Feasibility of investigating atomic collisions by the photon-echo technique

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An integral of elastic depolarizing collisions is obtained with account taken of the dependence of the velocity vector on the velocity of the exciting atom. The calculations are valid for any collision model involving a short-range interaction potential between two atoms, one of which is in the excited state and the other in the ground state. The result is presented in a form which is convenient in spectroscopy problems. Graphs of the dependence of the relaxation parameters on the modulus of the velocity of the exciting atom are given for quasiresonant collisions. The feasibility of determining experimentally the parameters characterizing elastic atomic collisions by investigating the photon echo is demonstrated for the case of resonant atomic transitions with change of the total angular momentum $1\rightleftarrows 0$. The information obtained in this manner is much more complete than that obtained by the existing experimental methods.

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The photon-echo method has been more and more widely used in recent years in nonlinear laser spectroscopy.[1–10] Its essence is the successive passage through the medium of two short exciting light pulses, separated in time by an interval τ_s. The photon echo arises at an interval of time of approximately τ_s after the second exciting pulse. The relaxation time of the excited atomic states is determined by the attenuation of the intensity of the echo as a function of τ_s, and the polarization properties of the echo permit us to identify the resonant atomic transitioms.

In the treatment of the results of the first experiments in a gas,[2,3,5,6] a start was made from an exponential law of attenuation of the intensity of the photon echo $I(\tau_s)\propto \exp(-2\tau_s/t_r)$, where t_r is the sought relaxation time and is due to the radiative decay of the excited state of the atom and gas-kinetic collisions. However, spectroscopic investigations[11–13] indicate that the collision matrix $\hat{\Gamma}$ depends both on the modulus and on the direction of the velocity **v** of the atom. Therefore, at great gas pressure, the function $I(\tau_s)$ has a more complicated form.

In theoretical researches,[7,8] under certain simplifying assumptions, the decay law of the echo intensity $I(\tau_s)$ has been determined with account of the dependence of $\hat{\Gamma}$ on the modulus of the velocity v. In Ref. 14 it was established that atomic collisions, even without account of the dependence of $\hat{\Gamma}$ on v, affects the polarization echo in the majority of atomic transitions, with the exception of $0\rightleftarrows 1$, $\frac{1}{2}\to\frac{1}{2}$, $1\to 1$ and $\frac{1}{2}\rightleftarrows\frac{3}{2}$. In the present work, we have calculated to the end the integral of the depolarizing elastic collisions, in which, in contrast to the generalized accepted approach,[11–13] the dependence on the direction of the velocity **v** of the exciting atom is preserved. In this case, the collision matrix $\hat{\Gamma}$ becomes anisotropic in velocity space. The anisotropy mentioned leads to a different rate of relaxation of the individual components of the polarization vector **P** and, as a consequence, to the rotation of **P** under the action of the collisions, a situation reflected in the polarization of the photon echo and its profile. In particular, after passage of two exciting pulses, polarized in mutually perpendicular planes, the appearance of the photon echo is due exclusively to elastic collisions. Moreover, the dependence of $\hat{\Gamma}$ on the direction of **v** leads to a splitting of the degenerate level. These phenomena have been established for resonance atomic transitions with change in the total angular momentum $1\rightleftarrows 0$, for which it was shown[14] that the echo polarization is independent of the atomic collisions.

The validity of this or that model of elastic atomic collisions can be tested against the experimentally measured decay law of the intensity $I(\tau_s)$, the angle of rotation of the plane of polarization and the shift in the maximum of the photon echo profile. These data enable us also to determine all the relaxation parameters of the collision integral if they are sufficiently smooth functions of the velocity of the atom.

The found photon echo singularities due to the dependence of the collision matrix on the velocity vector of the atom will also be observed in other optically resolved atomic transitions. Therefore, the use of the photon echo for the experimental study of atomic collisions is promising.

