Brownian motion of atoms and ions and nonlinear effects in the absorption of light

F. F. Baryshnikov and V. S. Lisitsa

I. V. Kurchatov Institute of Atomic Energy
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Nonlinear effects in the absorption spectra of atoms and ions experiencing stochastic (Brownian) accelerations are considered. The absorption of intense radiation by Brownian-moving atoms is considered by two methods: 1) by the method of integration over the Wiener-Feynman paths and 2) by the kinetic method with a Fokker-Planck collision integral. The two methods are shown to be equivalent. A nonlinear dependence of the absorbed power on the electromagnetic field strength, which results in a decrease of absorption in strong fields, is found.

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§1. INTRODUCTION

The kinetic equation for the density matrix $\hat{\rho}$ of the atom is the basis for the description of the interaction of resonant radiation with the atoms of ions in a gas or plasma. This equation contains in its right-hand side the collision integral $\hat{S}(v)$ describing the evolution of the atomic states in velocity space.[1-3] The sensitivity of the absorption effects to the change in the velocity of the atom is due to the fact that one of the basic mechanisms of broadening here is the Doppler effect, in which the basic contribution to the absorption is made by the points of resonance $kv = \Delta\omega$ ($k = \omega/c$ is the wave vector of radiation with frequency ω, v is the velocity of the atom, $\Delta\omega = \omega - \omega_0$ is the detuning from resonance between the frequency ω and the characteristic frequency of the transition ω_0).

The solutions of the equation for $\hat{\rho}$ in the case of different forms of the collision integral $\hat{S}(v)$ have been investigated in detail for the case of sufficiently weak intensity of the electromagnetic (EM) field E_0, i.e., in the linear approximation, see Refs. 1–3. We also note the recent work of Alekseev and Malyugin,[4] in which a systematic method of calculation of the absorption spectra was developed for an arbitrary form of $\hat{S}(v)$ at a sufficiently large value of the latter.

In the nonlinear problem, the situation is quite complicated, and the only actual example of an exact solution in this case is the strong-collisions model, see Ref. 2. In the present work, nonlinear effects in the absorption of light are considered for the strong-collisions model, which corresponds to Brownian motion of the absorbing atom or ion. The Brownian motion model describes an important circle of physical phenomena connected with the collisions both of a heavy absorbing particle in a light buffer gas and of ions in a plasma.[1-3]

Two approaches to the calculation of the absorption probability in the Brownian motion model should be noted here. The first approach is based on the solution of the equations for the amplitudes a_m of the atomic states, which depend parametrically on the stochastic parameter $v(t)$—the velocity of the atom. The general solutions that are obtained must be averaged over all possible trajectories of the velocity. Such an approach is similar to a known degree to the Feynman treatment in quantum mechanics.[5] This method was used in its linear approximation by Podgoretskii and Stepanov.[6] The calculation of succeeding corrections in the value E_0 of the EM field has been carried out by Rautian.[7]

The second (more traditional) approach is based on the solution of the kinetic equation, indicated above for the density matrix $\hat{\rho}$ with a collision integral of the Fokker-Planck type:[3,8]

$$\hat{S}(f) = \beta \frac{v_T^2}{2} \frac{d^2 f}{dv^2} + \beta \frac{d}{dv}(vf), \tag{1.1}$$

where β is the frequency of collisions of the particle, v_T is the most probable velocity.

This approach in similar in many ways to the Schrödinger treatment of quantum mechanics: $\hat{S}$ plays the role of the kinetic energy operator, and the Doppler frequency shift kv corresponds to the potential energy operator.

The solution of the kinetic equation with the integral (1.1) has been carried out by Smirnov and Shapiro[9] within the framework of perturbation theory in the magnitude of the field (see also Ref. 2). An approach outside the limits of perturbation theory has been given by Kofman and Burshteĭn[10] in the rate-equation approximation. The sense of this approximation is that the relaxation of the polarization is assumed to be the largest parameter of the problem, as a result of which it turns out to be proportional to the population of the states that change as a consequence of the Brownian motion of the atom.

In this research, in contrast to Ref. 10, the Brownian collisions are responsible for the relaxation of both the populations and of the polarization itself, so that the collision frequency β is the only relaxation parameter of the system. We restrict ourselves to the case $\beta/kv_0 \ll 1$, which corresponds to diffusion of the atoms in velocity space.

Since the relation between the two mentioned approaches which are based on path integrals and on the kinetic equation method, is of significant interest, we shall pay particular attention to their comparison.

The physical meaning of the nonlinear effects that

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arise is easily made clear by analyzing the linear kinetic equation with a collision integral of the diffusion type:

$$\hat{S}(f)=Dd^2f/dv^2$$

($D=v_0^2\beta$ is the diffusion coefficient in velocity space). Comparing $\hat{S}$ with the Doppler shift $(\Delta\omega-kv)$, it is easy to see that a narrow band of frequencies near the resonance $kv=\Delta\omega$, with effective width $\delta\omega_{\text{eff}}\sim kv_0(\beta/kv_0)^{1/3}$, is responsible for the absorption. The interaction with the strong field $G=\mathbf{d}_{12}\cdot\mathbf{E}_0$ ($\mathbf{d}_{12}$ is the dipole moment of the transition) leads to the forcing out of the frequency of the atomic oscillator $\Omega=[(\Delta\omega-kv)^2+4G^2]^{1/2}$ from this resonance region and, consequently, to a decrease in the absorption at $G\gtrsim\delta\omega_{\text{eff}}$.

