Optical and radiative collisions

V. S. Lisitsa and S. I. Yakovlenko

V. I. Kurchatov Institute of Atomic Energy, Moscow
(Submitted August 13, 1973)
Zh. Eksp. Teor. Fiz. **66**, 1550–1559 (May 1974)

Photon absorption by atoms colliding in an external electromagnetic field $E_0\cos\omega t$ is considered. The frequency ω is close either to the characteristic transition frequency of one of the atoms (optical collisions—OC) or to the frequency difference between transitions in both atoms (radiative collisions—RC). A unified approach is given to the theory of OC and RC. The results of the theory of broadening which are known for OC are used to find the RC line profile in weak fields, E_0. This approach is not restricted to the case of a weak field and can be used to investigate new nonlinear effects in the elementary process of photon absorption in OC and RC. It is shown that, in the general case, photon absorption and collisions between atoms cannot be separated, in contrast to the usual treatment where the two effects appear in the kinetic equations separately. The latter approximation is violated for fields E_0 above a certain critical field E_{crit}. When $E_0 > E_{crit}$, the absorption of light by the medium is sharply reduced and this leads to a new "transmission enhancement" which is unrelated to saturation of the medium. It is important to note that, for OC, $E_{crit} \sim 10^4$ V/cm in the region of the line wings, which can be achieved with existing tunable lasers. Some possible experiments are discussed which should yield new information on the properties of quasimolecules.

1. CROSS SECTIONS FOR OPTICAL AND RADIATIVE COLLISIONS

We consider a collision between atoms X and Y in an external electromagnetic field $E(t) = E_0\cos\omega t$. The field frequency ω is close either to the natural transition frequency in one of the atoms (X) or to the difference between transition frequencies in both atoms XY (Fig. 1). In the former case, we have the well known optical collisions (OC)[1)] leading to the broadening of the spectral lines[1] and, in the latter, we have radiative collisions (RC) which involve the transfer of excitation from one atom to the other with the simultaneous absorption (emission) of a photon.[2] We shall consider both OC and RC on the basis of a unified approach which may be summarized as follows:

1) OC and RC are looked upon as photon absorption reactions of the form

$$X(2)+Y(1)\overset{\text{OC}}{\rightleftharpoons}X(1)+Y(1)+\hbar\omega,$$
$$X(2)+Y(1)\overset{\text{RC}}{\rightleftharpoons}X(1)+Y(2)+\hbar\omega,\quad \omega\approx\omega_0. \tag{1.1}$$

In these expressions 1, 2 are the ground and an excited state of X or Y and $\hbar\omega_0$ is the energy difference between the transitions executed by the atoms on collision (Fig. 1).

2) The efficiency of the reactions (1.1) is characterized (by analogy with[2]) by the cross sections for optical and radiative collisions (denoted by σ). For classical motion of the nuclei, these cross sections are defined by

$$\sigma(\Delta\omega, E_0, v) = 2\pi\int d\rho\,\rho w(\Delta\omega, E_0, \rho, v). \tag{1.2}$$

3) The probability w is determined from the Schrödinger equation for the amplitudes a_1 and a_2 of the initial and final states of the resultant Hamiltonian for the compound system "atom X + atom Y + electromagnetic field":

$$i\dot{a}_1 = V_1a_1 + Ve^{i\Delta\omega t}a_2,\quad i\dot{a}_2 = V_2a_2 + Ve^{-i\Delta\omega t}a_1,$$
$$a_1(-\infty)=1,\quad a_2(-\infty)=0. \tag{1.3}$$

In these expressions, V_1 and V_2 are the diagonal matrix elements for the excitation of atoms X, Y (allowance for E_0 in these elements would merely lead to a shift of the line center[2]), V is the nondiagonal matrix element

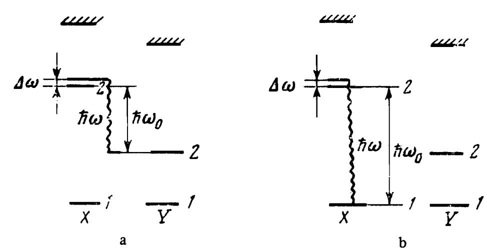

FIG. 1. Atomic terms in the case of radiative collisions (a) and optical collisions (b).

containing E_0, and $\Delta\omega = \omega - \omega_0$ is the detuning from resonance. Moreover $w = |a_2(\infty)|^2$.

