A nonlinear theory of broadening and a generalization of the Karplus-Schwinger formula

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A theory of broadening is developed which is nonlinear in the light intensity E_0^2. It is based on the following assumptions: *a*) The absorption of the light is due not to transitions in the atom, but to inelastic transitions in the compound "atom+field" system caused by collisions with the broadening particles, i.e., by optical collisions (OC). *b)* The OC-induced light absorption is determined by the OC cross sections, which depend on the collision parameters, as well as on the frequency detuning $\Delta\omega$ and the external-field strength E_0. *c*) The total absorbable light power Q is determined by the balance equations—with allowance for OC and inelastic (e.g., radiative) relaxation—for the level populations of the "compound system." The solution to the problem splits up into two types, a static solution and an impact solution, whose regions of applicability depend on $\Delta\omega$ and E_0. The nature of the dependence of the impact and static solutions on the field strength E_0 is considered. The kinetics of light absorption in a medium is considered on the basis of the solution to the dynamical problem determining the OC cross sections. A general formula for the absorbable power Q is derived and has the structure of the well-known Karplus–Schwinger formula, but contains an elastic-relaxation parameter T_2 that depends nonlinearly on $\Delta\omega$ and E_0. It yields the relations of the standard broadening theory for small E_0 and the Karplus–Schwinger result in the impact region. The dependence of T_2 on E_0 and $\Delta\omega$ leads to the appearance of new nonlinear effects in the absorption of light by a medium.

1. INTRODUCTION

The theory of spectral-line broadening [1-4] was developed primarily for the problems of radiation transfer and diagnostics. Usually the light field E_0 in these problems can be considered weak. More precisely, it can be assumed that: a) an atom "does not have time" in between collisions to undergo a field-induced transition; b) the field has no effect on a broadening-collision event. Therefore, in the theory of broadening the effect of the field E_0 on the shape (contour) of the spectral line is neglected, and light absorption is treated as a set of events of photon absorption by atoms broadened "beforehand" by collisions.

The problems connected with the interaction of powerful resonance radiation with matter became pressing with the appearance of lasers. For laser fields the condition (a) is usually not fulfilled. A theory free from this limitation was constructed by Karplus and Schwinger [5] (see also [6], Sec. 17). The result obtained by Karplus and Schwinger has, in our opinion, two main limitations. First, it does not yield in the weak-field limit all the results of the theory of broadening: It yields an expression for the line contour that is valid only in the region close to the center, i.e., in the impact region. Consequently, in the case of arbitrary fields E_0 this result also does not generalize the theory of broadening with sufficient completeness. Second, in the indicated theory is used the above-stated assumption (b), according to which the characteristics of the broadening collisions enter into the kinetic equations in the form of terms that do not depend on the field parameters (see, for example, [6], p. 126). The possibility of the violation of this approximation is examined in Pestov and Rautian's recent paper [7]. A similar problem concerning the scattering of an electron by an atom in a resonance field has been considered by Hahn and Hertel [8] (for a discussion of this work, see [9]). These investigations pertain, however, to only the impact region, and thereby do not describe the entire line contour.

In the present paper we attempt to construct a nonlinear broadening theory free from the limitations a) and b), i.e., a theory that generalizes the results of the standard broadening theory to the case of arbitrary fields and frequency shifts $\Delta\omega$. The analysis is based on a consistent use of the idea that the atom and the field constitute a compound system such that the transitions occurring in it are responsible for the absorption of light by the medium. We are able to derive for the total dissipatable light power an expression that coincides in form with the Karplus-Schwinger result (see Sec. 7). However, the fundamental parameters figuring in it are, in the general case, not relaxation constants, but complicated functions of the frequency and the field strength. In the weak-field limit the obtained expression yields the results of the standard (linear) theory of broadening; for the frequency region near the line center (i.e., in the impact region), the Karplus-Schwinger result; in the general case it describes complex nonlinear effects.

The analysis is carried out in the framework of the scheme usually adopted in broadening theory [1-4]: the motion of the broadening particles is assumed to be classical and rectilinear; the condition for the collisions to be binary encounters, i.e., the condition $N\rho_{eff}^3 \ll 1$, where N is the density of the broadening particles and ρ_{eff} is the effective impact parameter, which determines the contribution to the corresponding collision cross section, is assumed to be satisfied.

2. OPTICAL COLLISIONS. THE BASIC SYSTEM OF EQUATIONS

Let us consider the absorption of light energy by atomic particles X and Y colliding in the external electromagnetic field

$$E(t)=E_0\cos\omega t. \tag{2.1}$$

The frequency ω is close to the natural frequency ω_0 of the transition between the states 1 and 2 of the X atom (Fig. 1)[1)]. We shall henceforth call such collisions opti-

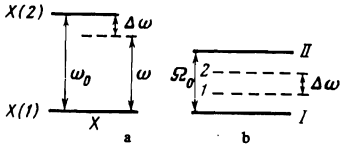

FIG. 1. Term diagram of: a) the atom X and b) the "X + E" compound system.

cal collisions (OC). This term is borrowed from the standard broadening theory, where it was introduced by Weisskopf for collisions that destroy the phase of the oscillations of the atom.

In the present paper we regard optical collisions as isolated events, in the course of which the atoms interact only with each other and with the electromagnetic field. The other interactions that occur in the course of an OC are assumed to be unimportant. This means, in particular, that we neglect the extraneous relaxation of the states X(1) and X(2) during an OC, i.e., we assume that in the absence of interactions with the atom Y and the field (2.1) these states are strictly discrete states.

In the framework of the assumed approximations we can speak of discrete states of some compound system composed of the atom X and the field E. The states of the "atom X + field E" compound system are more convenient for the description, since it is precisely the transitions induced between these states by the collisions with the Y atoms that are responsible for light absorption in the course of an OC.

Let the total Hamiltonian describing the states of the colliding atoms and the field have the form

$$\hat{H}=\hat{H}_X+\hat{H}_E+\hat{V}_{XE}+\hat{V}_{XY}(t). \qquad (2.2)$$

Here $\hat{H}_X$ and $\hat{H}_E$ are the Hamiltonians of the free X atom and the field E respectively, $\hat{V}_E$ is their interaction operator; $\hat{V}_{XY}$ is the X–Y interaction operator, the motion of the X and Y nuclei being, as has already been indicated, assumed to be classical and rectilinear: $R^2 = \rho^2 + v^2t^2$ (where ρ is the impact parameter and v is the relative velocity of the atoms). For convenience, we shall henceforth treat the electromagnetic field (2.1) as an ensemble of n_ω quanta of frequency ω; its Hamiltonian is included in $\hat{H}$. When, however, we go over in the specific expressions for the matrix elements to the classical limit $n_\omega \to \infty$, we shall use the quantity E_0.