It should be noted that the interaction of an atom with a strong EM field generally leads to violation of the resonance condition $\Delta\omega=kv$. Therefore, to obtain an absorption that is different from zero in this case, we must introduce some sort of relaxation mechanism. The usual introduction of a spontaneous relaxation mechanism γ leads to the well-known results of the theory of inhomogeneous broadening.[1-3] However, even at $\gamma=0$, the atom in the strong EM field, "being located in deep saturation," has the possibility of absorbing radiation if its velocity changes during the absorption process. Investigation of just these nonlinear effects, brought about by the relaxation of the velocity, is the purpose of the present work. We shall therefore limit ourselves below to the simplest setup of the problem: the atom is assumed to be a two-level system, and the only relaxation mechanism is the relaxation of its velocity as a result of the Brownian motion.

§2. METHOD OF THE AMPLITUDES OF THE STATES AND CONTINUOUS INTEGRATION

The system of equations for the amplitudes of the states of a two-level atom with resonance radiation has the form (see Ref. 2)

$$i\dot{a}_1=[kv(t)-\Delta\omega]a_1+Ga_2,\quad i\dot{a}_2=Ga_1,\quad a_2(v=v_0)=0. \tag{2.1}$$

The initial condition presented corresponds to pumping to level 1. The final expression for the absorbed power must be averaged over the pumping distribution $W(v_0)$, for example, over the Maxwell distribution.

The power $P(\omega)$ that is absorbed is expressed in terms of the probability w_{12} of the transition per unit time

$$P(\omega)=N\hbar\omega\langle w_{12}\rangle\equiv N\hbar\omega G^2I(\omega). \tag{2.2}$$

The angular brackets denote averaging over the realizations of random trajectories and over the pumping.

In our case, the law of motion of the absorbing particle or the dependence $v=v(t)$ is determined by the Langevin equation[8]

$$dv/dt+\beta v=n(t). \tag{2.3}$$

Here β is the damping parameter, $n(t)$ is a delta-correlated random force.

Thus, the system (2.1) contains the stochastic parameter $v(t)$ and is therefore a system of stochastic differential equations. Here and below, we shall understand by $v(t)$ the component of the velocity along the vector k, for which one-dimensional distributions are used everywhere.

To average in (2.2) over all the realizations of the random process, it is necessary to start out from the general solution of the system (2.1) without any assumptions on the character of the change in $v(t)$.

Such a general solution is easily obtained in the case of weak fields (perturbation theory). In the case of an arbitrary G, there is no general analytic solution. However, there does exist an approximate analytic solution of Vaĭnshteĭn, Presnyakov and Sobel'man (VPS),[11] which correctly describes the known limiting cases and is in good agreement with the numerical solutions of the system (2.1) for various potentials.

We use the VPS approximation for the determination of the probability w_{12}, obtaining (cf. the similar formula in Ref. 12)

$$I(\omega)=\left\langle\frac{1}{\pi}\operatorname{Re}\int_0^\infty dt\exp\left[i\int_0^t\Omega(\tau)d\tau\right]\right\rangle,$$
$$\Omega(\tau)=\{[kv(\tau)-\Delta\omega]^2+4G^2\}^{1/2}. \tag{2.4}$$

Equation (2.4) can be regarded as a natural generalization of the model of an oscillator with variable frequency[1] to the case of strong fields G when the phase of the oscillator becomes dependent on the value of G [as a consequence of the dependence on G of the Rabi frequency $\Omega(\tau)$].

Averaging over the realizations of the random process (2.3) can be carried out in the following way. We break up the interval of integration over τ in (2.4) into N identical parts. Then the average over the trajectories can be defined as

$$\left\langle\exp\left(i\int_0^t\Omega(\tau)d\tau\right)\right\rangle=\lim_{N\to\infty}\int\dots\int dv_0\dots dv_NW(v_0)W\left(v_0,v_1,\frac{t}{N}\right)$$
$$\dots W\left(v_k,v_{k+1},\frac{t}{N}\right)\dots W\left(v_{N-1},v_N,\frac{t}{N}\right)\prod_{k=0}^{N}\exp\left(i\frac{t}{N}\Omega[v_k(\tau)]\right). \tag{2.5}$$

Here $W(v_0)$ is the initial velocity distribution, $W(v_k, v_{k+1}, t/N)$ is the probability of the transition $v_k\to v_{k+1}$ within the time t/N, which, in the Brownian approximation (2.3), is equal to

$$W(v_0)=\frac{1}{(\pi v_T^2)^{1/2}}\exp[-(v/v_T)^2],$$
$$W(v_k,v_{k+1},\Delta t)=\frac{1}{[\pi v_T^2(1-e^{-2\beta\Delta t})]^{1/2}}\exp\left[-\frac{(v_{k+1}-v_ke^{-\beta\Delta t})^2}{v_T^2(1-e^{-2\beta\Delta t})}\right]. \tag{2.6}$$

The limiting transition in (2.5) determines the so-called continuous integral over the Gaussian measure,[5,13-15] which is given by the transition probability (2.6).