We must now note several points which are relevant to this approach.

The equations given by (1.3) have a form which is typical for the two-level approximation in the theory of atomic collisions. However, the concept of a transition is now used for the entire compound system (atoms + field). This means that when the solution of (1.3) is investigated we shall, for example, interpret the "terms" as being not the terms of the quasimolecule XY but the levels of the compound system as a whole, including the energy of the photon $\hbar\omega$. This approach ensures right from the outset that the collisions between the atoms and the absorption of light are part of the same process and cannot be separated.

The OC and RC cross sections given by (1.2) are not the usual cross sections in the sense that they depend not only on the parameters of the colliding atoms but also on the characteristics of the external field, namely, ω and E_0. The dependence of the efficiency of the reactions (1.2) on $\Delta\omega$ determines the line profile in OC or RC. The OC line profile is well known from broadening theory[1] but only for very weak fields E_0. For RC, on the other hand, the dependence on E_0 has been examined but only for the center of the line, $\Delta\omega = 0$.[2] In this paper we shall investigate the dependence of the OC and RC reactions on both $\Delta\omega$ and E_0.

In the case of weak fields (Sec. 2), the cross section given by (1.2) is a linear function of intensity: $\sigma \propto E_0^2$.

In this connection we are mainly interested in the determination of the RC line profile.

For large E_0 (Sec. 3), the dependence of σ on E_0^2 may be nonlinear. This is particularly interesting for the optical collisions which have frequently been considered. The nonlinear effects lead to a new feature in the kinetics of light absorption by a medium, namely, transmission by the medium which is not connected with its saturation (Sec. 4).

The matrix elements in (1.3) for rectilinear trajectories of the atoms $R^2(t) = \rho^2 + v^2t^2$ are given by

$$V_1-V_2=\varkappa=C_nR^{-n}(t),\quad V_{OC}=DE_0,\quad V_{RC}=B_3E_0R^{-3}(t). \tag{1.4}$$

In these expressions C_n, B_3 are constants which depend on the characteristics of the interacting atoms (see[1,2]), and D is a constant which characterizes the transition in atom X.

The nondiagonal matrix elements are different for OC and RC, and this reflects the difference between the reactions in (1.1). In the case of RC, there is a change in the state of both atoms, and the transition is in general impossible in the absence of interaction between them; in the case of the OC reaction, on the other hand, emission is possible even in the absence of the atom Y. In the latter case this occurs only at the frequency $\omega \approx \omega_0$. Henceforth we eliminate from our analysis the narrow region $\omega \approx \omega_0(\Delta\omega \approx 0)$ in the case of OC[2)].

2. WEAK FIELDS. RC LINE PROFILE

We begin by considering (1.3) in the case of weak fields (the relevant criterion will be established in Sec. 3). In this case, the solution can be obtained by perturbation theory.

For RC we have

$$w_{RC}=\left|\int_{-\infty}^{\infty} V_{RC}(t)\exp\left\{i\left[\Delta\omega t-\int_0^t \varkappa(\tau)d\tau\right]\right\}dt\right|^2. \tag{2.1}$$

For OC, it follows from (2.1) that for $\Delta\omega \gg \gamma$ we can exclude terms corresponding to the absorption of the field by atom X in the absence of interaction with atom Y which is unimportant for the ensuing analysis. This can be done by integrating (2.1) by parts. The first term is then responsible for absorption in the absence of absorption (and, consequently, vanishes for $\Delta\omega \neq 0$), and the second term represents absorption connected with the interaction between the atoms:[3)]

$$w_{OC}=\frac{V_{OC}^2}{\Delta\omega^2}\left|\int_{-\infty}^{\infty}\varkappa(t)\exp\left\{i\left[\Delta\omega t-\int_0^t\varkappa(\tau)d\tau\right]\right\}dt\right|^2. \tag{2.2}$$

It is clear from (2.1) and (2.2) that the dependence of w on E_0 for weak fields is trivial, i.e., $w \propto E_0^2$. The interesting aspect, therefore, is the evaluation of the dependence of σ on $\Delta\omega$, i.e., the OC and RC line profiles. Since the OC line profile is well known,[1] we shall confine our attention to the RC case.

