape does not depend on which of the levels has the longest lifetime. If the lower level has the longest lifetime, then this case corresponds to classical resonance scattering (the transition $1 \to 2 \to 1$). The physically more complicated situation is that in which the atom is excited at the lower short-lived level. Here the two-photon transition $1 \to 2 \to 1$ is unimportant and the principal contribution to the first state of the process is made by the gradual transitions $1 \to 2$ under the action of the external field.

In the following, the principal contribution is made by the two-photon transition $2 \to 1 \to 2$ which also determines the narrow part in the absorption line. It is not difficult to note that both the process $1 \to 2 \to 1$ and the process $2 \to 1 \to 2$ lead to a decrease in absorption. In strong fields, we shall connect the change in the line shape of stimulated radiation, as in [5], with the effect of splitting in a rapidly oscillating field.[13] In this case the basic contribution is made by atoms whose velocities satisfy the resonance condition for splitting. Consideration similar to that given in [5] enables us to determine the velocities of these atoms, which effectively absorb the weak field:

$$kv_{1,2} = \pm 2\sqrt{\Delta^2 - 4(dE)^2}.$$

[3)]We note that the back-scattering line represents a line of a stepwise transition. With twice the width of the Bennett dip. [6] This does not mean that the two-photon processes are absent in the irradiation of an individual moving atom. Here the contributions of the two-quantum and contribution from the interference of two quantum and gradual processes depend on the velocity of the atom. In averaging over the velocities, these contributions cancel each other.

It is thus seen that at $\Delta < 2dE$ there are no resonant atoms. This means that, in the region of detuning $\Delta < 2dE$, the absorption coefficient is equal to zero. The resonance atoms appear only for $\Delta > 2dE$, which leads to an increase in the absorption of the weak wave.

4. APPLICATIONS OF THE THEORY

A. Problems of stability. The results can be used, just as in [5], for the analysis of the stability closely-lying modes in a gas laser. Joint use of the results of [5] and of this research essentially solves the problem of the stability and selection of oscillation modes for $\Omega \gg \Gamma_0$ in lasers with nonlinear absorption. (For more detail, see [5].) An exception is the case of the strong field of a standing wave, when the generation frequency is located close to the center of the line, since this requires the consideration of the absorption line shape in the presence of two strong fields.

B. Spectroscopic applications. The results of the research open up qualitatively new possibilities for investigations with a view toward obtaining the fundamental spectroscopic constants. In experiments on the study of the Lamb dip, the basic spectroscopic information is connected with the measurement of the transition line width. Investigation of the line shape of stimulated forward scattering allows us to obtain not only the width of the line but also the relaxation constants of the individual levels. Evidently, this method can be very simple and reliable for the measurement of the lifetimes of the excited long-lived states.

We note that for strongly differing relaxation constants, it is convenient to study the forward scattering line. For slightly differing constants, it is better to use the method of oppositely-traveling waves (see [5]).

We emphasize still another new and important application, from our point of view. We have in mind the direct observation of level splitting in a gas in a strong, rapidly oscillating field and the measurement of the splitting quantity 2dE. The measurement of the absolute value of the field immediately gives the value of the matrix element d.

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