Direct calculation of the integrals (2.5) is not possible because of the nonlinear dependence on Ω on v. This calculation can be carried through only in the case of weak fields $G\to 0$, when $\Omega(\tau)=kv(\tau)-\Delta\omega$. Direct evaluation of the Gaussian integrals leads to the result:[16]

$$\Phi(\tau)=\left\langle \exp\left(ik\int_0^\tau v(t)\,dt\right)\right\rangle=\exp\left(-\frac{1}{2}k^2\langle r^2(\tau)\rangle\right)$$
$$=\exp\left[-\frac{k^2\bar{v}^2}{2\beta^2}(\beta\tau-1+e^{-\beta\tau})\right]. \qquad (2.7)$$

The argument in the exponential is simply the mean squared displacement $\langle r^2(\tau)\rangle$ calculated in the Brownian-motion model.[8] The characteristic $\Phi(\tau)$ dependence in the limit of small ($\beta\tau\ll 1$) and large ($\beta\tau\gg 1$) times has the form

$$\Phi(\tau)\approx\begin{cases}\exp\left(-\dfrac{k^2\bar{v}^2\tau^2}{4}\right), & \beta\tau\ll 1\\ \exp\left(-\dfrac{k^2\bar{v}^2}{2\beta}\tau\right), & \beta\tau\gg 1\end{cases} \qquad (2.8)$$

Taking the Fourier transform of (2.7), we get for the contour $I(\omega)$[1)]

$$I(\omega)\approx\begin{cases}\dfrac{1}{\pi^{1/2}kv_T}\exp\left\{-\left(\dfrac{\Delta\omega}{kv_T}\right)^2\right\}, & \dfrac{\beta}{kv_T}\ll 1\\ \dfrac{1}{\pi}\dfrac{\gamma_D}{\gamma_D^2+\Delta\omega^2}, & \dfrac{\beta}{kv_T}\gg 1\end{cases} \qquad (2.9)$$

Thus, the Doppler contour of the line at $\beta/kv_T\ll 1$ is replaced by a Lorentzian contour at $\beta/kv_T\gg 1$ with width $\gamma_D=(kv_T)^2/2\beta$, which is smaller by a factor $kv_T/\beta\ll 1$ in comparison with the Doppler width. This effect is known as the Diecke narrowing effect, see Refs. 1–3.

We now proceed to the case of strong fields G. The calculations here are very complicated in view of the nonlinear connection of Ω with the stochastic variable v. Therefore we shall limit ourselves below to the case $\beta/kv_T\ll 1$, which corresponds to small relaxation. In this case, the complete Fokker-Planck measure (2.6) can be replaced by the following much simpler form, which is obtained from (2.6) at $\beta t\ll 1$:

$$W(v_k,v_{k+1},\Delta t)=\frac{1}{\pi^{1/2}v_T}\exp\left[-\frac{(v_{k+1}-v_k)^2}{2\beta v_T^2\Delta t}\right]. \qquad (2.10)$$

The measure given by the probability of the transition (2.10) is known as the Wiener measure[13] and obviously determines the diffusion of the atoms in velocity space.

At $\beta t_{\text{eff}}\ll 1$, changes in the velocity as a result of this diffusion predominate over the regular velocity changes associated with viscous friction βv in Langevin's equation (2.3). Actually, let us estimate the mean square change in the velocity $\langle(v_k-v_{k+1})^2\rangle$ in the time t as a result of both effects. The change because of diffusion $\langle\Delta v^2\rangle_{\text{diff}}(t)$, which is obtained by direct calculation with the probability (2.10), yields

$$\Delta v_{\text{diff}}^2=\langle(v_k-v_{k+1})^2\rangle\propto\bar{v}^2\beta t, \qquad (2.11)$$

since the regular change (obtained by the method of expansion of the exponential $e^{-\beta t}$) gives

$$\Delta v_{\text{reg}}^2=(v_k-v_{k+1})_{\text{reg}}^2\propto\bar{v}^2(\beta t)^2. \qquad (2.12)$$

It follows from a comparison of (2.11) and (2.12) that

$$\Delta v_{\text{diff}}^2/\Delta v_{\text{reg}}^2\sim(\beta t)^{-1}\gg 1, \qquad (2.13)$$

which also justifies the use of the measure (2.10).

The result of Kac[13–15] is very important for our exposition; he expressed the continuous integral (2.5) with Wiener measure in terms of the fundamental solution of a differential equation of a parabolic type. That is, Kac showed that (the proof is contained in Ref. 13)

$$\left\langle\exp\left\{-\int_0^t V[x(\tau)]\,d\tau\right\}\right\rangle=\int_{-\infty}^{\infty}u(x,t)\,dx\quad(x(0)=0), \qquad (2.14)$$

here the function $u(x,t)$ is defined in the following way:

$$\frac{\partial u(x,t)}{\partial t}=\frac{1}{2}\frac{\partial^2 u}{\partial x^2}-V(x)u(x,t),$$
$$\lim_{x\to\pm\infty}u(x,t)=0,\quad\lim_{t\to 0}u(x,t)=\delta(x). \qquad (2.15)$$

Taking the Fourier transform

$$\psi(x)=\int_0^\infty u(x,t)e^{-pt}dt,$$

we obtain

$$\frac{1}{2}\frac{d^2\psi}{dx^2}-[p+Y(x)]\psi(x)=0,\quad\lim_{x\to\pm\infty}\psi(x)=0,\quad\psi'(-0)-\psi'(+0)=2. \qquad (2.16)$$

The information furnished on the continuous integral is sufficient for us to proceed to the solution of the stated problem.

