Nonlinear quantum theory of optical collisions and spectral-line contours

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A quantum-kinetic equation is derived for the density matrix using a new collision integral that characterizes the nonlocal character of the interaction and the effect of a strong electromagnetic field on the collision dynamics. A new method is developed on the basis of this equation for the analytic investigation of optical collisions, nonlinear dynamic effects, and the contours of atomic spectral lines resulting from collisions with multilevel particles of the impurity gas. The new method differs in principle from earlier ones in that it combines the dynamic and kinetic problems of the theory of atomic collisions. The following physical problems are solved by the proposed method: 1) a universal formula for the spectral-line contours is obtained, which is valid for the entire spectrum, including the shock, intermediate, and quasistatic frequency ranges in an electromagnetic field of arbitrary strength; 2) formulas are obtained for the optical-collision broadening and line-contour shifts in an electromagnetic field of arbitrary strength and frequency; 3) the absorption and emission of energy by the electromagnetic field is taken into account automatically, not only in the case of optical collisions, but also in the case of atoms in free flight; 4) the dependence of the spectral-line contour shift on the strength of the electromagnetic field is determined; and 5) the problem of the effect of a strong electromagnetic field on the dynamics of optical-collision transitions (the dynamic problem) is formulated and solved. It is shown that in special cases the universal formula obtained for the spectral-line contours yields generalized versions of the well-known Karplus-Schwinger and Lisitsa-Yakovlenko formulas and others.

1. INTRODUCTION

In an earlier paper,[1] the author and Rautian pointed out a limitation of the standard theory of spectral line broadening by a weak electromagnetic field that does not affect the dynamics of optical collisions (OC). To remove these limitations we subsequently[2] obtained quantum kinetic equations for the density matrix, in which the collision integrals are expressed in terms of the generalized Moller operator $\Omega(t)$ and the $\mathcal{T}(t)$ matrix, which depend on the parameters of the electromagnetic field and the time t. As a result we predicted a new physical effect—"field" narrowing of spectral lines associated with the influence of a strong electromagnetic field on the collision dynamics (nonlinear dynamical effects) and investigated it analytically.[2]

The dynamics of an individual OC event in a strong electromagnetic field was investigated by Lisitsa and Yakovlenko,[3] using differential equations for the probability amplitudes (in a basis of "field dressed" states[3,5]). Their results confirmed the field narrowing effect[2] and made it possible to predict the "brightening" of the medium,[3] which is a direct manifestation of field narrowing in the quasistatic region of the spectrum. These effects have recently obtained reliable experimental confirmation.[6,7]

Much attention has been given in recent years to the study of nonlinear dynamical effects. This is due, on the one hand, to new experimental possibilities resulting from the development of laser techniques, and on the other hand, to new prospects for obtaining additional information on the characteristics of the collisional interaction of particles, and on the selective initiation of new processes that do not take place in the absence of a strong electromagnetic field. As examples we may adduce radiative collisions[4] and laser-induced excitation exchange (see, e.g., Refs. 5 and 8).

A characteristic feature of the known methods for investigating the OC broadening of spectral lines (see, e.g., Refs. 3, 4, and 10) is the separation of the problem into two independent problems: the dynamical problem, and the kinetic problem. The solution of the dynamical problem yields the probability (cross section) for an OC transition, which may then be used to determine the populations of the energy levels and the shape of the "field saturated" spectral line.[3,4] In this case, because of the separation of the problem into two parts, one cannot obtain a general analytic expression for the line broadening and shift that would suitably describe the entire line contour. Moreover, the dynamical problems for the impact and quasistatic regions of the spectrum are solved by different methods, and it is not possible to "match" the results; the intermediate frequency region is therefore not discussed.

It accordingly seems natural to resort to the quantum kinetic equation for a single-particle density matrix when investigating the problem of optical collisions in a strong electromagnetic field, for in that method the dynamic and kinetic problems turn out to be automatically unified. Such a method was first developed in Ref. 2, but the Markovian collision integral obtained there does not provide an adequate description of the wings of the spectral lines. It therefore became necessary to obtain a new collision integral that would contain detailed information on the "nonlocality" of the collisions[9] and the effect of the electromagnetic field on their dynamics.

In this paper we develop a nonlinear quantum-kinetic

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theory of optical collisions and spectral-line contours resulting from collisions of active particles with impurities in a strong electromagnetic field. In Section 2 we obtain a collision integral of non-Markovian type in the binary approximation, which makes it possible to describe the entire contour, including the impact, intermediate, and quasistatic frequency regions. Its distinguishing feature is the presence of a new evolution operator $L(t,t')$ that characterizes the collision dynamics in a strong electromagnetic field. In Section 3 we derive a kinetic equation for the density matrix and use it to formulate the problem of investigating the spectral-line shapes in the two-level resonance approximation. In Section 4 we determine the dynamical evolution operator $L(t,t_0)$ in a strong electromagnetic field (the dynamical problem). In Section 5 we obtain for the first time a universal formula for the line shapes that describes the entire spectrum, including the shock, intermediate, and quasistatic frequency regions in the case of an arbitrarily strong electromagnetic field. The application of this formula to special cases yields generalizations of the well-known Lorentz-Weisskopf, Karplus-Schwinger,[11] Lisitsa-Yakovlenko,[3] Rautian,[12] and Vdovin-Galitskiĭ-Yakimets[13] formulas. In Section 6 we analyze some special cases, and in Section 7 we investigate a new physical effect: the dependence of the shift of the spectral-line contours on the strength of the electromagnetic field. The published papers most closely related to the subjects discussed here are Refs. 3–5 and 15–18.