1. THE COLLISION INTEGRAL

The elastic collision cross section of atoms, one of which is excited and the other in the ground state, is inversely proportional to their relative velocity raised to a fractional power and significantly exceeds in certain cases the usual gas-kinetic cross section.[15] Assuming this condition to be satisfied, we shall take into account the relaxation due to gas-kinetic inelastic collisions as well as radiative decay by means of the usual constants in the kinetic equation, while the intense relaxation due to elastic collisions will be taken into account in greater detail.

We consider below collisions of atoms between which there is a van der Waals or some other short-range interaction. The density of the excited working atoms (called simply atoms in what follows) is small in comparison with the density of unexcited atoms of the gas or of an impurity gas. In collisions of identical atoms, the van der Waals interaction exceeds the resonant dipole-dipole interaction.

For simplicity, we assume that the angular momenta of the considered excited states a and b of the atom are $j_a = 0$ and $j_b = 1$, and that the unexcited atoms are in the ground S state (the state a can also be unexcited). Following Refs. 16 and 17, we write down the collision integral that enters in the kinetic equation for the density matrix R_{m0} which describes transitions of the atom be tween states a and b[1)]:

$$\left(\frac{\partial}{\partial t}R_{m0}\right)_{\text{coll}}=-\frac{1}{2}(\gamma_a+\gamma_b)R_{m0}+\Gamma_{mm'}R_{m'0}; \tag{1}$$

Here $1/\gamma_a$ and $1/\gamma_b$ are the times of the excited-state relaxation due to gas-kinetic inelastic collisions and radiative decay. The indices m and m' take on the values 1, 0, and -1, labeling the state with a certain projection of the angular momentum.

The collision matrix $\Gamma_{mm'}$, which is nondiagonal in the states a and b, and which takes into account the relaxation of the polarization of the excited atom in elastic collisions with unexcited atoms, is the form

$$\Gamma_{mm'}=n_0\int(S_{mm'}-\delta_{mm'})\,|\mathbf{v}-\mathbf{v}_0|f(v_0)\,d\mathbf{v}_0\,d\rho. \tag{2}$$

Here $S_{mm'}$ is the matrix element of the S matrix of elastic scattering of an atom with angular momentum 1 by an unexcited atom, the atom velocities being $\mathbf{v}$ and $\mathbf{v}_0$. The quantity $d\rho$ denotes integration over the impact parameter ρ in a plane perpendicular to the relative velocity $\mathbf{v}-\mathbf{v}_0$. Further, n_0 is the density of unexcited atoms, which have a Maxwellian distribution in the velocities $\mathbf{v}_0$,

$$f(v_0)=\frac{1}{(\pi^{1/2}u_0)^3}\exp\left(-\frac{v_0^2}{u_0^2}\right),\quad u_0^2=\frac{2\varkappa T}{M_0},$$

where $\varkappa$ is the Boltzmann constant, T is the temperature, u_0 is the average thermal velocity, and M_0 is the mass of the unexcited atom. We have neglected in Eq. (2) the effect of elastic collisions on the wave function of the nondegenerate state a.

The density matrix ρ_a of an atom in the excited state a is subject to relaxation only because of inelastic collisions and radiative decay

$$(\partial\rho_a/\partial t)_{\text{coll}}=-\gamma_a\rho_a.$$

The collision integral $\Gamma_{mm'}^{nn'}$ in the kinetic equation for the density matrix $\rho_{mm'}$ of the atom in state b is determined in similar fashion:

$$\left(\frac{\partial}{\partial t}\rho_{mm'}\right)_{\text{coll}}=-\gamma_b\rho_{mm'}+\Gamma_{mm'}^{nn'}\rho_{nn'}, \tag{3}$$

where

$$\Gamma_{mm'}^{nn'}=n_0\int(S_{mn}S^*_{m'n'}-\delta_{mn}\delta_{m'n'})\,|\mathbf{v}-\mathbf{v}_0|f(v_0)\,d\mathbf{v}_0\,d\rho.$$