Using (2.4), we express the contour $I(\omega)$ in the form

$$I(\omega)=\int_{-\infty}^{+\infty}\frac{dv_0\exp\{-(v_0/v_T)^2\}}{(\pi v_T^2)^{1/2}}\frac{\mathrm{Re}}{\pi}\int_0^\infty dt\left\langle\exp\left\{i\int_0^t\Omega[v(\tau)]\,d\tau\right\}\right\rangle,$$
$$\Omega(\tau)=\{[\Delta\omega-kv_0-kv(\tau)]^2+4G^2\}^{1/2}. \qquad (2.17)$$

Here the angular brackets denote the continuous integral for a given value of v_0 (the conditional mean value[8,13]).

We introduce the following notation:

$$A=\frac{k\bar{v}_T}{\beta},\quad\frac{\Delta\omega-kv_0}{k\bar{v}_T}=\zeta,\quad\frac{v}{v_T}=x,\quad x(0)=0,$$
$$\tilde{\omega}=\frac{\Delta\omega}{kv_T},\quad\frac{v_0}{v_T}=y,\quad\frac{2G}{kv_T}=a. \qquad (2.18)$$

The condition of the smallness of the frequency $\beta\ll kv_T$ is assumed below, that is, $A\gg 1$.

Using (2.18), we express (2.17) in the form

$$I(\tilde{\omega})=\frac{\mathrm{Re}}{\beta\pi^{1/2}}\int_{-\infty}^{\infty}d\zeta e^{-(\tilde{\omega}-\zeta)^2}\int_0^\infty dt\left\langle\exp\left\{iA\int_0^t[(x-\zeta)^2+a^2]^{1/2}d\tau\right\}\right\rangle. \qquad (2.19)$$

In accord with Kac [see formulas (2.14)–(2.16)]

$$\int_0^\infty dt\langle\ldots\rangle=\int_{-\infty}^{\infty}dx\psi(x), \qquad (2.20)$$

where $\psi(x)$ is determined by the solution of the equation

$$\psi''-2iA[(x-\zeta)^2+a^2]^{1/2}\psi=0,$$
$$\psi(x)\underset{x\to\pm\infty}{\to}0,\quad[\psi(0)]_+=\psi(-0)-\psi(+0)=0,\quad[\psi'(0)]_+=2. \qquad (2.21)$$

Equation (2.21) will be used in the following sections for the solution of the given problem. For elucidation of the physical meaning of the function $\psi(x)$, it is of interest to obtain an equation similar to (2.21) by using another formal procedure—the equation for the density matrix.

It should be noted that the equivalence of the results based on the Langevin equation and on the Fokker-Planck equation is well known (see Ref. 8). In the problem considered, however, this equivalence is traced only in the linear case ($G\to 0$).

In the linear case, the general solutions are unknown both in the amplitude approach (using the continuous integral) and in the Fokker-Planck approach, which is used in equations for the density matrix. Therefore, their comparison even for the particular nonlinear solution (2.21) is nontrivial.

§3. EQUATIONS FOR THE DENSITY MATRIX AND THE EQUIVALENCE OF THE AMPLITUDE AND MATRIX APPROACHES

Following Refs. 1–3, we write down the equation for the elements of the density matrix

$$\begin{aligned}\left(\frac{d}{dt}+\gamma_{11}\right)\rho_{11}&=S_1(\rho_{11})+2G\,\mathrm{Im}\,\rho_{12}+Q_1W_\mu(v),\\ \left(\frac{d}{dt}+\gamma_2\right)\rho_{22}&=S_2(\rho_{22})-2G\,\mathrm{Im}\,\rho_{12}+Q_2W_\mu(v),\\ \left(\frac{d}{dt}+\gamma\right)\rho_{12}&=S(\rho_{12})+iG(\rho_{11}-\rho_{22}).\end{aligned}\tag{3.1}$$

We limit ourselves to the case of identical relaxation constants $\gamma_1=\gamma_2=\gamma$ and identical collision integrals $S_1=S_2=S$. We shall seek a stationary and spatially homogeneous solution of the system (3.1). Introducing $N=\rho_{11}-\rho_{22}$, $\rho\equiv\rho_{12}$ and $Q=Q_1-Q_2$, we write the system (3.1) in the form

$$(\hat{s}-\gamma)N=-4G\,\mathrm{Im}\,\rho+QW_\mu,\qquad(\hat{s}-\gamma)\rho=iGN+i(kv-\Delta\omega)\rho.\tag{3.2}$$

In the case of weak fields ($G\to 0$), the solution of the first equation of the set (3.2) has the form

$$N\equiv N_\mu=Q\gamma^{-1}W_\mu(v).\tag{3.3}$$

We substitute the solution for arbitrary fields in the form $N=N_\mu+\tilde{N}$. The system (3.2) in this case is written in the form

$$(\hat{s}-\gamma)\tilde{N}=-4G\,\mathrm{Im}\,\rho,\qquad(\hat{s}-\gamma)\rho=iG\tilde{N}+i(kv-\Delta\omega)\rho+iGQ\gamma^{-1}W_\mu.\tag{3.4}$$

Let $A=\mathrm{Re}\,\rho, B=\mathrm{Im}\,\rho$. Then the set (3.2) can be written in matrix form ($\rho=\{A,B,N\}$)

$$(\hat{s}-\gamma)\hat{E}\rho+\hat{Z}\rho=Q.\tag{3.5}$$

Here $\hat{E}$ is the unit matrix and $Q=\{0,GQ\gamma^{-1}N_\mu(v),0\}$

$$\hat{Z}=\begin{pmatrix}0&-\delta&0\\ \delta&0&-G\\ 0&4G&0\end{pmatrix},\qquad\delta\equiv kv-\Delta\omega.\tag{3.6}$$