1. Let us first consider the standard treatment of photon absorption in the course of an OC. Since in such a transition the number of photons before and after the collision is assumed to be fixed, it is natural to use the system of eigenfunctions $\varphi_1 = |X(1), n_\omega\rangle$ and $\varphi_2 = |X(2), n_\omega - 1\rangle$ of the Hamiltonian $\hat{H}_0 = \hat{H}_X + \hat{H}_E$. For the amplitudes a_1 and a_2 of these states we have the equations

$$ia_1=U_1a_1+Ve^{i\Delta\omega t}a_2, \quad ia_2=U_2a_2+Ve^{-i\Delta\omega t}a_1, \qquad (2.3)$$

where $U_k(t) = \langle\varphi_k|\hat{V}_{XY}(t)|\varphi_k\rangle$, $\hat{V} = |\langle\varphi_1|\hat{V}_{XE}|\varphi_2\rangle|$ and $\Delta\omega = \omega - \omega_0$; in a diapole transition $V = DE_0$, where $D = E_0^{-1}|(\mathbf{d}\cdot\mathbf{E}_0)_{12}|$.

The system (2.3) is the basic system of equations for the standard broadening theory, which is valid for sufficiently small field strengths E_0 (i.e., for sufficiently small V). Indeed, for arbitrary V it is difficult to even establish the initial conditions for (2.3), since V does not go to zero as $t \to \infty$ (V = const). Therefore, V is assumed to be small, so as to make the probability of a transition occurring in the absence of the broadening interaction (when $U_1(-\infty) = U_2(-\infty) = 0$) negligibly small in comparison with the transition probability in the course of an OC. Then setting one of the amplitudes equal to unity as $t \to -\infty$ and the other equal to zero (to be specific, $a_1(-\infty) = 1$ and $a_2(-\infty) = 0$), and solving (2.3) with the aid of perturbation theory, we find

$$|a_2(\infty)|^2=V^2\left|\int_{-\infty}^{\infty}dt\exp\left\{i\left[\Delta\omega t-\int^t\varkappa(\tau)d\tau\right]\right\}\right|^2, \qquad (2.4)$$

where $\varkappa = U_2 - U_1$ is the term shift due to the interaction.

The expression (2.4) determines the probability of emission (absorption) in one OC of a photon with a frequency shift $\Delta\omega$. The resulting contour arising from many OC is, generally speaking, not equal to the sum of the independent terms. This is due to the finiteness of the mean free time of the atom, which leads to the correlation of the individual events in which photons of frequency close to the line center (i.e., for which $\Delta\omega = 0$) are emitted. But this fact is important only in the narrow frequency region $\Delta\omega \lesssim \gamma_{im}$, where γ_{im} is the impact width of the line (see, for example, [2]). In the region $\Delta\omega \gg \gamma_{im}$ (which may be called the "single-particle" region[2)]), however, the line contour $I(\Delta\omega)$ is completely determined by the probability of a transition occurring in an OC with one Y particle and is proportional to the number of such OC that occur in unit time. Notice that the region of values $\Delta\omega \lesssim \gamma_{im}$ is, in the case of binary collisions, fairly narrow. It is located deep inside the impact region of the spectrum, so that practically all the characteristics of the line contour—in particular, the transition between the impact and static broadening mechanisms [2,3]—are describable by the single-particle approximation.

For the single-particle region it is convenient to transform (2.4) with the aid of integration by parts[3)], which corresponds to the exclusion of the nonbinary region $\Delta\omega \lesssim \gamma_{im}$. The first term is proportional to the delta function $\delta(\Delta\omega)$, and vanishes at $\Delta\omega \neq 0$, while the second yields

$$|a_2(\infty)|^2=\left|\int_{-\infty}^{\infty}dt\frac{V}{\Delta\omega}\varkappa(t)\exp\left\{i\left[\Delta\omega t-\int^t\varkappa(\tau)d\tau\right]\right\}\right|^2. \qquad (2.5)$$

It can be seen from a comparison of (2.5) and (2.4) that there has occurred in the single-particle region an effective replacement of the "transition potential" V by $V\varkappa/\Delta\omega$. This seemingly formal circumstance has a profound physical meaning in connection with the investigation of the transitions in the compound system (see below). Notice that the expression (2.5) is not quite correct. It diverges upon passage to the limit, i.e., $\lim(V/\Delta\omega) = \infty$ for $\Delta\omega \to 0$ and $\Delta\omega \gg \gamma_{im}$, which is a direct indication of the limited applicability of the conventional conception of strong fields.

2. Let us proceed to the case of arbitrary V. In this case we cannot speak of the absorption of an individual photon: to describe the dissipation of the light energy in the course of an OC, we must consider the transitions between the states of the Hamiltonian $\hat{H}_{XE} \equiv \hat{H}_X + \hat{H}_E + \hat{V}_{XE}$ of the compound system, states which are characterized by the eigenfunctions (see [12], p. 168; Eng. Transl., p. 135):

$$\psi_I=b_1\varphi_1+b_2\varphi_2, \quad \psi_{II}=b_2\varphi_1-b_1\varphi_2, \qquad (2.6)$$

where $b_{1,2} = 2^{-1/2}(1 \pm \Delta\omega/\Omega_0)^{1/2}$ with $\Omega_0 = \sqrt{\Delta\omega^2 + 4V^2}$. Everywhere below we shall use the capital letters (K, K′) and the Roman numerals (I, II) to denote quantities pertaining to the compound system; the small letters (k, k′)

and the Arabic numerals (1, 2) will be used to denote quantities characterizing the states φ_1 and φ_2.