In the impact-parameter approximation the equation for the S matrix in the relative coordinates $\mathbf{R}$ takes the simple form[18]

$$i\hbar(\mathbf{v}-\mathbf{v}_0)\nabla S_{mm'}(\mathbf{R})=V_{mm''}S_{m''m'}(\mathbf{R}) \tag{4}$$

with the additional condition $S_{mm_0}(-\infty)=\delta_{mm_0}$, where the index m_0 characterizes the state of the atom before the collision. The matrix $V_{mm'}=V_{mm'}(\mathbf{R})$ of the interaction operator of the colliding atoms is determined by the model assumed for the elastic collisions.

It is convenient to solve Eq. (4) in a system of coordinates with the Z axis directed along the relative velocity $\mathbf{v}-\mathbf{v}_0$ and the X axis along the vector $\boldsymbol{\rho}$ (the axes Z and X lie in the plane of the collisions). Moreover, it is expedient to transform from matrix to tensor notation, in which the interaction operator of the atoms is real. Having determined the S matrix in the given coordinate system, it is not difficult to find this same quantity in an arbitrary set of coordinates if we use the transformation at finite rotation.[19] We give the final result for (2) in matrix notation:

$$\Gamma_{mm'}=-\Gamma_1\delta_{mm'}-(\Gamma_0-\Gamma_1)\frac{v_m^*v_{m'}}{v^2}, \tag{5}$$

$$\Gamma_1=\frac{n_0}{2}\int d\mathbf{v}_0\,d\rho\,f(v_0)\,|\mathbf{v}-\mathbf{v}_0|\,[2-\tilde{S}_{00}-\tilde{S}_{11}+(\tilde{S}_{00}-\tilde{S}_{11})\cos^2\theta_0] \tag{6a}$$

$$\Gamma=n_0\int d\mathbf{v}_0\,d\rho f(v_0)\,|\mathbf{v}-\mathbf{v}_0|\,[1-\tilde{S}_{11}-(\tilde{S}_{00}-\tilde{S}_{11})\cos^2\theta_0], \tag{6b}$$

$$v_{\pm1}=\mp\frac{v_x\pm iv_y}{\sqrt{2}},\quad v_0=v_z,\quad \cos\theta_0=\frac{\mathbf{v}(\mathbf{v}-\mathbf{v}_0)}{v|\mathbf{v}-\mathbf{v}_0|}. \tag{7}$$

The tilde labels matrix elements of the S matrix after the collision, calculated in the set of coordinates with the Z axis parallel to the vector $\mathbf{v}-\mathbf{v}_0$. We emphasize that in the derivation of (5) the explicit form of the interaction potential of the atoms is not employed.

The collision matrix (5) depends both on the modulus and the direction of the atom velocity $\mathbf{v}$. Yet the complex quantities $\Gamma_1=\Gamma_1'+i\Gamma_1''$ and $\Gamma_0=\Gamma_0'+i\Gamma_0''$ depend only on the modulus v, the prime and double prime denoting the real and imaginary parts.

The elastic collisions cause relaxation of the polarization vector $\mathbf{P}=\mathbf{d}_{0m}R_{m0}$ of the atoms moving with velocity $\mathbf{v}$. This relaxation is described by the equation

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)P_\alpha=\Gamma_{\alpha\beta}P_\beta+i\frac{E_a-E_b}{\hbar}P_\alpha, \tag{8}$$

$$\Gamma_{\alpha\beta}=-\Gamma_1(\delta_{\alpha\beta}-v_\alpha v_\beta/v^2)-\Gamma_0 v_\alpha v_\beta/v^2, \tag{9}$$

where E_a and E_b are the energies of the atom in the states a and b, $\mathbf{d}_{0m}$ is the dipole moment of the considered atomic transition, and $\Gamma_{\alpha\beta}$ is the collision matrix (5) in tensor notation. The Greek subscripts take on three values, denoting the projections on the Cartesian axes.