The eigenvalues of the matrix $\hat{Z}$ are $\lambda_{1,3}=\pm i\Omega$, $\lambda_2=0$, $\Omega=\{(kv-\Delta\omega)^2+4G^2\}^{1/2}$. We find the eigenvectors of the matrix $\hat{Z}$ and form from them the matrix $\hat{T}$ that diagonalizes the matrix $\hat{Z}$ ($\rho=\hat{T}\sigma$):

$$\hat{T}=\frac{1}{\Omega}\begin{pmatrix}-\delta&G&-\delta\\ \lambda_1&0&\lambda_3\\ 4G&\delta&4G\end{pmatrix}.\tag{3.7}$$

The physical meaning of introducing matrices σ and $\hat{T}$ is a transition to the states of the mixed system "atom + field." In the absence of relaxation and collisions, the states of this system are stationary and the absorption of light does not take place. Account of the Fokker-Planck collisions leads to the appearance of stationary absorption in the system.

The absorbed power is determined by the imaginary part of the polarization $\mathrm{Im}\,\rho=B$:

$$P(\omega)=2\hbar\omega GB.\tag{3.8}$$

It follows from (3.7) that the imaginary part of the polarization is expressed in terms of the first and second components of the vector σ:

$$B=i(\sigma_1-\sigma_3).\tag{3.9}$$

We write down the equation which is satisfied by the components of the mixed density matrix σ:

$$(\hat{T}^{-1}\hat{S}\hat{T}-\gamma+\Omega)\sigma=\hat{T}^{-1}Q.\tag{3.10}$$

In Eq. (3.10), we must carry out commutation of the operators $\hat{S}$ and $\hat{T}$, since the latter depends parametrically on the variable v on which the operator $\hat{S}$ acts. Direct calculation shows that the noncommuting terms can be discarded upon satisfaction of the condition[2)]

$$G\beta/\Omega_{\mathrm{eff}}^2\ll 1\tag{3.11}$$

and the equations for the components σ_1 and σ_2 of the matrix take the form ($\hat{S}$ is the diagonal operator of the collision integral)

$$\begin{aligned}(\hat{S}-\gamma+i\Omega)\sigma_1&=-\frac{i}{2}\frac{QG}{\gamma}W_\mu,\\ (\hat{S}-\gamma-i\Omega)\sigma_3&=\frac{i}{2}\frac{QG}{\gamma}W_\mu.\end{aligned}\tag{3.12}$$

It follows from (3.12) that we can take $\sigma_3=\sigma_1^*$. We set $\sigma=-i\eta/2$; then

$$B=\mathrm{Re}\,\eta,\tag{3.13}$$

while the equation for η has the form

$$(\hat{S}-\gamma+i\Omega)\eta=QG\gamma^{-1}W_\mu.\tag{3.14}$$

For the solution, it is convenient to introduce a delta function on the right in place of $QG\gamma^{-1}W_\mu$, i.e., to find the fundamental solution $\tilde{\eta}$ instead of η. Here the final expression for the absorbed power takes the form

$$P=2\hbar\omega G^2Q\gamma^{-1}\langle\mathrm{Re}\,\tilde{\eta}\rangle.\tag{3.15}$$

The angular brackets here denote averaging over the Maxwell distribution. If we introduce the value of the jump in the derivative, which is equal not to 1 but to 2, as in (3.15), then

$$P=\hbar\omega G^2Q\gamma^{-1}\langle\mathrm{Re}\,\tilde{\eta}'\rangle,\tag{3.16}$$

which, after setting $Q/\gamma=N$, is completely identical with the expression obtained in the amplitude approach.

Thus, the functions ψ in the amplitude approximation are set in correspondence with the matrix elements $\sigma_{1,3}$ of the density matrix of the mixed system "atom + field." Solutions will be given below of the corresponding equations and new nonlinear absorption effects will be investigated. We note that the total weak-collisions operator enters into (3.12), in contrast with (2.21). However, in actuality, we used, in the derivation of (3.12) the condition $\beta/kv\ll 1$, which corresponds to the transition in the collision integral to the diffusion limit, keeping only the second derivative d^2/dv^2 (cf. Sec. 2).

§4. SOLUTION OF THE EQUATIONS AND INVESTIGATION OF NONLINEAR EFFECTS IN ABSORPTION

We shall start out from Eq. (2.21). Substitution in it of $x_1=x-\xi$ yields

$$\psi''-2iA(x_1^2+a^2)^{1/2}\psi=0,\quad \psi\to 0,\quad x_1\to\pm\infty,\quad [\psi'(\xi)]_+^-=2;$$
$$I=\frac{\mathrm{Re}}{\beta\pi^{1/2}}\int_{-\infty}^{\infty}d\xi\exp\{-(\xi-\bar\omega)^2\}\int_{-\infty}^{\infty}dx_1\psi(x_1). \tag{4.1}$$

Further, we set $p(t)=(t^2+a^2)^{1/2}$, and define the function $p(t)$ in the plane with a cut from $t=-ia$ to $t=+ia$. On the real axis we have then $\mathrm{sign}\,p(t)=\mathrm{sign}\,t$. This choice of the branch of the analytic function guarantees a valid transition to the limit of weak fields ($a=0, p(t)=t$).