2. THE COLLISION INTEGRAL

Let us consider a two-component gaseous mixture (particles a and b) at low pressures and derive the collision integral $\sigma(t)$ as a result of their interactions with the impurity particles (b). We shall start from the chain of Bogolyubov equations broken off at the two-particle density matrix $F_{ab}(t)$ (the binary approximation)[2,14]:

$$i\hbar\frac{\partial F_{ab}}{\partial t}=[H_a+H_b+V(t)+W, F_{ab}(t)],$$
$$i\hbar\frac{\partial F_a}{\partial t}=[H_a+V(t), F_a(t)]+i\hbar\sigma(t), \tag{2.1}$$

$$\sigma(t)=\frac{n_b}{i\hbar}\mathrm{Sp}_b[W, F_{ab}(t)]. \tag{2.2}$$

Here the $F_{a,b}(t)$ are single-particle density matrices, H_a and H_b are the Hamiltonians for the isolated particles, $V(t)$ and W are the energy operators for the interactions of particles a with the field and with the particles b, respectively, and n_b is the concentration of the impurity particles. We express the two-particle matrix $F_{ab}(t)$ as the sum of the correlation matrix $g(t)$ and the product of the single-particle matrices[1]

$$F_{ab}(t)=F_a(t)F_b(t)+g(t). \tag{2.3}$$

From the differential equation (2.1) for F_{ab} and Eq. (2.3) we obtain the following equation for the correlation matrix $g(t)$:

$$i\hbar\frac{\partial g}{\partial t}=[H_a+H_b+V(t)+W, g(t)]+[W, F_aF_b]. \tag{2.4}$$

Analysis showed that the differential equation (2.4) can be solved with the aid of the evolution operator $G(t,t_0)$, which has the form

$$G(t, t_0)=L(t, t_0)S(t, t_0)F(t, t_0). \tag{2.5}$$

The operators F, L, and S satisfy the differential equations

$$\frac{\partial F}{\partial t}=\frac{i}{\hbar}(H_a+H_b)F, \qquad \frac{\partial S}{\partial t}=\frac{i}{\hbar}S[FV(t)F^+], \tag{2.6}$$

$$\frac{\partial L}{\partial t}=\frac{i}{\hbar}L[YFWF^+Y^+], \qquad L(t,t)=I, \tag{2.7}$$

in which I is the operator for the identity transformation (the unit matrix). The unitary-transformation matrix $Y(t)$ satisfies the same differential equation as $S(t,t_0)=Y(t)Y^+(t_0)$, so we can use the operator $S(t,0)$ in place of $Y(t)$ in the equation (2.7) for L.

The evolution operators G, S, and L have the standard transformation properties:

$$L(t, t_1)L(t_1, t_0)=L(t, t_0), \quad L(t, t_0)=L^+(t_0, t).$$

The operator $F(t,t_0)$ transforms all the operators to the interaction representation. The operator $S(t,t_0)$ characterizes the evolution of the correlation matrix and other operators under the action of the electromagnetic field. The evolution operator $L(t,t_0)$ represents the collisional interaction in a strong electromagnetic field; we shall call it the dynamic-evolution operator.

Using the operator $G(t,t_0)$, we obtain the following formula for the correlation matrix $g(t)$ from Eq. (2.4):

$$g(t)=G^+(t,t_0)g(t_0)G(t,t_0)$$
$$-\frac{i}{\hbar}\int_{t_0}^{t}G^+(t,t')[W, F_a(t')F_b(t')]G(t,t')\,dt'. \tag{2.8}$$

On substituting (2.8) into (2.3) and (2.2) we obtain the following expression for the collision integral in the interaction representation:

$$\sigma(t)=\frac{n_b}{i\hbar}\mathrm{Sp}_b\Big\{[U, \rho(t)\rho_b(t)]+[U, G_0^+(t,t_0)g(t_0)G_0(t,t_0)]$$
$$-\frac{i}{\hbar}\Big[U, \int_{t_0}^{t}S^+(t,t')L_0^+(t,t')[U(t'), \rho(t')\rho_b(t')]$$
$$\times L_0(t,t')S(t,t')\,dt'\Big]\Big\};$$

$$U(t)=FWF^+, \quad \rho(t)=FF_a(t)F^+,$$
$$L_0(t,t')=Y^+(t')L(t,t')Y(t'). \tag{2.9}$$

The collision integral (2.9) differs from others in the presence of the dynamic evolution operator $L(t,t')$, which describes the effects of the strong electromagnetic field and the nonlocal character of the interaction[9] on the OC dynamics. When $L(t,t')=I$ (the Born approximation), Eq. (2.9) yields the result obtained by Apanasevich and Nizovtsev,[15] while in the absence of an electromagnetic field ($S(t,t')=I$) it yields a collision integral that is similar in form to the non-

Markovian collision integral of Ref. 16 but differs essentially from it in content.