For the amplitudes of the states I and II of the compound system we obtain from (2.3) with allowance for (2.6) the expressions

$$i\dot{a}_I=U_I a_I+V_{I\,II}e^{i\omega_0 t}a_{II},\quad i\dot{a}_{II}=U_{II}a_{II}+V_{II\,I}e^{-i\omega_0 t}a_I,$$

where

$$U_I=b_1^2U_1+b_2^2U_2;\quad U_{II}=b_2^2U_1+b_1^2U_2;\quad V_{I\,II}=V_{II\,I}=b_1b_2(U_2-U_1)$$

After the substitution

$$b_K=a_K\exp\left(i\int U_K\,d\tau\right)$$

we have

$$i\dot{b}_I=b_{II}\varkappa\frac{V}{\Omega_0}\exp\left\{i\left[\Omega_0 t-\frac{\Delta\omega}{\Omega_0}\int^t\varkappa(\tau)\,d\tau\right]\right\},$$

$$i\dot{b}_{II}=b_I\varkappa\frac{V}{\Omega_0}\exp\left\{-i\left[\Omega_0 t-\frac{\Delta\omega}{\Omega_0}\int^t\varkappa(\tau)\,d\tau\right]\right\}. \qquad (2.7)$$

The system (2.7) is the basic system of equations describing the transitions connected with the dissipation of the energy of the field in course of an OC.

The role of the potential inducing the transition in the compound system is, as can be seen from (2.7), played by the quantity $\kappa V/\Omega_0$, which, for $\Delta\omega \gg 2V$, goes over into the effective potential $\kappa V/\Delta\omega$ of the formula (2.5). It is clear from this that the Spitzer transformation, which separates out the term responsible for light absorption in the course of an OC, corresponds to a transition to the states of the compound system in the weak-field case when $V \ll \Delta\omega$. The above-noted incorrectness of the formula (2.5) is connected with its inapplicability when $V \gtrsim \Delta\omega$.

If we are interested in the single-particle region $\Omega_0 \gg \gamma_{im}$, then the initial conditions for the basic system (2.7) can be formulated for $t \to -\infty$. Choosing them in the form $b_I(-\infty) = 1$ and $b_{II}(-\infty) = 0$ for the $I \to II$ transition probability w, we have: $w = |b_{II}(\infty)|^2$.

It is clear from (2.7) that, in contrast to the results (2.4) and (2.5) of the standard broadening theory, the probability w of a transition occurring in an OC is, generally speaking, not proportional to the light intensity E_0^2 ll V^2. This is a direct indication of the inseparability in the general case of the two elementary events: the broadening-collision and the light-absorption events. It is clear from the foregoing that nonlinear effects should appear when $V \gtrsim \Delta\omega$. As will be shown below, however, they can appear in fairly weak fields $V \gtrsim \Delta\omega$.

Let us derive an expression for the energy dissipated in one OC event. The computation of the energy dissipated in any transition (including OC) amounts to finding the mean energy of the field in the states of the compound system before and after the transition. If some state of the compound system is characterized by the wave function $\psi(t) = b_I(t)\psi_I + b_{II}(t)\psi_{II}$, then the mean (the quantum-mechanical average taken over arbitrary initial phases of the statistical ensemble) value $\langle H_E\rangle$ of the field energy is given by the relation

$$\langle H_E\rangle=|b_I|^2\langle H_E\rangle_I+|b_{II}|^2\langle H_E\rangle_{II},\quad \langle H_E\rangle_K\equiv\langle K|\hat{H}_E|K\rangle. \qquad (2.8)$$

In the optical collision the compound system was initially in the state $b_I(-\infty) = 1$, $b_{II}(-\infty) = 0$. After the collision the system turned out to be in the state $b_I(\infty)$, $b_{II}(\infty)$. For the change in the mean energy of the field occurring in one OC we have

$$\langle H_E(-\infty)\rangle-\langle H_E(\infty)\rangle=\hbar\omega(b_1^2-b_2^2)\,|b_{II}(\infty)|^2=\hbar\omega\frac{\Delta\omega}{\Omega_0}w. \qquad (2.9)$$

Thus, the energy dissipated in one OC event is proportional to the probability w of the $I \to II$ transition in the compound system. Therefore, to compute the energy absorbed by the medium as a result of OC, we need simply find the number of OC occurring per unit time. For this purpose it is convenient to introduce the concept of an "optical-collision cross section:"

$$\sigma(v, E_0, \Delta\omega)=2\pi\int_0^\infty d\rho\,\rho w(\rho, v, E_0, \Delta\omega). \qquad (2.10)$$

These cross sections are generalizations of the OC cross sections introduced by Weisskopf in the conventional theory of broadening (see [1], as well as [3], p. 465).

In the single-particle region $\Omega_0 \gg \gamma_{im}$ we can speak of populations N_I and N_{II} of the states I and II of the "X + E" compound system. Then, introducing with the aid of (2.10) the optical-collision rate Γ_{OC} defined by

$$\Gamma_{OC}=N_Y\langle v\sigma(\Delta\omega, v, E_0)\rangle, \qquad (2.11)$$

where N_Y is the density of the broadening atoms Y and $\langle\ldots\rangle$ denotes averaging over the velocity, we can easily find the light power Q_{OC} absorbed as a result of OC by a unit volume of the medium:

$$Q_{OC}=\hbar\omega\frac{\Delta\omega}{\Omega_0}\Gamma_{OC}(N_I-N_{II}). \qquad (2.12)$$

In computing the total light energy absorbed by the medium, it is necessary to bear in mind, besides the OC, another dissipation channel connected with the mixing of the states of the atom and the field. Let us illustrate the role of this channel with the aid of a simple example.

Let the interaction with the field be momentarily switched on at the moment of time t = 0, and let all the atoms be in the lower state X(1) at $t < 0$. The energy of the field before the interaction is switched on is equal to $n_\omega\hbar\omega = \langle H_E(t < 0)\rangle$. At $t > 0$ the wave function of the compound system has the form: $\psi(t) = b_1 e^{i\Omega_0 t/2}\psi_I + b_2 e^{-i\Omega_0 t/2}\psi_{II}$. The mean energy of the field in this state is, according to (2.8), equal to $\langle H_E(t > 0)\rangle = \hbar\omega(n_\omega - 2V^2/\Omega_0^2)$. Hence the change in the mean energy of the field due to the mixing is equal to

$$\langle H_E(t<0)\rangle-\langle H_E(t>0)\rangle=2V^2\hbar\omega/\Omega_0^2. \qquad (2.13)$$