It is seen that $1/\Gamma_1'$ and $1/\Gamma_0'$ are the relaxation times of the transverse and longitudinal (relative to $\mathbf{v}$) components of the polarization vector, and $\hbar\Gamma_1''$ and $\hbar\Gamma_0''$ are connected with the shifts of the sublevels of the split level of the atom with $j_b=1$.

In order to understand the singularities of the behavior of the quantities $\Gamma_1=\Gamma_1(v)$ and $\Gamma_0=\Gamma_0(v)$ as functions of the modulus of the velocity v, we consider quasiresonant collisions, in which the energy E_k of one of the intermediate states is very close to the energy E_0 of the system of two colliding atoms $|E_0 - E_k| = \xi \ll E_0$. Then, of the entire sum over the intermediate states in the expression $V_{mm'}$ for the van der Waals interaction potential, there remains only the single quasi-resonant component

$$V_{mm'} = -Q(R^2\delta_{mm'} + 3R_m^* R_{m'})/R^8,$$

where $Q = |d_b|^2 |d_0|^2/9\hbar\xi$, and d_b and d_0 are the reduced dipole moments of the optically resolved transitions of the excited and ground states of the atoms.

For quasi-resonant collisions, the S matrix was found in Ref. 15, and made it possible, with the help of a high-speed computer, to plot the quantities (6a) and (6b) against the modulus v of the velocity of the atom (Figs. 1 and 2). It is seen that there exists a region[2] $\alpha^2 \equiv M_0/M_1 \gg 1$ where the dependence of the quantities Γ_1 and Γ_0 on v is significant, while at $\alpha^2 \ll 1$ these quantities are almost constant in the neighborhood of the mean thermal velocity $u = (2\varkappa T/M_1)^{1/2}$. Here M_1 is the mass of the excited atom.

2. BASIC EQUATIONS

We consider the formation of the photon echo in a gas following the action of two ultrashort exciting pulses with electric field intensities

$$\mathbf{E}_1 = \mathbf{e}_1 \exp[i(\omega t - kz + \Phi_1)] \quad \text{at} \quad 0 \leq t - z/c \leq T_1, \tag{10}$$

$$\mathbf{E}_2 = \mathbf{e}_2 \exp[i(\omega t - kz + \Phi_2)] \quad \text{at} \quad \tau_s + T_1 \leq t - z/c \leq \tau_s + T_1 + T_2. \tag{11}$$

The amplitudes $\mathbf{e}_1$ and $\mathbf{e}_2$ as well as the phase shifts Φ_1 and Φ_2 are constant, and the carrier frequency ω is at resonance with the frequency $\omega_0 = (E_a - E_b)/\hbar > 0$ of the atomic transition with change in the total angular momentum $j_a = 0 \to j_b = 1$. The durations T_1 and T_2 of the pulses are short in comparison with the time τ_s between them and the times of irreversible relaxations. The term z/c takes into account the delay of the electromagnetic signal propagating along the Z axis with the velocity c.

For the determination of the intensity E of the electric field of the photon echo, we use the d'Alembert equation

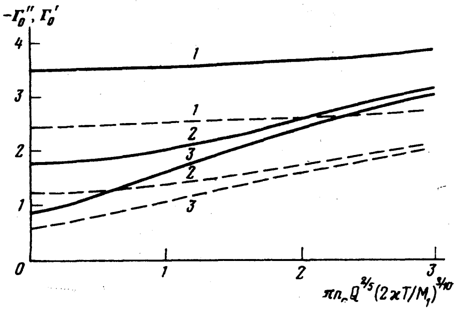

FIG. 1. Values of Γ_0' (continuous curves) and Γ_0'' (dashed curves) (Γ_0' and Γ_0'' are in units of u) at the values $\alpha^2 = 0.1$, 1 and 10—curves 1, 2, 3, respectively.