When (2.9) is taken into account, the quantum kinetic equation for the single-particle density matrix $\rho(t)$ takes the form[2,12]

$$\left(\frac{\partial}{\partial t}+\frac{\mathbf{p}}{m_a}\nabla\right)\rho(t)=\frac{1}{i\hbar}[V(t),\rho(t)] -\frac{1}{2}[\Gamma\rho+\rho\Gamma]+\sigma(t)+Q(t). \quad (2.10)$$

Here we have introduced the matrix $Q(t)$, which characterizes the pumping, and a supplementary term containing the diagonal matrix Γ, which describes the spontaneous transitions[12]; $V(t)\equiv FV(t)F^{+}$; and $\mathbf{p}$ and m_a are the momentum and mass of the active particle. The generalization of (2.9) and (2.10) to the case of collisions of identical particles $(a-a)$ is trivial.[1]

The non-Markovian collision integral (2.9) makes it possible substantially to extend the range of applicability of quantum kinetic equations of the form of (2.10). In particular, it can be used to investigate new nonlinear dynamical effects due to the influence of a strong electromagnetic field on the OC dynamics[2,3] and to obtain a universal formula for the shapes of gaseous absorption lines (induced emission) that makes it possible to describe the entire spectrum, including the impact, intermediate, and quasistatic frequency regions.

3. THE KINETIC EQUATION FOR THE DENSITY MATRIX

Let us consider the evolution of a two-level quantum system (an atom) in a strong electromagnetic field under collisions with multilevel unpolarized ($\rho_{b\lambda\mu}=\rho_\mu\delta_{\lambda\mu}$) impurity particles. The formulation of the problem corresponds to the nonlinear OC theory[3,4] and differs from the known variants of the theory[3-5] in the method of investigation and in yielding general results for arbitrary electromagnetic-field strengths and arbitrary mismatch between the frequency ω_0 of the electromagnetic-field and the transition frequency ω_{mn}, in accordance with the model[12] based on relaxation of constants.

We shall use the quantum-kinetic equation (2.10), which leads to the following set of differential equations for the matrix elements $\rho(t)$ in the two-level approximation:

$$\left(\frac{\partial}{\partial t}+\frac{\mathbf{p}}{m_a}\nabla+\gamma\right)\rho_{mn}=iVe^{-i(\omega t-\mathbf{kr})}(\rho_{mm}-\rho_{nn})+\sigma_{mn}(t)$$
$$\left(\frac{\partial}{\partial t}+\frac{\mathbf{p}}{m_a}\nabla+\gamma_s\right)\rho_{ss}=\gamma_s q_s\pm 2\,\mathrm{Re}[iV^{*}e^{i(\omega t-\mathbf{kr})}\rho_{mn}(t)]+\sigma_{ss}(t). \quad (3.1)$$

Here $\mathbf{k}$ is the wave vector and $\mathbf{r}(t)$ the radius vector of the particle. In the equations for the diagonal matrix elements $\rho_{ss}(t)$ the plus and minus signs correspond to $s=m$ and $s=n$, respectively, while the $q_s(\mathbf{r},\mathbf{p},t)$ characterize the incoherent pumping and determine the steady-state populations of the energy levels in the absence of an electromagnetic field. The constants γ, γ_m, and γ_n describe the spontaneous transitions,[12] while $V=V_{mn}/\hbar$ and $\omega=\omega_0-\omega_{mn}$.

In the subsequent investigations we shall need the explicit form of the collision integral (2.9). To simplify the formulas we shall assume that $\mathbf{p}=0$, since in the final formulas for the characteristics of the particles having nonvanishing momenta, the $\mathbf{p}$ dependence can be recovered with the aid of the standard substitution[12] $\omega\to\omega-\mathbf{kv}$, $\mathbf{v}=\mathbf{p}/m_a$.

Since the off-diagonal elements of the matrix $U(t)$ are rapidly oscillating functions of time in the interaction representation, their effect on collisions with unpolarized impurity particles turns out to be negligibly small (in the two-level approximation). We shall accordingly assume the matrix $U(t)$ to be diagonal: $U_{ss'}=\hbar U_s\delta_{ss'}$ (elastic processes).