The RC line profile is analogous to the OC profile[1] in that it can be divided into two regions, i.e., the static and the impact regions, corresponding to large and small values of $\Delta\omega$, respectively. The results for small $\Delta\omega$ were obtained in[2]; here we shall confine our attention to the case of large $\Delta\omega$. In this limit, the final result is determined by the relative signs of $\Delta\omega$ and κ, which determine the presence or otherwise of a stationary phase point $\kappa(t_0) = \Delta\omega$ in the exponential function in (2.1). In the first case, if we evaluate the integral in (2.1) by the method of steepest descents on the real axis, we have

$$w_{RC}^{(-)}=4\pi V_{RC}^2(t_0)/|\dot\varkappa(t_0)|,\quad \dot\varkappa\equiv d\varkappa/dt. \tag{2.3}$$

This result is identical with the transition probability for atomic collisions in the presence of term crossing (this is the Landau case,[6] Section 90). Hence, in accordance with Section 1, the stationary-phase condition $\kappa(t_0) = \Delta\omega$ can be looked upon as the condition for term crossing in the compound system (atoms X + Y + field).

In the second case (in the absence of a stationary phase point or crossing point), the integral in (2.1) is determined by the singularity in the potential V(t) which is nearest to the real axis. This is $t^* = i\rho/v$ which gives (see[5])[4)]

$$w_{RC}^{(+)}\propto\exp\left\{-\frac{2\rho}{v}[|\Delta\omega|+|C_n\rho^{-n}|]\right\}. \tag{2.4}$$

Using (2.1), (1.4), and (1.2), we can now write down the general expression for σ_{RC}:

$$\sigma_{RC}^{(\pm)}=8\pi\frac{B_3^2}{\rho W^2v^2}E_0^2\chi^{(\pm)}\left(\frac{\Delta\omega}{\Omega}\right), \tag{2.5}$$

$$\chi^{(\pm)}(\beta)=\int_0^\infty\frac{dy}{y^3}\left|\int_{-\infty}^{\infty}\frac{dx}{(1+x^2)^{3/2}}\times\exp\left\{i\left[\beta yx\pm y^{-n+1}\int_0^x\frac{dz}{(1+z^2)^{n/2}}\right]\right\}\right|^2. \tag{2.6}$$

The signs ± correspond to equal and opposite signs of $\Delta\omega$ and κ, respectively, $\rho_W = (C_n/v)^{1/(n-1)}$ is the Weisskopf radius,[1] and $\Omega = v/\rho_W$ is the characteristic frequency scale.

The regions $\Delta\omega \ll \Omega$ and $\Delta\omega \gg \Omega$ will be referred to as the impact and static regions by analogy with the theory of broadening. For the static region, we have using (2.3) and (2.4)

$$\sigma_{RC}^{(-)}=8\pi^2\frac{B_3^2}{nC_n}\frac{\rho_{\Delta\omega}^{n-3}}{v}E_0^2,\quad \rho_{\Delta\omega}=\left(\frac{C_n}{\Delta\omega}\right)^{1/n}, \tag{2.7}$$

$$\sigma_{RC}^{(+)}\propto\exp[-2\alpha_n\rho_{\Delta\omega}|\Delta\omega|/v],\quad \alpha_n\sim1. \tag{2.8}$$

For the impact region $\sigma_{RC}^{(-)} \approx \sigma_{RC}^{(+)}$.

Therefore, the RC line profile is asymmetric (just as in the case of OC[5]): there is a gradual fall on one wing and an exponential fall on the other. We note that the characteristic impact parameter ρ_{eff} for the static wing is $\rho_{\Delta\omega}$ (2.7), whilst for the impact case $\rho_{eff} = \rho_W$.