Notice that the mixing of the states of the atom X and the field occurs in a time $\sim\Omega_0^{-1}$, and, if $\Omega_0 \gg \Gamma_{OC}$, then the optical collisions can be neglected during this time (i.e., it can be assumed that the field instantaneously mixes the states). Then in solving the kinetic equations in which the OC are allowed for, it can be assumed that at the initial moment of time $N_I(0) = b_1^2N$ and $N_{II}(0) = b_2^2N$, where N is the total X-atom concentration. In this case we have for $N_I(t)$ and $N_{II}(t)$ the balance equations

$$dN_I/dt=\Gamma_{OC}N_I-\Gamma_{OC}N_{II},\quad N_I+N_{II}=N. \qquad (2.14)$$

Their solution under the indicated initial conditions with allowance for (2.12) yields

$$Q_{OC}=\hbar\omega\frac{\Delta\omega^2}{\Omega_0^2}\frac{N}{2}2\Gamma_{OC}e^{-2\Gamma_{OC}t}. \qquad (2.15)$$

The total energy W_{OC} absorbed by the medium as a result of OC is equal to

$$W_{OC}=\int_0^\infty Q_{OC}(t)\,dt=\hbar\omega\frac{N}{2}\frac{\Delta\omega^2}{\Omega_0^2}. \qquad (2.16)$$

Thus, in the considered simple example the dissipation of the field energy occurs in two stages: first, during a time $t \sim 1/\Omega_0$ an amount of energy equal to $(4V^2/\Omega_0^2)N\hbar\omega/2$ is dissipated as a result of the mixing (2.13), and then during a time $t \sim \Gamma_{OC}^{-1}$ the energy $W_{OC} = \hbar\omega N\Delta\omega^2/2\Omega_0^2$ is dissipated owing to the OC. The total dissipated light energy is equal simply to $N\hbar\omega/2$, a result which corresponds to the total equalization of the populations of the atomic states.

3. THE NATURE OF THE FREQUENCY AND FIELD-STRENGTH DEPENDENCES OF THE OC CROSS SECTIONS

The nature of the solution to the basic system (2.7) is determined by the relationship between the three quantities: Ω_0, the level spacing in the compound system; $\kappa\Delta\omega/\Omega_0$, the diagonal interaction matrix element; $\kappa V/\Omega_0$, the off-diagonal matrix element. The fundamental role in this case is played by the Weisskopf frequency $\Omega_W = v/\rho_W$ and the Weisskopf radius ρ_W. The quantity ρ_W can be found as the impact parameter for which the change in the phase of the wave function (owing to the matrix elements $\kappa V/\Omega_0$ and $\kappa\Delta\omega/\Omega_0$) during the flight is equal to unity. For the power-law interaction $\kappa = C_n R^{-n} = C_n(\rho^2 + v^2t^2)^{-n/2}$, for which concrete results are given below, we have $\rho_W = (C_n/v)^{1/(n-1)}$.

1. If $\Omega_0 \ll \Omega_W$, then we can neglect Ω_0 in (2.7) in comparison with the other parameters. In this case (2.7) reduces to a differential equation with constant coefficients. The solution of this equation yields, after substituting it into (3.4), the expression

$$\sigma = \frac{2V^2}{\Omega_0^2}\sigma_W, \quad \sigma_W = 2\pi\int_0^\infty d\rho\,\rho\left[1-\cos\int_{-\infty}^{\infty}\varkappa(\tau)\,d\tau\right]. \tag{3.1}$$

The region of values $\Omega_0 \ll \Omega_W$ corresponds to fast transits. In analogy to broadening theory, we shall call it the impact region.

Notice that the system (2.7) for $\Delta\omega \ll 2V$ is equivalent to the resonance case in atomic collisions; the result of the standard impact theory of broadening follows from (3.1) when $\Delta\omega \gg 2V$.

2. The region $\Omega_0 \gg \Omega_W$, the quasi-static region, corresponds to slow transits. Here two main cases can be presented: a) the term crossing case, when there exists a point t_0 at which $\kappa(t_0) = \Omega_0^2/\Delta\omega$; b) the term repulsion case, when there is no point of intersection.

The case (a) corresponds to the Landau-Zener approximation [12,13], which is valid under the conditions that $\kappa\Delta\omega/\Omega_0 \sim \Omega_0 \gg \kappa V/\Omega_0$, Ω_W and $\kappa\Delta\omega > 0$, or

$$\Delta\omega \gg \Omega_W, V; \quad \varkappa\Delta\omega > 0. \tag{3.2}$$

For the OC cross section in this case, we have (cf. [14])

$$\sigma = 4\pi(C_n/\Delta\omega)^{2/n}\Lambda(E_0^2/E_{cr}^2). \tag{3.3}$$

Here $E_{cr} = V_{cr}/D$,

$$V_{cr} = [n(\Delta\omega)^{(n+1)/n}v/2\pi C_n^{1/n}]^{1/2} \equiv V_{LZ} = DE_{LZ} \tag{3.4}$$

is the critical value of the field;

$$\Lambda(x) = \frac{1}{2}\int_0^1 dy\, e^{-x/\sqrt{1-y}}(1-e^{-x/\sqrt{1-y}}) \approx \begin{cases} x, & x \ll 1 \\ x^{-1}e^{-x}, & x \gg 1 \end{cases}.$$

Notice that the broadening-theory result for the static wing of the line follows from (3.3) in the weak-field limit, when $E_0 \ll E_{cr}$; in the case when $E_0 \gg E_{cr}$, the cross section is observed to fall off exponentially.

The case b) corresponds to the situation, first considered by Stückelberg (see [13], p. 651), when $\kappa\Delta\omega/\Omega_0 \ll \kappa V/\Omega_0$, i.e.,

$$V \gg \Delta\omega, \Omega_B. \tag{3.5}$$

The OC cross section is then given by the relation

$$\sigma \propto \exp[-(V/V_{cr})^{(n-1)/n}], \tag{3.6}$$

where

$$V_{cr} = (C_n^{-1/n}v)^{n/(n-1)}. \tag{3.7}$$

The exponential decrease of the OC cross section with increasing E_0 in both the Stückelberg and Landau-Zener cases is due to the fact that the external field pushes the terms of the compound system apart.

Notice that the critical field $E_0 \sim \Delta\omega/D$, E_{LZ}, E_S is, by definition, much smaller than the magnitude of the characteristic atomic field $E_{at} = 0.5 \times 10^{10}$ V/cm. The most interesting nonlinear effects connected with OC-induced light absorption appear in fields $E_0 \sim E_W = \Omega_W/D$, which, under normal gas-kinetic conditions, are relatively low ($\sim 10^4$–10^5 V/cm).