The operators $Y(t)$ and $S^{+}(t,t-\tau)$ have the following form[12,15] for a two-level system in a monochromatic electromagnetic field:

$$Y(t)=\frac{1}{\sqrt{2}}\begin{Vmatrix} c_{+}^{1/2}e^{i\omega_{-}t} & -c_{-}^{1/2}e^{-i(\omega_{+}t+\varphi_0)} \\ c_{-}^{1/2}e^{i\omega_{+}t} & c_{+}^{1/2}e^{-i(\omega_{-}t+\varphi_0)}\end{Vmatrix}, \quad (3.2)$$

$$S^{+}(t,t-\tau)=Y^{+}(t)\,Y(t-\tau)=\frac{1}{2}\begin{Vmatrix} a_{11}(\tau) & a_{12}(t,\tau) \\ -a_{12}^{*}(t,\tau) & a_{11}^{*}(\tau)\end{Vmatrix}, \quad (3.3)$$

where we have used the notation

$$a_{11}(\tau)=c_{+}e^{-i\omega_{-}\tau}+c_{-}e^{-i\omega_{+}\tau}, \qquad c_{\pm}=1\pm\frac{\omega}{\nu},$$
$$a_{12}(t,\tau)=\frac{2|V|}{\nu}(e^{i\omega_{-}\tau}-e^{i\omega_{+}\tau})e^{-i(\omega t+\varphi_0)}, \quad (3.4)$$
$$\omega_{\pm}=\frac{1}{2}(\omega\pm\nu), \qquad \nu^2=\omega^2+4|V|^2;$$

and φ_0 is the phase constant of the matrix element $V=\frac{1}{2}(\mathbf{d}_{mn}\mathbf{E})=|V|e^{i\varphi_0}$.

Analysis showed that the final results do not depend on the phase φ_0, so for simplicity we shall assume it to be zero ($\varphi_0=0$) in the intermediate formulas.

Using the matrix (3.2), we obtain the differential equation for the dynamic evolution operator $L^{+}(t,t_0)$:

$$i\frac{\partial L^{+}(t,t_0)}{\partial t}=U_1(t)L^{+}(t,t_0), \qquad L^{+}(t,t)=I; \quad (3.5)$$

where

$$U_1(t)=\frac{1}{\hbar}Y(t)UY^{+}(t) =\frac{1}{2}\begin{Vmatrix} \frac{\omega}{\nu}\Delta U+U_0 & 2\frac{|V|}{\nu}\Delta Ue^{-i\nu t} \\ 2\frac{|V|}{\nu}\Delta Ue^{i\nu t} & -\frac{\omega}{\nu}\Delta U+U_0\end{Vmatrix};$$
$$\Delta U=U_m-U_n, \quad U_0=U_m+U_n. \quad (3.6)$$

Equation (3.5) has the same form as the differential equation for the matrix identified in Ref. 18 with the scattering matrix in a basis of dressed states.[3,5]

The basic and most complicated problem of nonlinear OC theory is to determine the dynamic evolution operator $L^{+}(t,t_0)$. This is due to the time dependence of the matrix $U_1(t)$, which, in turn, is due both to the dependence of the collision interaction potential on the distance $|\mathbf{r}_a(t)-\mathbf{r}_b(t)|$

between the particles and to the effect of the strong electromagnetic field. The following section is devoted to the solution of the dynamical problem.

4. THE DYNAMIC EVOLUTION OPERATOR

It is not difficult to show, using (3.6), that the dynamic evolution operator $L^+(t,t_0)$ can be expressed as a product of two operators:

$$L^+(t,\ t_0)=L_1^+(t,\ t_0)L_2^+(t,\ t_0), \tag{4.1}$$

where $L_1^+(t,t_0)$ is a diagonal matrix with the matrix elements

$$L_{1ss}^+(t,t_0)=\exp\left\{\frac{i}{2}\left[\pm\eta(t,t_0)-\int_{t_0}^{t}U_0(\tau)\,d\tau\right]\right\};$$

$$\eta(t,t_0)=\frac{\omega}{\nu}\int_{t_0}^{t}\Delta U(\tau)\,d\tau, \tag{4.2}$$

while the operator matrix $L_2^+(t,t_0)$ satisfies the differential equation

$$i\frac{\partial L_2^+(t,t_0)}{\partial t}=U_2(t,t_0)L_2^+(t,t_0),\quad L_2^+(t,t)=I. \tag{4.3}$$

Here we have used the notation

$$U_2(t,t_0)=\begin{Vmatrix}0 & \varkappa(t,t_0)\\ \varkappa^*(t,t_0) & 0\end{Vmatrix};$$
$$\varkappa(t,t_0)=\frac{|V|}{\nu}\Delta U(t)\exp\{-i[\nu t-\eta(t,t_0)]\}. \tag{4.4}$$

To solve the differential equation (4.3) we transform to equations for the matrix elements $l_{ss'}=(L_2^+)_{ss'}$, which form two pairs of coupled equations with the same coefficients. To investigate the set of differential equations for $l_{mm}(t,t_0)$ and $l_{nm}(t,t_0)$ we introduce the new complex functions $\varphi_m(t,t_0)$ and $\varphi_n(t,t_0)$:

$$l_{mm}(t,\ t_0)=\exp\varphi_m(t,\ t_0);$$
$$il_{nm}(t,\ t_0)=\exp\varphi_n(t,\ t_0). \tag{4.5}$$