3. STRONG FIELDS. NONLINEAR DEPENDENCE OF CROSS SECTIONS ON LIGHT INTENSITY

We now consider the case of a sufficiently strong field E_0 whose interaction with the atoms is not small in comparison with the interatomic interaction (as, for example, in the case of a collision in the field of strong laser radiation). Equations (1.3) cannot be solved by perturbation theory in this case. The general solution of (1.3), on the other hand, is not available, and we shall therefore consider some interesting special cases. This will yield some important nonlinear effects for both OC and RC.

We begin with the case where we can retain only the terms containing the field E_0 and reject V_1 and V_2 in (1.3). This case is interesting for RC.[5)] In the case of exact resonance ($\Delta\omega = 0$) we obtain a system analogous to the equations for the resonance excitation transfer,[8] the solution of which is[2]

$$w_{RC}=\sin^2(2B_3E_0/v\rho^2), \quad \sigma_{RC}=\pi^2B_3E_0/v. \qquad (3.1)$$

It is clear from (3.1) that in the impact region ($\Delta\omega \to 0$) the dependence of σ_{RC} on E_0 for strong fields is different, i.e., $\sigma_{RC} \propto E_0$ instead of $\sigma_{RC} \propto E_0^2$ [see (2.5)]. Equating (3.1) and (2.5), we obtain the critical field E_{crit} for which this change takes place:

$$E_{crit} = v\rho_W^2/\pi B_3 = C_n^{2/(n-1)} v^{(n-3)/(n-1)}/B_3\pi \equiv E_W. \qquad (3.2)$$

We note that the characteristic impact parameter ρ_{eff} for a strong field is $\rho_{eff} \approx \pi(B_3E_0/v)^{1/2} \gg \rho_W$. The condition $E_0 \gg E_W$ is equivalent to the condition that V_1, V_2 are small in comparison with V [used in the derivation of (3.1)].

The expression for E_W for $n > 3$ contains the small parameter $v^{(n-3)/(n-1)}$ and, therefore, the field E_0 remains much less than the atomic field $E_{at} = 0.5 \times 10^{10}$ V/cm and may not be small in comparison with E_W.

In the case of strong fields, the cross section $\sigma_{RC}(\Delta\omega)$ is very sensitive to the detuning $\Delta\omega$. This can be verified for the above case of $\kappa = 0$ by writing down the expression for $\sigma_{RC}(\Delta\omega)$ in accordance with the result given in [8]

$$\sigma_{RC} \propto \exp[-(E_0/E_{crit})^{1/2}], \quad E_0 \gg E_{crit}, \qquad (3.3)$$

where

$$E_{crit} = v^2/B_3\Delta\omega^2 = E_W(\Omega/\Delta\omega)^2 \equiv E_S. \qquad (3.4)$$

Comparison of (3.2) and (3.4) shows that the transition to the exponential fall in the cross section occurs for $\Delta\omega \gtrsim \Omega$.

We now consider another special case of the solution of (1.3) which determines the change in the static profile of the OC and RC lines in strong fields. It is connected with the presence of a crossing point and, for weak fields, is described by the Landau formula (2.3). Generalization of this case to arbitrary E_0 can be obtained by replacing the Landau result (2.3) by the more general result obtained by Zener [6]

$$w=2e^{-p}(1-e^{-p}), \quad p=2\pi V^2(t_0)/|\dot{\varkappa}(t_0)|. \qquad (3.5)$$

Substituting (3.5) in (1.2), we obtain

$$\sigma=2\pi\rho_{\Delta\omega}^2\Lambda(E_0^2/E_{crit}^2), \qquad (3.6)$$

$$\Lambda(\alpha)=\frac{1}{2}\int_0^1 dy\exp(-\alpha/\sqrt{1-y})[1-\exp(-\alpha/\sqrt{1-y})]$$

$$=\begin{cases} \alpha & \text{when } \alpha\ll1 \\ \alpha^{-1}e^{-\alpha} & \text{when } \alpha\gg1 \end{cases}, \qquad (3.7)$$

when E_{crit} is the critical value of the field which has the following form for OC and RC:

$$E_{crit}^{RC}=\left(\frac{nC_n}{2\pi}\frac{v}{\rho_{\Delta\omega}^{n-5}}\right)^{1/2}\frac{1}{B_3}=\left[\frac{nC_n^{5/n}}{2\pi}(\Delta\omega)^{(n-5)/n}v\right]^{1/2}\frac{1}{B_3}\equiv E_{\Delta\omega}^{RC}, \qquad (3.8)$$

$$E_{crit}^{OC}=\left(\frac{nC_n}{2\pi}\frac{v}{\rho_{\Delta\omega}^{n+1}}\right)^{1/2}\frac{1}{D}=\left[\frac{n(\Delta\omega)^{(n+1)/n}v}{2\pi C_n^{1/n}}\right]^{1/2}\frac{1}{D}\equiv E_{\Delta\omega}^{OC}. \qquad (3.9)$$