The regions of applicability of the above-considered approximations are shown in Fig. 2. The impact approximation, $\Omega_0 \ll \Omega_W$, is valid inside a circle of radius Ω_W in the plane of the variables $\Delta\omega$ and V, and the static approximation, $\Omega_0 \gg \Omega_W$, is valid outside this circle. When $V > \Delta\omega$, there arise nonlinear effects in both the impact region (the resonance case) and the static region (the Stückelberg case). When $V \ll \Delta\omega$, the conventional (linear) theory of broadening is valid almost everywhere. An exception is the Zener region, which lies above the curve defined by $E_0 = E_{LZ}$, E_{LZ} being given by (3.4). The right- and left-hand sides of the figure are distinguished by the sign of the quantity $\kappa\Delta\omega$ (i.e., by the presence or absence of a point of intersection of the terms). Our treatment is valid outside a circle of radius $\gamma_{im} = N_Y\langle\sigma_W v\rangle$ (i.e., when $\Omega_0 \gg \gamma_{im}$), which is represented by the blackened semicircle in Fig. 2.

If we draw in Fig. 2 the straight line V = const parallel to the $\Delta\omega$ axis, then this line will determine the line contour for a given field strength E_0. Notice that what we have in mind here is the contour corresponding to only the OC-induced light absorption; to obtain the line contour corresponding to the total energy absorption, we must, generally speaking, take the kinetic effects into account (see below). The principal distinctive feature of the OC contour is, as can be seen from Fig. 3, the appearance of a "transparency window," which is responsible for the decrease in the light-absorption probability

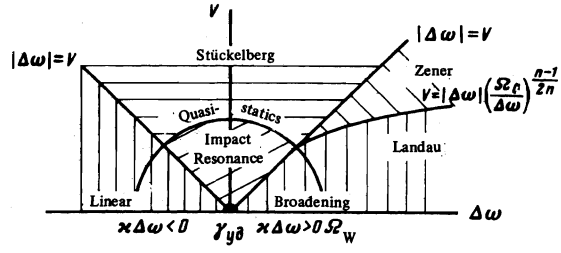

FIG. 2. Composite picture of the physical domains of variation of the OC cross section.

in the region $V < V_{cr}$. The shape of the "transparency window" is determined by the value of V. Thus, for $V < \Omega_W$ (the straight line V = const passes through the circle $\Omega_0 = \Omega_W$), a "plateau" is formed near the line center (the curve 1 in Fig. 3); for $V > \Omega_W$ exponential decays are observed in the region $\Delta\omega < V_{cr}$. It is characteristic that the dip in the central region (the "transparency window") broadens with increasing V.

The straight line $\Delta\omega$ = const in Fig. 2 determines the dependence of the OC cross section on the field strength E_0. The nature of such a dependence for different values of $\Delta\omega$ is shown in Fig. 4. The principal distinctive feature of such a dependence is the exponential decrease of the OC cross section in fields stronger than the critical field. In a number of cases this decrease can be smoothed out as a result of an additional averaging over the angle between the vectors $\mathbf{d}_{12}$ and $\mathbf{E}_0$, which can lead to a power-law decrease for the same critical fields.

4. THE KINETICS OF LIGHT ABSORPTION IN THE PRESENCE OF INELASTIC RELAXATION

Let us consider the problem of light absorption in a medium composed of the atoms X and Y, when there can occur between the states 1 and 2 of the X atom inelastic relaxation characterized by the rates: γ_{12} for the X(1) → X(2) transition and γ_{21} for the X(2) → X(1) transition. This relaxation can be due to spontaneous transitions, as well as to inelastic electron impacts. Let us make two assumptions, on the basis of which the kinetic model discussed below is constructed:

a) We shall assume the inelastic relaxation in the course of an OC event to be unimportant, which amounts to the imposition of the condition

$$v/\rho_{\text{eff}} \gg 2\gamma_{\text{ir}} = \gamma_{12}+\gamma_{21} \quad (\Omega_W \gg \gamma_{\text{ir}}); \tag{4.1}$$

b) we shall assume that the field E_0 does not influence the inelastic relaxation, i.e., that the transitions 2 → 1 and 1 → 2 occur in intervals of time much shorter than Ω_0^{-1}. For inelastic electronic collisions this implies that the X-atom—electron collision time is short compared to Ω_0^{-1}; for spontaneous decay this condition amounts to the requirement that $\Omega_0 \ll \omega_0$. Both conditions are usually easy to fulfill.

Generally speaking, the inelastic relaxation takes the "X + E" compound system into new states I_+, II_+, I_-, and II_- differing from I and II in the number of quanta (see Fig. 5). The states in Fig. 5a correspond to the wave functions $\varphi_1 = |X(1), n_\omega\rangle$, $\varphi_2 = |X(2), n_\omega - 1\rangle$, $\varphi_1^+ = |X(1), n_\omega + 1\rangle$, $\varphi_2^+ = |X(2), n_\omega\rangle$, $\varphi_1^- = |X(1), n_\omega - 1\rangle$, and $\varphi_2^- = |X(2), n_\omega - 2\rangle$. The states in Fig. 5b correspond to the wave functions $\psi_{I_\pm} = b_1\varphi_1^\pm + b_2\varphi_2^\pm$ and $\psi_{II_\pm} = b_1\varphi_2^\pm - b_2\varphi_1^\pm$. Notice that, owing to the very inelastic transition, the energy of the field does not change[4]: only the energy of the X atom changes, and the dissipation of the light energy occurs during the remixing of the new atomic states with the field, i.e., during the formation (after the transition) of the states $K_\pm$ of the "X + E" compound object. The differences in population of the states K, $K_\pm$ of the compound object can be neglected in the balance equations, since in the classical-field limit of interest to us here $n_\omega \gg 1$, so that $n_\omega \pm 1 \approx n_\omega$. Then the light power dissipated on account of the inelastic relaxation is given by the expression

$$\begin{aligned} Q_{\text{ir}} = {} & N_{\text{I}}(\langle H_E\rangle_{\text{I}\,\text{I}_+}\gamma_{\text{I}\,\text{I}_+} + \langle H_E\rangle_{\text{I}\,\text{I}_-}\gamma_{\text{I}\,\text{I}_-} + \langle H_E\rangle_{\text{I}\,\text{II}_+}\gamma_{\text{I}\,\text{II}_+} \\ & + \langle H_E\rangle_{\text{I}\,\text{II}_-}\gamma_{\text{I}\,\text{II}_-}) + N_{\text{II}}(\langle H_E\rangle_{\text{II}\,\text{II}_+}\gamma_{\text{II}\,\text{II}_+} + \langle H_E\rangle_{\text{II}\,\text{II}_-}\gamma_{\text{II}\,\text{II}_-} \\ & + \langle H_E\rangle_{\text{II}\,\text{I}_+}\gamma_{\text{II}\,\text{I}_+} + \langle H_E\rangle_{\text{II}\,\text{I}_-}\gamma_{\text{II}\,\text{I}_-}). \end{aligned} \tag{4.2}$$