From (4.3) we obtain the following differential equation for the difference between the arguments of the exponentials in (4.5):

$$\frac{d\varphi_{nm}}{dt}\equiv\dot\varphi_{nm}=\varkappa e^{\varphi_{nm}}+\varkappa^*e^{-\varphi_{nm}},\qquad \varphi_{nm}=\varphi_n-\varphi_m. \tag{4.6}$$

On separating the real parts in equations of the form of (4.6) and solving the resulting equations by the Vainshtein-Presnyakov-Sobel'man (VPS) method[19] we obtain an expression for the matrix elements of the operator $L_2^+(t,t_0)$:

$$l_{ss}(t,\ t_0)=[\cos\lambda(t,\ t_0)]e^{i\psi_s(t,\ t_0)},\quad s=m,\ n,$$
$$l_{ss'}(t,\ t_0)=-i[\sin\lambda(t,\ t_0)]e^{i\psi_s(t,\ t_0)},\quad s\neq s', \tag{4.7}$$

where we have used the notation

$$\lambda(t,t_0)=\int_{t_0}^{t}\mathrm{Re}[\varkappa(t',t_0)e^{i\psi(t',t_0)}]dt',$$
$$\psi_s(t,\ t_0)=\mathrm{Im}\,\varphi_s(t,\ t_0),\ \psi(t,\ t_0)=\mathrm{Im}\,\varphi_{nm}(t,\ t_0). \tag{4.8}$$

To determine the phases $\psi_s(t,t_0)$, we deviate from the VPS method and use second-order differential equations that are equivalent to the initial matrix equation (4.3):

$$\ddot\varphi_s+\dot\varphi_s^2-\alpha_s\dot\varphi_s+|\varkappa|^2=0,\quad s=m,\ n;$$
$$\alpha_m=\frac{1}{\varkappa}\frac{d\varkappa}{dt},\qquad \alpha_n=\alpha_m^*. \tag{4.9}$$

It is not difficult to verify that the solutions of these equations are the functions

$$\dot\varphi_s=\frac{1}{2}\alpha_s\pm i\left\{J_s^2\pm i\frac{d}{dt}\left[J_s^2\pm i\frac{d}{dt}(J_s^2\pm\ldots)^{1/2}\right]^{1/2}\right\}^{1/2},$$
$$J_m^2=\frac{1}{4}[(\omega-\Delta U)^2+4|V|^2]-\frac{1}{2}i\nu\frac{\Delta\dot U}{\Delta U}+\frac{1}{2}\left(\frac{\Delta\dot U}{\Delta U}-\frac{3}{2}\frac{\Delta\dot U^2}{\Delta U^2}\right),$$
$$J_n=J_m^*. \tag{4.10}$$

Taking (4.4) and (4.8) into account, we obtain the following expressions for the phases $\psi(t,t_0)$ and $\psi_s(t,t_0)$:

$$\psi(t,t_0)=\nu(t-t_0)-\eta(t,t_0)-2\,\mathrm{Re}\int_{t_0}^{t}J(\tau)\,d\tau, \tag{4.11}$$

$$J(\tau)=\left\{J_m^2+i\frac{d}{dt}\left[J_m^2+i\frac{d}{dt}(J_m^2+\ldots)^{1/2}\right]^{1/2}\right\}^{1/2}, \tag{4.12}$$
$$\psi_n(t,\ t_0)=-\psi_m(t,\ t_0)={}^1/_2\psi(t,\ t_0).$$

The method of obtaining the phases $\psi_s(t,t_0)$ from equations of the form of (4.9) is a generalization of the WKB and VPS methods[19,21] to the case in which the nonadiabatic character of the collisions and the effect of the electromagnetic field on their dynamics are taken into account. The generalization also involves the fact that the condition $|J_m|^2\gg1$, which is characteristic of the asymptotic methods[19,21] does not apply to the quantity $|J_m|$. We note that the VPS method corresponds to the $J(\tau)\approx J_m(\tau)$. approximation.

Formulas (4.2), (4.7), and (4.11) completely solve the problem of determining the dynamic evolution operator $L^+(t,t_0)$:

$$L^+(t,t_0)=\begin{Vmatrix}l_1(t,t_0) & -ir_1(t,t_0)\\ -ir_1^*(t,t_0) & l_1^*(t,t_0)\end{Vmatrix}, \tag{4.13}$$

where we have used the notation

$$l_1(t,t_0)=[\cos\lambda(t,t_0)]e^{i\varphi(t,\ t_0)},\ r_1(t,t_0)=[\sin\lambda(t,t_0)]e^{i\varphi(t,\ t_0)}, \tag{4.14}$$

and

$$\varphi(t,t_0)=\frac{1}{2}\left[\pm\nu(t-t_0)+2\,\mathrm{Re}\int_{t_0}^{t}J(\tau)\,d\tau\right]. \tag{4.15}$$

The plus and minus signs in (4.15) correspond to the cases $\omega<0$ and $-\omega>0$, respectively. In the adiabatic approximation,[10] in which the components proportional to $\Delta\dot U$ and $\Delta\ddot U$ are neglected in (4.12), the expression for the phase $\varphi(t,t_0)$ takes the form

$$\varphi(t,t_0)=\frac{1}{2}\left[\pm\nu(t-t_0)+\int_{t_0}^{t}[(\omega-\Delta U)^2+4|V|^2]^{1/2}d\tau\right]. \tag{4.16}$$

The relation (4.13) obtained for the dynamic evolution operator $L^{+}(t,t_0)$ enables us to proceed to the development of a nonlinear quantum kinetic theory of spectral line contours.