When $E_0 \ll E_{crit}$ the expression given by (3.6) coincides with (2.7) in the case of RC, and with the well known result of the quasi-static theory of broadening in the case of OC. [1]

When $E_0 \gg E_{crit}$ the cross section (3.6) falls rapidly

$$\sigma=4\pi\rho_{\Delta\omega}^2(E_{crit}/E_0)^2\exp[-(E_0/E_{crit})^2]. \qquad (3.10)$$

This rapid fall is connected with term repulsion, as in the usual theory of collisions. In the present case, this repulsion is due to the "external" factor, i.e., the field E_0.

Therefore, in strong fields there is a radical change in the static OC and RC profiles. This must be specially noted for OC for which the static profile has frequently been investigated. [1] However, this was done in weak fields E_0, in contrast to (3.10).

From (3.1), (3.3), and (3.10) it follows that, beginning with a certain critical value $E_0 \sim E_{crit}$, the cross sections σ_{OC} and σ_{RC} deviate from the usual E_0^2 law which is valid for weak fields. Moreover, for all the cases which have been considered, there is an increase in the transparency of the medium for $E_0 > E_{crit}$ (transmission effect). This effect is particularly clearly defined for the case of term crossing, (3.10).

The above nonlinear effect is of direct interest because it can be seen even in fields much lower than the characteristic atomic field $E_{at} = 0.5 \times 10^{10}$ V/cm. Let us now estimate the order of magnitude for the characteristic parameters utilized above. Thus, for n = 6, $v = 10^5$ cm/sec, and assuming that $C_n \sim B_n \sim 10$ at. units, we obtain $\rho_W \sim 10a_0 = 10^{-7}$ cm, $\Omega \sim 10^{12}$ sec^{-1} ≈ 0.001 eV, $E_W = 10^7$ V/cm, $E_{\Delta\omega}^{RC} \sim 10^7 (\Delta\omega/\Omega)^{1/2}$ V/cm, and $E_{\Delta\omega}^{OC} \approx 0.5 \times 10^4$ V/cm.

Fields $E_0 \sim 10^7$ V/cm can be obtained by focusing high-power laser beams and, therefore, the above effects may become significant, for example, in the case of laser-induced breakdown in a medium. The possibility of being able to investigate line profiles broadened by optical collisions, using a tunable laser beam, is also very interesting. Variable-frequency laser beams with power per pulse ~1 MW are now available. [9] By focusing this beam into a spot of radius 10^{-2} cm it is possible to achieve $E_0 \approx 10^4$ V/cm.

4. EFFECT OF NONLINEARITY IN THE ELEMENTARY EVENTS ON THE KINETICS OF LIGHT ABSORPTION BY A MEDIUM

The main aim of the foregoing discussion was to analyze the dynamic problem, i.e., to solve the Schrödinger equation (1.3) and hence determine the cross section (1.2). As we have seen, even at this stage, one encounters nonlinear effects, the significance of which is that the absorption of a photon and the broadening collision cannot be separated. We then have the natural question: what is the effect of this on the kinetics of the population of atomic levels and the absorption of light? The nonlinearities which arise during this ("kinetic") stage are known from the usual (simplified) theory in which the collision and absorption events appear in the kinetic equation separately ([10], page 126). Departure from the framework of this approximation, i.e., simultaneous allowance for the nonlinearities of both types, naturally leads to a substantial complication of the theory. In this paper we shall consider this problem within the framework of a kinetic model which is relatively simple but at the same time enables us to exhibit the interrelation between the two nonlinearities and, in particular, between the known saturation effect and the transmission effect established above (Section 3).