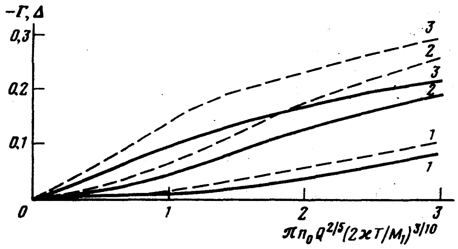

FIG. 2. The values of Γ (continuous curves) and Δ (dashed curves) (Γ and Δ are in units of u) at α^2 equal to 0.1, 1 and 10—curves 1, 2, 3, respectively.

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{E} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\int \mathbf{P}\, d\mathbf{v} \tag{12}$$

and the quantum-mechanical equation for the density matrix

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right)\hat{\rho} = \frac{i}{\hbar}(\hat{\rho}H - H\hat{\rho}) + \left(\frac{\partial \ddot{\rho}}{\partial t}\right), \tag{13}$$

in which H is the Hamiltonian of an atom located in the electromagnetic field, and the last term on the right side of (13) takes into account the collision relaxation according to (1)–(3), and also the radiative transition to the lower resonance level, due to spontaneous radiation at the upper level.

We separate in the sought functions the rapidly oscillating factors

$$\mathbf{E} = \mathbf{e} \exp[i(\omega t - kz + \Phi)],$$

$$\mathbf{P} = \mathbf{p} \exp[i(\omega t - kz + \Phi)],$$

where $\mathbf{e}$ and $\mathbf{p}$ are slowly varying amplitudes and Φ is a constant phase shift. In the resonance approximation, the equations for the slow functions follow from (12) and (13):

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\mathbf{e} = -2i\pi\omega \int \mathbf{p}\, d\mathbf{v}, \tag{14}$$

$$\left[\frac{\partial}{\partial t} + i(\Delta\omega - kv_z) + \gamma\right]p_\alpha - \Gamma_{\alpha\beta}p_\beta = \frac{i|d|^2}{\hbar}\left(\frac{1}{3}\rho_a e_\alpha - e_\beta \rho_{\beta\alpha}\right), \tag{15}$$

$$\left(\frac{\partial}{\partial t} + \gamma_a\right)\rho_a = \frac{i|d|^2}{\hbar}(p_\alpha e_\alpha^* - p_\alpha^* e_\alpha), \tag{16}$$

$$\left(\frac{\partial}{\partial t} + \gamma_b\right)\rho_{\alpha\beta} - \Gamma_{\alpha\beta}^{\sigma\delta}\rho_{\sigma\delta} - \frac{4|d|^2\omega_0^3}{9\hbar c^3}\delta_{\alpha\beta}\rho_a = \frac{i|d|^2}{3\hbar}(p_\alpha^* e_\beta - e_\alpha^* p_\beta), \tag{17}$$

where we have introduced the notation

$$p_\alpha = d_{0m}{}^\alpha R_{m0} \exp[i(kz - \omega t - \Phi)], \quad \Delta\omega = \omega - \omega_0,$$

$$\gamma = (\gamma_a + \gamma_b)/2, \quad \rho_{\alpha\beta} = d_{0m}{}^\beta \rho_{mm'} d_{m'0}^\alpha/|d|^2,$$

$$\Gamma_{\alpha\beta}^{\sigma\delta} = 9 d_{0m}{}^\beta d_{m'0}^\alpha \Gamma_{mm'}^{nn'} d_{n0}{}^\delta d_{0n'}{}^\sigma/|d|^4.$$

Here d is the reduced dipole moment of the atomic transition $j_a = 0 \to j_b = 1$, and the other quantities are as defined above. The last term on the left side of (17) is due to the spontaneous radiation of the atom at the upper level, which is accompanied by transition to the state b.