5. A UNIVERSAL FORMULA FOR SPECTRAL LINE CONTOURS

On substitution the matrices (3.3) and (4.13) for $S^{+}(t,t-\tau)$ and $L^{+}(t,t-\tau)$ into Eq. (2.9) we obtain, in the limit $t_0 \to -\infty$ with $g(t_0)=0$, the matrix elements for the collision integral $\sigma(t)$:

$$\sigma_{mn}(t) = -P_1\rho_{mn}(t) + P_2 e^{-2i(\omega t+\varphi_0)}\rho_{mn}^{*}(t),$$
$$\sigma_{nm}(t) = \sigma_{mn}^{*}(t), \quad \sigma_{mm} \approx \sigma_{nn} \approx 0. \tag{5.1}$$

Here we have introduced the following notation:

$$P_1(|V|,\omega) = i\Delta\bar{U} + n_b\Big\langle \int_{-\infty}^{\infty} \rho_b \Delta U(t)\,dt \int_{0}^{\infty} \Delta U(t-\tau) \times [d_1(\tau) l_1^{2}(t,t-\tau) + d_2(\tau) r_1^{*2}(t,t-\tau)]\,d\tau \Big\rangle; \tag{5.2}$$

$$P_2(|V|,\omega) = -n_b \Big\langle \int_{-\infty}^{\infty} \rho_b \Delta U(t)\,dt \int_{0}^{\infty} \Delta U(t-\tau) \times [d_1(\tau) r_1^{2}(t,t-\tau) + d_2(\tau) l_1^{*2}(t,t-\tau)]\,d\tau \Big\rangle,$$

where in turn, we have used the notation

$$d_1(\tau) = \frac{2|V|^2}{v^2} + \frac{1}{4}\left(1+\frac{\omega}{v}\right)^2 e^{iv\tau} + \frac{1}{4}\left(1-\frac{\omega}{v}\right)^2 e^{-iv\tau},$$
$$d_2(\tau) = \frac{|V|^2}{v^2}(e^{iv\tau}+e^{-iv\tau}-2), \quad \Delta\bar{U} = n_b\Big\langle \int_{-\infty}^{\infty} \Delta U \rho_b\,d\tau \Big\rangle. \tag{5.3}$$

The angle brackets $\langle \cdots \rangle$ denote averaging over the relative velocities v of the particles and the impact parameter b and summing over the index μ.

In deriving Eqs. (5.1) and (5.2) we considered only the quasiclassical approximation,[10] although the collision integral (2.9) contains information on the changes in the momenta of the particles during collisions.[2,12]

To determine the absorption-line contour (induced emission) we must find the off-diagonal element of the density matrix $\rho_{mn}(t)$. Using formulas (5.1)–(5.3) we obtain the following expression for the steady-state value of $\rho_{mn}(t)$ from the set of differential equations (3.1):

$$\rho_{mn}(t) = \frac{iVe^{-i\omega t}(q_m-q_n)(\gamma+P_1^{*}-P_2+i\omega)}{|P_1+\gamma-i\omega|^2+2|V|^2(\gamma+\gamma_{oc})(\gamma_m^{-1}+\gamma_n^{-1})-|P_2|^2}, \tag{5.4}$$

where we have used the notation

$$\gamma_{oc}(|V|,\omega) = \operatorname{Re} P(|V|,\omega),$$
$$P(|V|,\omega) = P_1(|V|,\omega) - P_2(|V|,\omega)$$
$$= i\Delta\bar{U} + n_b\Big\langle \int_{-\infty}^{\infty} \rho_b \Delta U(t)\,dt \int_{0}^{\infty} \Delta U(t-\tau) \times [d_1(\tau)e^{2i\varphi} + d_2(\tau)e^{-2i\varphi}]\,d\tau \Big\rangle, \tag{5.5}$$

and $\gamma_{oc}(|V|,\omega)$ is the half width of the line contour due to OC transitions.

Equation (5.4) enables us to determine the entire contour of an atomic absorption line in the presence of collisions with multilevel particles in a strong electromagnetic field:

$$I(|V|,\omega) \sim \operatorname{Re}[-iV^{*}(t)\rho_{mn}(t)] = \frac{|V|^2(q_m-q_n)[\gamma+\gamma_{oc}(|V|,\omega)]}{[\omega-\Delta_{oc}(|V|,\omega)]^2+[\gamma+\gamma_{oc}(|V|,\omega)]^2+2|V|^2(\gamma+\gamma_{oc})(\gamma_m^{-1}+\gamma_n^{-1})+pq}, \tag{5.6}$$

where

$$\Delta_{oc}(|V|,\omega) = \operatorname{Im} P_1(|V|,\omega);$$
$$pq = 2\operatorname{Re} P_2(\gamma+\gamma_{oc}) - (\operatorname{Im} P_2)^2. \tag{5.7}$$

Formally, Eq. (5.6) has the same form as the Karplus-Schwinger formula[11] as generalized by Lisitsa and Yakovlenko[3] to the case in which a strong electromagnetic field is present, but there are also substantial differences.