Equation (4.1) contains the large parameter $A\gg 1$, which allows us to use the quasiclassical method for its solution. We use here for the solution of Eq. (4.1) a method based on its reduction to the standard Airy equation (see Ref. 16), by introducing the auxiliary function $v(\eta)$:

$$\psi(x_1)=[\eta/p(x_1)]^{1/4}v(\eta),\quad \eta=\left\{\frac{3}{2}\int_0^{x_1}p^{1/2}(t)\,dt\right\}^{2/3}. \tag{4.2}$$

The equation for the function $v(\eta)$ is of the form

$$\frac{d^2v}{d\eta^2}-2iA\eta v=0,$$
$$[v(\eta_0)]_+^-=0,\quad \left[\frac{dv}{d\eta}\right]_+^-\Big|_{\eta=\eta_0}=2[\eta_0/p(\xi(\eta_0))]^{1/4},\quad v(\eta\to\pm\infty)\to 0, \tag{4.3}$$

where

$$\eta_0=\left\{\frac{3}{2}\int_0^{\xi}p^{1/2}(t)\,dt\right\}^{2/3}. \tag{4.4}$$

We introduce a new variable δ, for which Eq. (4.3) reduces directly to the Airy equation:

$$C\delta=\eta,\quad C\delta_0=\eta_0,\quad C\equiv e^{-i\pi/6}(2A)^{-1/3}\ll 1. \tag{4.5}$$

For the function $v(\delta)$ we get

$$\frac{d^2v}{d\delta^2}-\delta v=0,$$
$$[v(\delta_0)]_+^-=0,\quad \left[\frac{dv}{d\delta}\right]_+^-=2C^{3/4}[\delta_0/p(\xi(\delta_0))]^{1/4}. \tag{4.6}$$

Taking it into account that

$$d\eta_0/d\xi=\{p[\xi(\eta_0)]/\eta_0\}^{1/2},\quad d\eta/dx_1=\{p(x_1(\eta))/\eta\}^{1/2},$$

and substituting the function ψ in (4.1), we obtain for the contour $I(\omega)$

$$I=\frac{\mathrm{Re}}{\beta\pi^{1/2}}\int_{-\infty}^{\infty}d\eta_0\left(\frac{\eta_0}{p[\xi(\eta_0)]}\right)^{1/2}\exp\{-(\xi(\eta_0)-\bar\omega)^2\}\int_{-\infty}^{\infty}d\eta\left(\frac{\eta}{p[x_1(\eta)]}\right)^{1/2}v(\eta). \tag{4.7}$$

We construct the solution of Eq. (4.6) from two independent solutions $A^+(\delta)$ and $A^-(\delta)$ of Eq. (4.6). This new solution possesses the specified jump in the derivative. Then the expression (4.7) for the contour $I(\omega)$ takes in terms of the variables δ the form

$$I=\frac{2}{\pi^{1/2}kv_T}\mathrm{Re}\left\{e^{-i\pi/3}\int_{-\infty e^{i\pi/6}}^{+\infty e^{i\pi/6}}d\delta_0\left[\frac{C\delta_0}{p(\xi(C\delta_0))}\right]^{3/4}\exp\{-(\xi(C\delta_0)-\bar\omega)^2\}\right.$$
$$\times\left[A^+(\delta_0)\int_{-\infty e^{i\pi/6}}^{\delta_0}A^-(\delta)\left(\frac{C\delta}{p[x_1(C\delta)]}\right)^{3/4}d\delta\right.$$
$$\left.\left.+A^-(\delta_0)\int_{\delta_0}^{+\infty e^{i\pi/6}}A^+(\delta)\left(\frac{C\delta}{p[x_1(C\delta)]}\right)^{3/4}d\delta\right]\right\}. \tag{4.8}$$

As the functions $A^+(\delta)$ and $A^-(\delta)$, it is convenient to choose $\mathrm{Ai}(\delta)$ and $\mathrm{Ai}(\delta)+i\mathrm{Bi}(\delta)$, respectively, where Ai and Bi are the standard Airy functions.[16] Turning around the path of integration over δ and δ_0 in (4.8) to the real axis, we get

$$I=2(I_1+I_2)/\pi^{1/2}kv_T,$$
$$I_1=\mathrm{Re}\int_{-\infty}^{\infty}d\delta_0\varphi(\delta_0)\exp\{-[\xi(C\delta_0)-\bar\omega]^2\}\mathrm{Ai}(\delta_0)\int_{-\infty}^{\delta_0}d\delta\,\mathrm{Ai}(\delta)\varphi(\delta),$$
$$I_2=\mathrm{Im}\int_{-\infty}^{\infty}d\delta_0\varphi(\delta_0)\exp\{-[\xi(C\delta_0)-\bar\omega]^2\}\left\{\mathrm{Ai}(\delta_0)\int_{-\infty}^{\delta_0}d\delta\mathrm{Bi}(\delta)\varphi(\delta)\right. \tag{4.9}$$
$$\left.+\mathrm{Bi}(\delta_0)\int_{\delta_0}^{\infty}d\delta\,\mathrm{Ai}(\delta)\varphi(\delta)\right\},$$
$$\varphi(\delta)=(C\delta/p[x_1(C\delta)])^{1/4},\quad \varphi(\delta_0)=(C\delta_0/p[\xi(C\delta_0)])^{1/4}.$$