When we consider the effect of the reactions (1.1) on the kinetics of population of atomic levels, it is sufficient to introduce into the population balance equation terms which are proportional to the transition rate:

$$q = \langle \sigma(\Delta\omega, E_0, v) v \rangle \tag{4.1}$$

where the angle brackets represent averaging over the relative velocities. The change in the light-wave intensity is taken into account through the expression for the power absorbed (emitted) per unit volume:

$$Q_{RC} = \hbar\omega_0 q_{RC} (N_1^X N_2^Y - N_2^X N_1^Y),$$
$$Q_{OC} = \hbar\omega_0 q_{OC} [N_1^Y (N_1^X - N_2^X)], \tag{4.2}$$

where $N_{1,2}^{X,Y}$ are the populations of states 1, 2 of atoms X, Y.

The next problem in the kinetics of the process reduces to the determination of the equilibrium populations $N_{1,2}^{X,Y}$. In the usual approach, the collision cross sections of atoms X and Y enter the kinetic equations in the form of relaxation constants which are independent of the field E_0. This approach does not take into account the possibility of absorption of light in the course of the elementary collision event itself.[6)] Taking this into account is equivalent to the appearance of a new relaxation channel in which the absorbed energy excess ($\hbar\Delta\omega$) is converted into translational degrees of freedom of the atoms X and Y.

To be specific, let us consider OC. Suppose that a uniform gas of two-level atoms X is broadened by atoms Y and undergoes inelastic (for example, radiative) relaxation characterized by rates γ_{12} and γ_{21} of the transitions $X(2) \rightleftarrows X(1)$. The population-balance equations for this case have the form

$$dN_1^X/dt = 0 = (\gamma_{12} + qN_Y) N_2^X - (\gamma_{21} + qN_Y) N_1^X,$$
$$N_1^X + N_2^X = \text{const}. \tag{4.3}$$

If we now introduce the inelastic relaxation time through the formula $T_1^{-1} = (\gamma_{12} + \gamma_{21})/2$, and consider the number of active molecules per unit volume $N = N_1^X - N_2^X$, we find that $N = T_1^{-1} N_0/(T_1^{-1} + qN_Y)$ where N_0 is the value of N for $E_0 = 0$. The power Q absorbed per unit volume is then given by

$$Q = \hbar\omega \frac{T_1^{-1} q N_Y}{T_1^{-1} + qN_Y} N_0. \tag{4.4}$$

Let us begin by considering the impact limit $\Delta\omega \ll \Omega$. In this case, the quantity qN_Y can readily be related to the usual impact width γ which is defined in terms of the phase relaxation time by the formula $T_2^{-1} = \gamma$. In fact, using (1.2), (1.4), (2.2), and (4.1) we obtain $qN_Y = (DE_0/\Delta\omega)^2 T_2^{-1}$, and (4.4) becomes identical with the well known expression for the absorbed power ([10], p. 367). We note that our result does not contain the term T_2^{-2} which should be additive to $\Delta\omega^2$. This is natural because in the derivation of (2.2) it was assumed that $\Delta\omega \gg T_2^{-1}$. Therefore, the adopted scheme (4.3) takes into account both the inelastic (T_1) and elastic (T_2) relaxation, where the latter multiplies the intensity E_0^2 in the impact limit.

The fact that E_0^2 and T_2^{-1} multiply one another corresponds to the approximate mutual separation of the photon-absorption and atomic-collision events. This, however, is valid only in sufficiently weak fields. In the general case, on the other hand, the collision and absorption events cannot be separated, as already noted. We shall demonstrate this by considering the example of the inhomogeneous broadening region $\Delta\omega \gg \Omega$.

Since for $\Delta\omega \gg \Omega$ the atomic collisions have a quasi-static character, they do not lead to the dephasing of the oscillations, which is well known from the theory of broadening.[1] Other causes of phase relaxation will be assumed to be absent. In this formulation, the problem which we are considering differs from the well known case of absorption in inhomogeneously broadened lines[10] (Sec. 17) where phase relaxation is supposed to be present even against a background of stationary atoms ($T_2^{-1} \neq 0$).