First, the collisional shift $\Delta_{oc}(|V|,\omega)$, which depends on the strength of the electromagnetic field and the frequency ω, appears in Eq. (5.6). Formula (5.7) for $\Delta_{oc}(|V|,\omega)$ describes a new nonlinear dynamic effect for optical collisions in a strong electromagnetic field.

Second, the term in the denominator proportional to $|V|^2$ contains the coefficient $(\gamma_m^{-1}+\gamma_n^{-1})$, which reflects the difference between the lifetimes of particles on levels m and n (an open system) in place of the factor $\gamma_{i.r.}^{-1}$, which characterizes[3,11] the inelastic relaxation.[3,11] It is evident from (5.6) that the description of the line contour with the aid of the two constants[3,11] γ_{oc} and $\gamma_{i.r.}$ is valid when the conditions $\gamma_m = \gamma_n = 2\gamma$ and $\gamma = \gamma_{i.r.}$ are satisfied; but these conditions are usually not satisfied[12] in real systems.

Thus, the component $(\gamma+\gamma_{oc})^2$ in the denominator of the Lisitsa-Yakovlenko formula[3] was introduced artificially, since the theory developed in Ref. 3 is applicable only for frequencies $v^2 \gg (\gamma+\gamma_{oc})^2$. In Eq. (5.6), however, the component $(\gamma+\gamma_{oc})^2$ appears along with a new additive term pq in a natural manner, the additional term pq indicating that it is permissible to introduce the component $(\gamma+\gamma_{oc})^2$ into the denominator of (5.6) only for values of $|V|^2$ and ω for which $pq \ll (\gamma+\gamma_{oc})^2$.

An important property of Eq. (5.6) is the fact that in the absence of collisions ($\gamma_{oc} = \Delta_{oc} = 0$) it, unlike the known formulas of Refs. 3 and 11, leads to the absorption-line contour due to spontaneous relaxation in a strong electromagnetic field.[12] This means that (5.6) contains information on the absorption of radiation not only during OC transi-

tions,[3,11] but also during the free flight of the atoms.

We note that two nonlinear spectroscopic effects that differ in nature find their expression in Eq. (5.6). The first is associated with the leveling of the populations of the energy levels in a strong electromagnetic field (the saturation effect), which is well known from the theory of lasers (see, e.g., Ref. 12). The second effect is comparatively new; it is due to an effect of a strong electromagnetic field on the OC dynamics that leads to a dependence of the characteristics γ_{oc} and Δ_{oc} of the collisional relaxation on the strength of the electromagnetic field (a nonlinear dynamical effect). The specific physical manifestation of this effect was first investigated in Ref. 2 and in Refs. 3 and 4.

We have analyzed the formal distinctive features of Eq. (5.6), which is a generalization of the well-known Lorentz-Weisskopf, Karplus-Schwinger,[11] and Lisitsa-Yakovlenko[3] formulas, and others. A more thorough analysis of the results can be carried through by considering the physical content of the functions $\gamma_{oc}(|V|, \omega)$, $\Delta_{oc}(|V|, \omega)$, and pq as applied to different ranges of variation of the parameters ω and $|V|^2$.

Formula (5.6) characterizes the contour of a spectral line of an individual atom. To obtain the absorption-line contour for an ensemble of atoms (in a unit volume) one must average (5.6) over the velocities $\mathbf{v}$ of the active particles (with allowance for the substitution $\omega \to \omega - \mathbf{kv}$), over the orientations of the dipole moments,[22] and over the spatial distribution of the active particles.

6. APPROXIMATE FORMULAS FOR THE WIDTH AND SHIFT

Analysis of Eqs. (5.5) and (5.7) for $\gamma_{oc}(|V|, \omega)$ and $\Delta_{oc}(|V|, \omega)$ showed that, as applied to the impact region of the spectrum ($\nu\tau_c \ll 1$, where τ_c is the duration of a collision), they yield the classical formulas for γ_{imp} and Δ_{imp} provided $\rho_b = 1$ (Ref. 10, p. 252).

In the quasistatic frequency region corresponding to the intersection of the difference of the "terms" ΔU and ω, in the case of a weak electromagnetic field and a multipole interaction,[10] Eqs. (5.5) and (5.6) yield the well known quasistatic distribution in the line wing (Ref. 10, p. 254).