We define the distorting function $J(\omega, G)$:

$$I(G,\omega)=I(0,\omega)J(\omega,G),\quad I(0,\omega)=\pi^{-1/2}\exp(-\bar\omega^2). \tag{4.10}$$

Here the functions $J_{1,2}$ are determined by Eqs. (4.9) with the obvious substitution

$$\exp[-(\xi(C\delta_0)-\bar\omega)^2]\to\exp[2\bar\omega\xi(C\delta_0)-\xi^2(C\delta_0)].$$

Function $\xi(C\delta_0)$, is determined in accordance with Eq. (4.4) from the relation

$$\frac{2}{3}(C\delta_0)^{1/4}=\int_0^{\xi}(t^2+a^2)^{1/4}dt\quad (C\equiv(2A)^{-1/3}e^{-i\pi/6}). \tag{4.11}$$

The limiting values of the function ξ are the following:

$$\xi\approx\begin{cases}\dfrac{2}{3\sqrt{a}}(C\delta_0)^{3/4}, & |C\delta_0|\ll a\\ C\delta_0, & |C\delta_0|\gg a\end{cases} \tag{4.12}$$

The convergence of the integrals in (4.9) is determined, as is not difficult to show, by the Airy functions, so that effectively $\delta_{\mathrm{eff}}\sim 1$. Taking (4.12) into account, we find that $\xi_{\mathrm{eff}}\ll 1$. Therefore, under the condition

$$\bar\omega\xi_{\mathrm{eff}}\ll 1 \tag{4.13}$$

the exponentials in the distorting function $J(\omega, G)$ can be set equal to unity. Then the function $J(\bar\omega, G)=J(0, G)$ turns out to be independent of frequency. Thus, the deformation of the contour by the EM field turns out to be uniform over the spectrum in the most important range of frequencies $\bar\omega=\Delta\omega/kv_T\lesssim 1$, where the condition (4.13) is satisfied. We shall show that just this range of frequencies makes the fundamental contribution to the normalization of the spectrum, so that

$$\int_{-\infty}^{\infty}d\omega I(G,\omega)=J(0,G). \tag{4.14}$$

With account of the above, formula (4.10) for the distorting function $J(0, G)$ takes the form[3)]

$$J(0,G)=\mathrm{Re}\left[\int_{-\infty}^{\infty}d\delta\,\mathrm{Ai}(\delta)\varphi(\delta)\right]^2+2\,\mathrm{Im}\int_{-\infty}^{\infty}d\delta_0\varphi(\delta_0)\mathrm{Bi}(\delta_0)\int_{\delta_0}^{\infty}d\delta\varphi(\delta)\mathrm{Ai}(\delta). \tag{4.15}$$

It follows from the definition of the functions φ (4.9) and $p(\varphi)$ (4.1) that

$$\varphi(\delta)=[C\delta/[\xi^2(C\delta)+a^2]^{1/4}]^{3/4}. \tag{4.16}$$

With account of (4.12), the limiting expressions for $\varphi(\delta)$ are the following:

$$\varphi(\delta)\approx\begin{cases}(C\delta/a)^{3/4}, & C\delta\ll a\\ 1, & C\delta\gg a\end{cases}. \tag{4.17}$$

It is seen from (4.17) that the function $\varphi(\delta)$ which determines the distortions of the contour by the laser field

reduces to unity in weak fields ($a \to 0$), and to a small factor $(C\delta_{\text{eff}}/a)^{3/4}$ in strong fields. Assuming $\delta_{\text{eff}} \sim 1$, we see that the parameter of the transition from weak to strong fields is the critical value of the field G_{cr}, equal to

$$G_{\text{cr}} = kv_T|C| \approx kv_T(\beta/kv_T)^{1/3}. \tag{4.18}$$

It follows from formulas (4.15) and (4.16) that the field distortion of the contour is determined in order of magnitude by the square of the modulus the distorting function $|\varphi(\delta_{\text{eff}})|^2$ at the values $\delta_{\text{eff}} \sim 1$, which make the major contribution to the integral (4.15). Thus, at fields $G \gg G_{\text{cr}}$, the distorting factor turns out to be of the order of

$$J(0, G \gg G_{\text{cr}}) \sim (G_{\text{cr}}/G)^{1/2}. \tag{4.19}$$

§5. CONCLUSION

We pause to discuss the interrelation of the considered effect with the usual saturation, for example, in the presence of the quenching of an atomic oscillator characterized by the frequency γ. For this purpose we compare the time T_{ph} of loss of phase coherence by the oscillator (at a given value of the velocity v_0) as a consequence of both effects–phase diffusion and quenching. In the case of diffusion equating the phase shift to a quantity of the order of unity on the basis of the time evolution of the amplitudes in Sec. 2, or estimating the collision integral in Sec. 3, see also Sec. 1), we find

$$T_{\text{ph}}^{\text{diff}} \sim (k^2D)^{-1/3} \sim (kv_T)^{-2/3}\beta^{-1/3},$$

in the case of quenching obviously, $T_{\text{ph}}^{\text{qu}} \sim \gamma^{-1}$. Setting $T_{\text{ph}}^{\text{diff}} \ll T_{\text{ph}}^{\text{qu}}$, we obtain the condition

$$\gamma \ll kv_T(\beta/kv_T)^{1/3} \approx G_{\text{cr}},$$

which determines the region of manifestation of the effect considered above. Numerical estimates of the parameter β/kv_T for optical transitions in a gas of atoms or molecules with density $N \sim 10^{18}$ cm^{-3} and also for the plasma of an argon laser[9] ($N_i \sim 10^{14}$ cm^{-3}) lead to a quantity of the order of 10^{-2}–10^{-3}. Under these conditions, the considered nonlinear effects, which correspond to the condition $G \gtrsim G_{\text{cr}}$, for a typical value $\Delta\omega_D \sim 10^{10}$ s^{-1} manifest themselves at laser field intensities $E_0 \gtrsim 10^3$–10^4 V/cm.