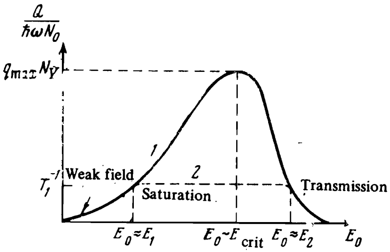

FIG. 2. Absorbed power Q as a function of the field E_0 for two values of the density N_Y of the broadening particles: (1) $N_Y \ll N_{crit}$, (2) $N_Y \gg N_{crit}$.

Analysis of the result given by (4.4) for the static region reduces to the following.

When $qN_Y \ll T_1^{-1}$ we have

$$Q = \hbar\omega q N_Y N_0, \tag{4.5}$$

i.e., the absorption line shape is determined by the function $q(\Delta\omega, E_0)$.

When $qN_Y \gg T_1^{-1}$ we reach saturation of the medium, i.e., Q is independent of the field E_0 (see [10], p. 367), so that

$$Q = \hbar\omega N_0 / T_1. \tag{4.6}$$

The fact that the absorption and collision events cannot be separated is most clearly seen in the following important fact: if the nonlinearity of the elementary event is ignored, then $q \propto E_0^2$ so that the condition $qN_Y \geq T_1^{-1}$ (and hence the saturation effect itself) can always be realized; in reality, on the other hand, the function q is bounded $[q \lesssim q_{max} \sim q(E_{crit})]$ and, therefore, in the region $N_Y \ll N_{crit}$ where $N_{crit} q_{max} \approx T_1^{-1}$ there is in general no saturation (we recall that we are dealing with the line wing). We now estimate the order of magnitude of N_{crit}: When $T_1 \sim 10^8$ sec^{-1}, $v \sim 10^5$ cm/sec, $\rho_{\Delta\omega} \sim 10^{-7}$, we have $N_{crit} \sim 10^{17}$ cm^{-3}.

The behavior of Q as a function of E_0 is qualitatively illustrated in Fig. 2. As can be seen, when $N_Y \gg N_{crit}$ the straight line T_1^{-1} = const intersects the $q(E_0)$ curve at two points: at $E_0 \sim E_1$ and at $E_0 \sim E_2$ where

$$E_1^2 = E_{crit}^2 \frac{N_{crit}}{N_Y} = \frac{\Delta\omega D^{-2}}{N_Y \rho_{\Delta\omega}^3 T_1}, \quad E_2^2 = E_{crit}^2 \ln\left[\left(\frac{E_2}{E_{кр}}\right)^2 \frac{1}{N_Y v \rho_{\Delta\omega}^2 T_1}\right]. \tag{4.7}$$

Therefore, when $E_0 \sim E_1$ we have saturation, but for still larger fields $E_0 \sim E_2 \sim E_{crit}$ the medium becomes transparent.

It is clear from the foregoing that nonlinear absorption in the collision event itself becomes appreciable in the line wing where the oscillation dephasing during the X—Y collision is unimportant, and there are no other phase-relaxation mechanisms ($T_2^{-1} = 0$). In the usual inhomogeneous broadening,[10] the results depend on T_2 and do not explicitly contain the velocity v of the atoms. In our analysis, on the other hand, the parameter T_2 is absent but v is present. In weak fields, the results of both analyses are the same because, in this limit, they do not contain either T_2 or v. This agreement is connected with the following fact, which is well known in broadening theory: in the static region, averaging over the impact parameters is equivalent to averaging over term shifts by the nearest neighbor. However, this is valid only for sufficiently weak fields E_0. When the fields are so strong that they affect the population kinetics ($E_0 \gtrsim E_1$) or the probability of the elementary absorption event itself ($E_0 \gtrsim E_{crit}$) then the line shape is no longer determined only by the level shift. Thus, in this case, the results of the theory of inhomogeneous broadening are fundamentally dependent on T_2 (and vanish for $T_2^{-1} \to 0$; see[10]); the results obtained in the present work, in their turn, are fundamentally dependent on v (and, correspondingly, the effect vanishes for $v \to 0$). Hence it is clear that the effect which we are considering is essentially different in sufficiently strong fields from the usual saturation in an inhomogeneous broadened line. The essence of the effect is that, in strong fields, the collision and absorption events can no longer be separated, as already noted.