In the case of a strong electromagnetic field, for the Landau-Ziner region[3,23] with allowance for the identity

$$(\omega-\Delta U)^2+4|V|^2 \equiv \frac{4|V|^2}{\nu^2}(\Delta U)^2+\left(\nu-\frac{\omega}{\nu}\Delta U\right)^2$$

we obtain[20] the following expression in the adiabatic approximation (4.16):

$$\gamma_{oc}(|V|, \omega)=n_b\left(1+\frac{\omega}{\nu}\right)\langle \rho_b \exp[-\delta(|V|, \omega, v)]\rangle,$$
$$\delta=\frac{2\pi|V|^2}{|D|v}\left(1+\frac{4|V|^2}{\omega^2}\right), \quad D=\left.\frac{d\Delta U}{dr}\right|_{r=r_0}, \quad \omega>0. \tag{6.1}$$

When $\omega^2 \gg 4|V|^2$ and $\rho_b = 1$, Eqs. (6.1) and (5.6) lead to the results of Ref. 3. Both the field narrowing[2] of the spectral line contours and the brightening of the medium in the quasistatic frequency region[3] follow from Eqs. (5.5), (5.6), and (6.1).

A comparison of relations (5.5), (5.7), and (6.1) with the known results of Refs. 3, 4, 10, 20, and 23 shows that Eqs. (5.5)–(5.7) describe the entire contour of atomic spectral lines from unified positions for an electromagnetic field of arbitrary strength, while the following approximate expressions for $\gamma_{oc}(|V|, \omega)$ and $\Delta_{oc}(|V|, \omega)$ (for $\rho_b = 1$) can be used to analyze various limiting cases:

$$\gamma_{oc}(|V|, \omega)=\mathrm{Re}\, P_0(|V|, \omega), \quad \Delta_{oc}(|V|, \omega)=\mathrm{Im}\, P_0(|V|, \omega), \tag{6.2}$$

where we have used the notation

$$P_0(|V|, \omega)=i\Delta\overline{U}+n_b\left\langle \int_{-\infty}^{\infty}\Delta U(t)\,dt\int_{-\infty}^{t}\Delta U(t')\,e^{i\varphi_0(t,t')}\,dt'\right\rangle;$$
$$\varphi_0(t, t')=\int_{t'}^{t}[(\omega-\Delta U)^2+4|V|^2]^{1/2}\,d\tau. \tag{6.3}$$

These formulas are not rigorous enough to describe the intermediate frequency region in which the coefficients $d_1(\tau)$ and $d_2(\tau)$ may play an important part.

7. NONLINEAR SHIFT OF THE SPECTRAL-LINE CONTOURS

For a qualitative study of the new nonlinear dynamical effect—the dependence of the shift $\Delta_{oc}(|V|, \omega)$ on the strength of the electromagnetic field—we use Eqs. (6.2) and (6.3), which yield the following expression valid at $\omega = 0$ (the center of the contour):

$$\Delta_{oc}(|V|)\approx n_b\left\langle \int_{-\infty}^{\infty}dt\Delta U(t)\left[1+\int_{0}^{\infty}\Delta U(t-\tau)\right.\right.$$
$$\left.\left.\times\sin\varphi_0(t, t-\tau)\,d\tau\right]\right\rangle. \tag{7.1}$$

Equation (7.1) yields the classical formula for the impact shift in the limit $|V| \to 0$:

$$\Delta_{imp}=n_b\left\langle \sin\int_{-\infty}^{\infty}\Delta U(t)\,dt\right\rangle.$$

In the other limiting case of a strong electromagnetic field ($|V|^2 \gg (\Delta U)^2$ and $|V|\tau_c \gg 1$) the integrand in the nonlinear component of (7.1) is a rapidly oscillating function of time and its contribution to $\Delta_{oc}(|V|)$ turns out to be negligible. In that case the shift $\Delta_{oc}(|V|)$ reaches its maximum value

$$\Delta_{oc}^{max}(|V|)\approx\Delta\overline{U}\equiv n_b\left\langle \int_{-\infty}^{\infty}\Delta U(t)\,dt\right\rangle.$$

We note that the divergence of $\Delta U(r)$ as $r \to 0$ is compensated by the nonlinear shift in (7.1), so that the following condition is satisfied in the region where $r = (b^2 + v^2t^2)^{1/2}$ is small:

$$[(-\Delta U)^2+4|V|^2]^{1/2}\approx(-\Delta U).$$

In the linear approximation in $|V|$ (when $|V|\tau_c \ll 1$ we obtain

$$\Delta_{oc}(|V|)\approx\Delta_{imp}+2|V|\tau_c\gamma_0,$$

$$\gamma_0 = n_b \left\langle \int_{-\infty}^{\infty} \Delta U(t)\, dt \int_0^{\infty} \Delta U(t-\tau)\, \tau\tau_c^{-1} \left(\cos \int_{t-\tau}^{t} \Delta U\, d\tau_1 \right) d\tau \right\rangle .$$

from Eq. (7.1). Consequently, the shift $\Delta_{oc}(|V|)$ increases with increasing strength of the electromagnetic field; moreover, in the general case $\Delta_{oc}(|V|)$ increases nonlinearly, the character of the nonlinearity being determined by the form of the potential difference $\Delta U(r)$. The increase in the shift $\Delta_{oc}(|V|)$ may be attributed to the enhancement of the effect of collisions with large impact parameters as a result of the field splitting of the energy levels in a strong electromagnetic field.

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