In conclusion, we consider the case of the simultaneous action on the atom of constant (a) and Brownian accelerations in the linear approximation. Equation (3.1) describes in this case the ion in a buffer gas or in a plasma acted upon by a regular force (for example, an electric field).

For the population $N(v)$ we have

$$adN/dv - S_v(N) = 0, \tag{5.1}$$

where $S_v(N)$ is given as before in the model of weak collisions:

$$S_v = \beta \frac{d}{dv}\left(\frac{v_T^2}{2}\frac{d}{dv} + v\right).$$

It is obvious that by carrying out the change of variables $v = \tilde{v} + a/\beta$, we obtain in the new variables $\tilde{v}$ an equation that does not contain terms with acceleration:

$$S_{\tilde{v}}(N) = 0, \tag{5.2}$$

and has the obvious solution

$$N \propto W_\mu(\tilde{v}) = W_\mu(v + a/\beta). \tag{5.3}$$

Thus, the ionic component in the gas acquires a directed velocity $v = a/\beta$ in the direction of the acceleration. It is significant that this velocity depends on such characteristics of the ion as its mass and the collision frequency.

The equation for the polarization in (3.1), which determines the absorption of the EM field, reduces, through the obvious substitutions $v = \tilde{v} + a/\beta$, $\Delta\omega = \Delta\tilde{\omega} + ka/\beta$ to an equation without acceleration, but with the shifted frequency

$$\Gamma\rho + i(k\tilde{v} - \Delta\tilde{\omega})\rho = S_{\tilde{v}}(\rho) - iGNW_\mu(\tilde{v}). \tag{5.4}$$

If ions of different species are contained in the medium, then the frequency shift turns out to be dependent on the ion species.

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[1)] The Gaussian contour in (2.9) corresponds to not very high frequencies $\Delta\omega \sim kv_T$ (see Ref. 7).

[2)] The quantity Ω_{eff} is determined below from the solution of the corresponding equations. The criterion (3.11) here reduces to the condition $\beta/kv_v \ll 1$ (see the effective regions of change of the parameters $(\Delta\omega - kv_0)/kv_T \equiv \zeta$ and δ below).

[3)] The two terms entering into the integral I_2 in (4.9) are transformed into one by integration by parts.

[1] L. A. Vaĭnshteĭn, I. I. Sobel'man, and E. A. Yukov, Vozbuzhdenie atomov i ushirenie spektral'nykh linii (Excitation of Atoms and the Broadening of Spectral Lines) Moscow, Nauka, 1979.

[2] S. G. Rautian, G. I. Smirnov, and A. M. Shalagin, Nelinneĭnye rezonansy v spektrakh atomov i molekul (Nonlinear Resonances in the Spectra of Atoms and Molecules) Novosibirsk, Nauka, 1979.

[3] Yu. L. Klimontovich, Kineticheskaya teoriya elektromagnitnykh protsessov (Kinetic Theory of Electromagnetic Processes) Moscow, Nauka, 1980.

[4] V. A. Alekseev and A. V. Malyugin, Zh. Eksp. Teor. Fiz. **80**, 897 (1981) [Sov. Phys. JETP **53**, 456 (1981)].

[5] R. Feynman and A. R. Hibbs, Quantum Mechanics and Path Integrals, McGraw (1965).

[6] M. I. Podgoretskiĭ and A. V. Stepanov, Zh. Eksp. Teor. Fiz., **40**, 561 (1961) [Sov. Phys. JETP **13**, 393 (1961)].

[7] S. G. Rautian, Trudy FIAN **43**, 3 (1968).

[8] S. Chandrasekar, Stochastic Problems in Physics and Astronomy, Rev. Mod. Phys. **15**, 1–89 (1943).

[9] G. I. Smirnov and D. A. Shapiro, Zh. Eksp. Teor. Fiz. **76**, 2084 (1979) [Sov. Phys. JETP **49**, 1054 (1979)].

[10] A. F. Kofman and A. I. Burshteĭn, Zh. Eksp. Teor. Fiz. **76**, 2011 (1979) [Sov. Phys. JETP **49**, 1019 (1979)].

[11] L. A. Vaĭnshteĭn, L. P. Presnyakov, and I. I. Sobel'man, Zh. Eksp. Teor. Fiz. **43**, 518 (1962) [Sov. Phys. JETP **16**, 370 (1963)].

[12] V. S. Lisitsa, Zh. Eksp. Teor. Fiz. **69**, 195 (1975) [Sov. Phys. JETP **42**, 101 (1975)].

[13] M. Kac, Probability and Related Topics in Physical Sciences,

Interscience, N. Y. (1959).
[14]I. M. Gel'fand and A. M. Yaglom, Usp. Mat. Nauk **11**, 77 (1956).
[15]I. M. Koval'chik, Usp. Mat. Nauk **18**, 97 (1963).
[16]M. A. Abramowitz and I. A. Stegun, Handbook of Mathematical Functions, Dover (1964).

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