At the initial instant of time $t - z/c = 0$, the polarization vector $\mathbf{p}$ is equal to zero and the density matrices $\rho_a = \rho_a(\mathbf{v}, t - z/c)$ and $\rho_{\alpha\beta} = \rho_{\alpha\beta}(\mathbf{v}, t - z/c)$ take the form

$$\rho_a(\mathbf{v}, 0) = n_a f(v), \quad \rho_{\alpha\beta}(\mathbf{v}, 0) = n_b f(v)\delta_{\alpha\beta}/9,$$

where n_a and n_b are the densities of the atoms at the upper and lower levels at $t - z/c = 0$. According to the assumed normalization, the quantities **p**, ρ_α and $\rho_{\alpha\beta}$ refer to a set of atoms moving with velocity **v**.

If we neglect the reaction of the medium on the exciting pulses, then Eqs. (14)–(17) become linear and it becomes possible to determine the photon echo in the given-field approximation (10) and (11). For this purpose, the conditions

$$4\omega|d|^2L|N_0|T_i/9\hbar c \ll 1 \quad \text{at} \quad T_0 \gg T_i,$$
$$4|d|^2L|N_0|/9\hbar u \ll 1 \quad \text{at} \quad T_0 \ll T_i,$$

must be satisfied, where $i = 1$ or 2, $N_0 = n_a - n_b/3$ is the repopulation density of the levels, L is the dimension of the gaseous medium, and the time T_0 of the reversible Doppler relaxation is expressed in terms of the mean thermal velocity u of the atoms of the gas, $T_0 = 1/\text{ku}$.

3. PHOTON ECHO ON A NARROW SPECTRAL LINE

We first consider the case of a narrow spectral line ($1/T_0 \ll 1/T_i$, $i = 1, 2$), assuming the pulses (10) and (11) to be linearly polarized; $\mathbf{e}_1 = e_1\mathbf{l}_1$ and $\mathbf{e}_2 = e_2\mathbf{l}_2$. Since the lengths of the exciting pulses are small in comparison with the inverse relaxation time, the effect of collisions in time intervals on the order of T_1 and T_2 will be neglected. Then the solution of Eqs. (14)–(17) without account of collisions in the region $0 \leq t - z/c \leq T_1$ is the same as in the previous work.[4]

After passage of the first exciting pulse $T_1 \leq t - z/c$, the polarization vector **P** of the set of atoms moving with velocity **v** is different from zero at each point z of the gas medium

$$\begin{gathered} \mathbf{P}(t-T_1-z/c) = p_0\mathbf{L}(t-T_1-z/c) \\ \times \exp[i(\omega+kv_z)(t-T_1-z/c)+i(\omega T_1+\Phi_1)], \\ p_0 = i|d|^2 e_1 N_0 f(v)\sin\Omega_1 T_1/3\hbar\Omega_1, \\ \Omega_1^2 = 4|d|^2 e_1^2/3\hbar^2. \end{gathered} \tag{18}$$

Information on the elastic collisions is contained in the complex vector $\mathbf{L}(t)$, which has the following components:

$$L_x(t) = h(t)\cos\psi - g(t)\cos(2\varphi+\psi),$$
$$L_y(t) = h(t)\sin\psi + g(t)\sin(2\varphi+\psi).$$

Here ψ is the angle between the vector $\mathbf{l}_1$ and the X axis, and the functions $h(t)$ and $g(t)$ are damped in time as a consequence of the collisional relaxation and radiative decay:

$$h(t) = \tfrac{1}{2}[(1+\cos^2\theta)e^{-\Gamma_1 t} + \sin^2\theta e^{-\Gamma_0 t}]e^{-\gamma t},$$
$$g(t) = \tfrac{1}{2}\sin^2\theta(e^{-\Gamma_1 t} - e^{-\Gamma_0 t})e^{-\gamma t}.$$

The quantity $\mathbf{L}(t)$ depends essentially on the polar (θ) and azimuthal (φ) angles of the vector **v**, i.e., on the mutual orientation of the vectors v, k and $\mathbf{l}_1$, where the polar axis Z is chosen along the wave vector **k**. Because of the presence of preferred directions of **k** and $\mathbf{l}_1$, the relaxation of the polarization vector (18) follows a more complicated law in comparison with the solution of Eq. (8).