In conclusion, let us consider some experimental aspects. First of all, the connection between OC and RC enables us to use the same experimental procedures for both of them. For RC this means that when the line profile is examined we can use the usual method of spectral analysis. For OC this means that we can have an essentially new type of experiment based on the use of a tunable laser, as suggested previously for RC.[2] The scheme of the experiment is as follows: the gas is irradiated with a laser pulse of frequency $\omega \approx \omega_0$ and, as a result of the reaction (1.1), the state X(2) is populated. The integrated intensity of the decay line (for example, to some intermediate state) can be used to determine the relative efficiency of population of the state X(2) as a function of the laser-frequency detuning $\Delta\omega$. By varying the laser frequency from pulse to pulse, it is possible to examine the entire line profile. In this way, we can measure not only the line profile for weak fields but also (by increasing E_0) the critical value of the field, E_{crit}. At the present time the tunable laser power is not high enough and measurements of E_{crit} are possible only for OC for which $E_{crit} \sim 10^4$ V/cm.

The above transmission effect is interesting in itself and also from the standpoint of the determination of the interaction potential C_nR^{-n}. In fact, if we know the line profile in a weak field, and the value of E_{crit}, we can use (3.6) and (3.8) to find n and C_n. We note that the transmission effect appears at relatively low gas densities and low fields E_0 for which the laser breakdown is usually impossible.

The authors are grateful to L. I. Gudzenko, V. I. Kogan, and O. B. Firsov for valuable discussions.

1)This term was introduced by Weisskopf.

2)The width of this region is of the order of the impact linewidth γ, see [1].

3)These transformations were first carried out in [3] in the case of hydrogen-line broadening. Similar results were obtained by the Green function formalism [4] (the relation between those two methods is discussed in [5]).

4)A similar evaluation of the integral in the case of broadening is given in [7].

5)The case $v_1 = v_2 = 0$ for OC corresponds to the interaction between the resonance field and the atom in the absence of collisions, and will not be considered here.

6)In this sense, the processes considered in this paper are in many ways analogous to the processes considered in [11] where the transition cross sections depend on the rate of radiative decay.

1 R. Breene, Rev. Mod. Phys. **29**, 94 (1957). I. I. Sobel'man, Vvedenie v teoriyu atomnykh spektrov (Introduction to the Theory of Atomic Spectra), Fizmatgiz, 1963. M. Baranger, Atomic and Molecular Processes (ed. D. Bates) (Russ. Transl., Mir, 1963), p. 429.

2 L. I. Gudzenko and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. **62**, 1686 (1972) [Sov. Phys.-JETP **35**, 877 (1972)]. S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. **64**, 2020 (1973) [Sov. Phys.-JETP **37**, 1019 (1973)].

3 L. Spitzer, Phys. Rev. **55**, 699 (1939); **56**, 39 (1939); **58**, 348 (1940).

4 V. V. Yakimets, Zh. Eksp. Teor. Fiz. **51**, 1469 (1966) [Sov. Phys.-JETP **24**, 990 (1967)].

5 V. I. Kogan and V. S. Lisitsa, JQSRT **12**, 881 (1972).

6 L. D. Landau and E. M. Lifshitz, Kvantovaya Mekhanika (Quantum Mechanics), Fizmatgiz, 1963 [Addison-Wesley, 1965].

7 S. D. Tvorogov and V. V. Fomin, Opt. Spektrosk. **33**, 413 (1971).

8 N. F. Mott and H. S. W. Massey, Theory of Atomic Collisions, 1949 (Russ. Transl., Mir, 1969).

9 A. I. Kovrigin and P. V. Nikles, ZhETF Pis'ma Red. **13**, 440 (1971) [JETP Lett. **13**, 313 (1971)].

10 V. M. Faĭn, Kvantovaya radiofizika (Quantum Radiophysics), Vol. 1, Sov. Radio, 1972.

11 V. I. Kogan, V. S. Lisitsa, and A. D. Selidovkin, Zh. Eksp. Teor. Fiz. **65**, 152 (1973) [Sov. Phys.-JETP **38**, 75 (1974)].

Translated by S. Chomet
159