Here $\langle H_E\rangle_{KK'_\pm} \equiv \langle H_E\rangle_K - \langle H_E\rangle_{K'_\pm}$ is the difference between the field energies in the respective states of the compound object (cf. Sec. 3), i.e., the energy dissipated in the transition $K \to K'_\pm$; the quantities $\gamma_{KK'_\pm}$ are the respective transition rates. The $K \to K'_\pm$ transition rates $\gamma_{KK'_\pm}$ are determined by the squares of the respective inelastic-interaction matrix elements taken between the wave functions ψ_K, $\psi_{K'_\pm}$ of the compound system. To compute them, it is sufficient to multiply the transition rates γ_{12} and γ_{21} in the atom by the squares of the amplitudes, b_K^2 and $b_{K'}^2$, with which the wave functions of the atom enter into the wave functions of the initial and final states of the compound object.

Let us discuss separately the inelastic relaxation induced by spontaneous radiative transitions. The spontaneous transitions produce the following three lines: the principal line at frequency $\omega_{sp} = \omega$, to which the transitions $I \to I_-$ and $II \to II_-$ contribute, as well as two satellite lines at frequencies $\omega_{sp} = \omega \pm \Omega_0$ produced respectively by the transitions $II \to I_-$ and $I \to II_-$ (see Fig. 5b). Multiplying the rate of the appropriate transition by the light energy dissipated in that transition, we obtain for the spontaneous-radiation power of the components of the triplet the expressions:

$$Q_{\text{cr}} = \hbar\omega\gamma_{\text{cr}}\frac{V^2}{\Omega_0^2}\begin{cases} \left(1-\dfrac{\Delta\omega}{\Omega_0}\right)N_{\text{I}}, & \omega_{\text{cn}} = \omega - \Omega_0, \\ N = N_{\text{I}} + N_{\text{II}}, & \omega_{\text{cn}} = \omega, \\ \left(1+\dfrac{\Delta\omega}{\Omega_0}\right)N_{\text{II}}, & \omega_{\text{cn}} = \omega + \Omega_0, \end{cases} \tag{4.3}$$

where γ_{sp} is the radiative X(2) → X(1) decay rate.

If we neglect the influence of the inelastic relaxation

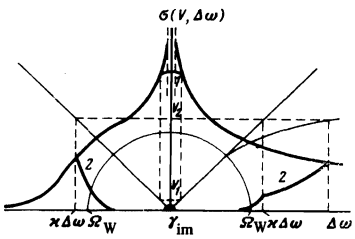

FIG. 3

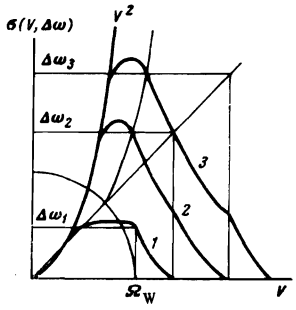

FIG. 4

FIG. 3. Dependence of the OC cross section on $\Delta\omega$ for different V: the curve 1 corresponds to the case when $V \equiv V_1 < \Omega_W$, the curve 2, to the case when $V \equiv V_2 > \Omega_W$.

FIG. 4. Dependence of the OC cross section on V for different $\Delta\omega$: the curve 1 corresponds to the case $\Delta\omega \equiv \Delta\omega_1 < \Omega_W$, the curves 2 and 3, to the cases $\Delta\omega = \Delta\omega_2, \Delta\omega_3 > \Omega_W$.

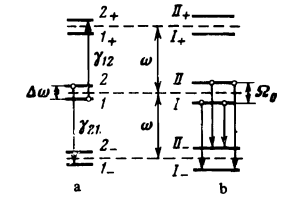

FIG. 5. Energy-level diagram of the "X atom + field E" compound system: a) without allowance for the interaction V between the atom and the field; b) with allowance for the interaction V. The arrows in the figure (b) correspond to spontaneous transitions.

on the population, then $N_I = N_{II} = N/2$ (see below) and the entire picture coincides with the corresponding results of the theory of spontaneous emission in a strong field (see [15], as well as [6]).

We shall obtain the subsequent results on the basis of the balance equations for the two-level scheme in the stationary case:

$$\frac{dN_I}{dt}=0=(\gamma_{II\,I}+\Gamma_{OC})N_{II}-(\gamma_{I\,II}+\Gamma_{OC})N_I, \quad N_I+N_{II}=N. \tag{4.4}$$

Here $\gamma_{III} = \gamma_{III_+} + \gamma_{III_-}$ and $\gamma_{III} = \gamma_{III_+} + \gamma_{III_-}$. The transitions $K \to K_\pm$ are neglected here, although $\gamma_{KK_\pm}$ are different from zero. This is connected with the above-noted fact that the states K, $K_\pm$ are indistinguishable in the limit $n_\omega \to \infty$ (when the transitions $K \to K_\pm$ are formally taken into account, their contributions to the population balance cancel each other out).

Solving (4.4) and substituting the solution into (4.2), we obtain

$$Q_{ir}=2\hbar\omega\gamma_{ir}\Delta N\frac{V^2}{\Omega_0^2}\frac{2\Gamma_{OC}+\gamma_{ir}}{\Gamma_{OC}+{}^1/_2\gamma_{ir}(1+\Delta\omega^2/\Omega_0^2)}, \tag{4.5}$$

where $\Delta N = (\gamma_{21} - \gamma_{12})N/(\gamma_{21} + \gamma_{12})$ is the number of active atoms in zero field and $\gamma_{ir} = (\gamma_{21} + \gamma_{12})/2$ is the inelastic-relaxation rate in the two-level system. For the field power dissipated on account of OC we have from (2.12) and (4.4) the expression

$$Q_{OC}=\hbar\omega\gamma_{ir}\Delta N\frac{\Delta\omega^2}{\Omega_0^2}\frac{\Gamma_{OC}}{\Gamma_{OC}+{}^1/_2\gamma_{ir}(1+\Delta\omega^2/\Omega_0^2)}. \tag{4.6}$$

Thus, the total light power $Q = Q_{ir} + Q_{OC}$ dissipated in the medium is given by the expression

$$Q=\hbar\omega\gamma_{ir}\Delta N\frac{\Gamma_{OC}+2V^2\gamma_{ir}/\Omega_0^2}{\Gamma_{OC}+{}^1/_2\gamma_{ir}(1+\Delta\omega^2/\Omega_0^2)}. \tag{4.7}$$

5. ANALYSIS OF THE NONLINEAR EFFECTS IN LIGHT-ABSORPTION KINETICS

Let us consider the expression (4.7). Notice, to begin with, that the inelastic and OC-induced relaxations can be characterized by the same type of quantities after introducing the notation

$$\Gamma_{ir}=\frac{2V^2}{\Omega_0^2}\gamma_{ir}, \quad \gamma_{OC}=\frac{\Omega_0^2}{2V^2}\Gamma_{OC}. \tag{5.1}$$

Then in the impact region ($\Omega_0 \ll \Omega_W$) $\gamma_{OC} = \gamma_{im} = N_Y\langle\sigma_W v\rangle$ (see (3.1)); in the static region ($\Omega_0 \gg \Omega_W$) $\gamma_{OC} < \gamma_{im}$. With the aid of (5.1) we can rewrite the expression for the total absorbed power (4.7) in the form

$$Q=Q_{sat}\frac{\Gamma_{OC}+\Gamma_{ir}}{\Gamma_{OC}+\Gamma_{ir}(1+\Delta\omega^2/2V^2)}=Q_{sat}\frac{\gamma_{OC}+\gamma_{ir}}{\gamma_{OC}+\gamma_{ir}(1+\Delta\omega^2/2V^2)}. \tag{5.2}$$

Here $Q_{sat} = \hbar\omega\gamma_{ir}\Delta N$ is the power absorbed in the saturation regime; it is the maximum power that can be absorbed by the medium for a given inelastic relaxation.

The relation (5.2) formally has the same form as the Karplus-Schwinger result [5]. The only difference consists in the fact that (5.2) does not contain the additive correction $(\gamma_{OC} + \gamma_{ir})^2$ to the quantity $\Delta\omega^2$. This is quite understandable, since in deriving (5.2) we used the relation $\Omega_0 \gg \max(\gamma_{OC}, \gamma_{ir})$, which allows us to consider the balance equation for the populations and not the kinetic equations for the density matrix. As has already been indicated in Sec. 2, because the collisions are binary encounters, the difference in question is important only in the narrow central part of the impact region where $\Omega_0 \leq \gamma_{ir} + \gamma_{im} \ll \Omega_W$.

In order to clearly demonstrate the connection between the result (for Q) obtained here and the known result, let us rewrite (5.2) in the notation used in the book [6], introducing for this purpose the indicated correction to the frequency shift

$$Q=2\hbar\omega\Delta N\frac{T_2^{-1}(\Delta\omega, V)V^2}{\Delta\omega^2+T_2^{-2}+4V^2T_1T_2^{-1}(\Delta\omega, V)}. \tag{5.3}$$

Here $T_1^{-1} = 2\gamma_{ir}$ and $T_2^{-1} = \gamma_{OC} + \gamma_{ir}$. The relation (5.3) formally coincides with the formula (17.71) in [6]. In contrast to the well-known result, however, the quantity T_2, which plays the role of a generalized elastic-relaxation parameter, is here a complex function of the field characteristics $\Delta\omega$ and V. Therefore, in actual fact, the relation (5.3) coincides with the Karplus-Schwinger formula only in the impact region $\Omega_0 \ll \Omega_W$, where $\gamma_{OC} = \gamma_{im}$, and does not depend on $\Delta\omega$ and V. For weak fields $V \to 0$ (see the linear-broadening-theory region in Fig. 2), if the inelastic-relaxation rate is negligible compared to the OC rate (i.e., if $\gamma_{ir} \ll \gamma_{OC}$), then the relation (5.3) yields the well-known result of the binary-collision broadening theory [11,16], which describes the entire line contour with allowance for the transition from impact to static broadening. Thus, the formula (5.3) is a general result of the nonlinear broadening theory, and is valid for arbitrary field strengths E_0 and frequency detunings $\Delta\omega$. It is interesting that just as the general result of the conventional broadening theory has the structure of the (impact) Lorentz formula with a "variable width" [11,16], the result (5.3) of the nonlinear broadening theory has the structure of (also the impact) Karplus-Schwinger formula [5] with a variable "elastic-relaxation" parameter $T_2(\Delta\omega, V)$.

Of interest is the analysis of not only the expression for the total power Q, but also of the dependence on $\Delta\omega$ and V of the quantities Q_{ir} and Q_{OC}, since they, like Q, can be directly measured in experiments (see Sec. 6). Let us, therefore, consider the dependence on $\Delta\omega$ and V of the expression (5.2), as well as of the ratio Q_{OC}/Q, which illustrates the contribution of OC to light absorption. In investigating these expressions, we shall not introduce the additive correction to $\Delta\omega^2$, limiting ourselves to the region $\Omega_0 \gg \gamma_{im} + \gamma_{ir}$, since it is precisely in this region that the new nonlinear effects appear. The expressions in question can be represented in the simple forms

$$Q=Q_{sat}\frac{U}{U+R}, \quad \frac{Q_{OC}}{Q}=1-R, \tag{5.4}$$

after introducing the dimensionless parameters

$$U=\frac{2V^2}{\Delta\omega^2}, \quad R=\frac{\gamma_{ir}}{\gamma_{ir}+\gamma_{OC}}. \tag{5.5}$$

By definition, $R \leq 1$. Consequently, for $2V^2 \gg \Delta\omega^2$, the phenomenon of saturation is observed independent of the medium characteristics and no matter what the relation between the contributions of the OC and the inelastic relaxation is: the absorbable power attains its maximum value ($Q = Q_{sat}$) and ceases to depend on the characteristics of the field (let us recall that $\Omega_0 \gg \gamma_{im} + \gamma_{ir}$). When the contribution of the optical collisions is large, i.e., when $\gamma_{OC} \gg \gamma_{ir}$ (the pressure-induced broadening is large), then $R \ll 1$, and the saturation sets in considerably earlier when $2V^2 \gg R\Delta\omega^2$. The behavior of the quantity R is of the following nature. For small $\Omega_0 \lesssim \Omega_W$, usually, $R \ll 1$ under gas-kinetic conditions, since $\gamma_{OC} \sim \gamma_{im} \gg \gamma_{ir}$. As Ω_0 increases, the quantity

γ_{OC} decreases and, consequently, R increases. The dependences $R(\Delta\omega, V)$ and $Q(\Delta\omega, V)$ can, in each specific case, be comparatively simply derived on the basis of the results obtained in Sec. 3. Here we investigate in greater detail the qualitative form of the V dependence of Q for the values of $\Delta\omega$ corresponding to the static wing (see Fig. 6).

As follows from Sec. 3, the quantity γ_{OC} does not depend on V when $V < V_{cr}$. Let $\gamma_{OC} \gg \gamma_{ir}$ in weak fields. Then, for $V \ll V_{cr}$, $\Delta\omega\sqrt{\gamma_{OC}/\gamma_{ir}}$, the quantity $Q \propto V^2$; saturation sets in when $V \sim \Delta\omega\sqrt{\gamma_{OC}/\gamma_{ir}}$. However, when $V > V_{cr}$, the quantity γ_{OC} decreases exponentially with increasing V, and therefore no saturation sets in. The absorbable power begins to exponentially decrease, the decrease slowing down when $\gamma_{OC}(V) \sim \gamma_{ir}$ and $Q \sim Q_{sat} 2V_{cr}^2/\Delta\omega^2$. As V increases further, the quantity $Q \propto V^2$ and attains saturation when $V \sim \Delta\omega$. Of interest is the case $\gamma_{im} \sim \gamma_{ir}$, i.e., when $R \sim 1$ in weak fields. In this case saturation sets in only when $V \sim \Delta\omega$ (see Fig. 6). The possibility of the decrease of the absorbable power owing to the nonlinear dependence of an OC event on the light intensity was pointed out in [14], where this effect is called medium "brightening" (in contrast to the saturation effect).

6. CONCLUSION

Let us discuss some of the results of the present paper. Notice, first of all, that the above-employed approach, which is based on the consideration of the compound system, allows the formulation of a nonlinear broadening theory in closed form. Indeed, the dynamical part of the problem is given by the solution of the basic system of equations (2.7), while the kinetic effects are taken into account by the balance equations (4.4), where the relaxation parameters are determined by the OC cross sections computed in Sec. 3. We stress that in the above-developed approach the most interesting region is, in contrast to the conventional approach [6], analyzed on the basis of elementary population-balance equations. This is due to the fact that the phase relaxation, which complicates the kinetics, is automatically taken into account at the dynamical stage in the Eqs. (2.7). The solution of these latter equations is, however, a standard problem of the theory of atomic collisions. Thus, the consistent use of the compound-system concept allows us to not only generalize the problem, but to simplify it as well.

The results of the inhomogeneous broadening theory for strong fields, which is used in quantum radiophysics (see, for example, [6], Sec. 17.5), do not follow from the formula (5.3). At the same time, for weak fields, these results, which correspond to the quasi-static theory, are contained in (5.3). The indicated discrepancy is due to the fact that the criterion (4.1) used in the present paper is the converse of the assumption on which the conventional approach is implicitly based. Indeed, the condition (4.1) implies that we neglect any relaxation that occurs in the course of an OC. Meanwhile, the averaging that is carried out in the theory of inhomogeneous broadening over the static shifts (see [6], Sec. 17.5) corresponds to the assumption that the atom has in the course of a collision "time" to relax, i.e., that the converse of the condition (4.1) is fulfilled.

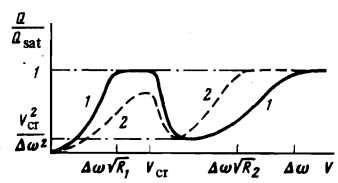

FIG. 6. Dependence of the absorbable light power Q on V in the static wing of the line (see Secs. 4 and 5). The curve 1 corresponds to the case $R(V = 0) \equiv R_1 < V_{cr}^2/\Delta\omega^2$; the curve 2, to the case $R(V = 0) \equiv R_2 > V_{cr}^2/\Delta\omega^2$.

Notice that under gas-kinetic conditions the condition (4.1) is usually fulfilled for spontaneous relaxation. In this case, as can be seen from (5.3), the parameters T_1 and T_2^{-1} enter into the result multiplicatively. In the general case, however, this may not be the case at sufficiently large values of T_1^{-1} (e.g., at high electron densities): T_2 may be nonlinearly related to T_1 (cf., for example, [17][5]).

Let us make some remarks about the experimental aspects of the problem. Here it is, in principle, possible to independently measure the three quantities relating the field characteristics to the parameters of the colliding atoms: 1) the total absorbable power Q from the attenuation of the light intensity; 2) the spontaneous-radiation intensity (4.3), by, for example, carrying out observations in the direction perpendicular to the initial direction of the beam (in the absence of electronic relaxation this corresponds to the measurement of Q_{ir}); 3) the heating of the heavy particles of the gas and, consequently, the scattering of the atoms in OC.

It is possible in all the three cases to observe nonlinear effects as the light intensity is increased. The experimental observation of these effects is of interest both in itself and in connection with the possibility of the measurement of the characteristics of the interaction of the atoms, as well as from the point of view of the direct experimental verification of the approximations used in the theory of atomic collisions.

[1)] Below we shall, for brevity, call the atomic particles X and Y simply atoms.

[2)] In the case of broadening by electrons this region is called the "single-electron" region [2].

[3)] This transformation was first carried out by Spitzer [10]. Its connection with other approaches is considered in [11].

[4)] We distinguish between the quanta produced as a result of spontaneous transitions and those of the field (2.1) (by, for example, their polarization.

[5)] Similar questions have been considered by I. Bel'gman.

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