The physically observed polarization vector $\int \mathbf{P}d\mathbf{v}$ is rapidly attenuated in time:

$$\int \mathbf{P}\,d\mathbf{v} \sim \mathbf{l}_1 \exp\left[-\frac{(t-T_1-z/c)^2}{4T_0^2}\right] \tag{19}$$

as a consequence of the Doppler dephasing of its component parts, which have different values of the velocity **v**. However, the set of atoms moving with velocity **v** and making a contribution to (18), preserves the "memory" of the first exciting pulse during a time interval

$$T_1 \leq t - z/c \leq t_r, \tag{20}$$

where t_r is the time of inverse relaxation, identical with the smallest of the quantities $1/\Gamma_1$, $1/\Gamma_0$ and $1/\gamma$ ($T_0 \ll t_r$). The collisional relaxation obliterates this memory, and the degree of memory destruction is significantly reflected in the polarization and the profile of the photon echo. This serves as the source of information on the atomic collisions.

During the time

$$\tau_s + T_1 \leq t - z/c \leq \tau_s + T_1 + T_2$$

a second exciting pulse (11) passes through the medium, and its vector $\mathbf{l}_2$ on entering the medium is parallel to the X axis and makes an angle ψ with the vector $\mathbf{l}_1$. The linearized equations (14)–(17) are easily solved in this case, too, with account taken of the initial condition that follows from (18) at $t = \tau_s + T_1$. At the time $t = \tau_s + T_1 + T_2 + z/c$, when the second pulse (11) leaves the point z of the gaseous medium, the polarization vector which makes a contribution to the echo takes on the value

$$\begin{gathered} \mathbf{P}(T_2) = \tfrac{1}{2}\mathbf{l}_2 p_0^* L_x^*(\tau_s)(1-\cos\Omega_2 T_2) \\ \times \exp\{i[\omega(\tau_s+T_1+T_2)+(\Delta\omega-kv_z)\tau_s+2\Phi_2-\Phi_1]\}, \\ \Omega_2^2 = 4|d|^2 e_2^2/3\hbar^2. \end{gathered} \tag{21}$$

The quantity (21) serves as the initial condition for the solution of Eqs. (14)–(17) in the region $\tau_s + T_1 + T_2 \leq t - z/c$ after passage of the second exciting pulse, when the relaxation phenomena again affect the formation of the echo.

According to (18), (19), and (20), after passage of the first exciting pulse the macroscopic electromagnetic field decays rapidly after an interval of time T_0, as a consequence of dephasing and losses of coherence of the individual radiators. Therefore, in the region $T_0 \leq t - z/c$, the excited atoms radiate independently, and their lifetime is determined by the radiative decay and the atomic collisions. The second exciting pulse leads to the result that, at the instant of time $t = 2\tau_s + T_1 + T_2 + z/c$, synchronization of the radiators with different velocities **v** takes place and the set of excited atoms undergoes a transition to the superconducting state, which is rapidly destroyed with emission of a photon echo of an intensity proportional to the square of the density of the excited atoms.

Omitting the intermediate calculations, we write out the final equation for the intensity of the electric field of the photon echo:

$$\mathbf{E}^e=\mathbf{e}^e\frac{\pi|d|\omega LN_0}{4\sqrt{3}c}\sin\Omega_1T_1(\cos\Omega_2T_2-1)$$
$$\times\exp\{i[\omega_0t-kz+2\Phi_2-\Phi_1+\Delta\omega(2\tau_s+T_1+T_2)]\}, \tag{22}$$

$$e_x^e=\cos\psi\int d\mathbf{v}f(v)[2h^*(\tau_s)h(t-\tau_s-T_1-T_2-z/c)$$
$$+g^*(\tau_s)g(t-\tau_s-T_1-T_2-z/c)]\exp[ikv_z(t-2\tau_s-T_1-T_2-z/c)]. \tag{23}$$

$$e_y^e=\sin\psi\int d\mathbf{v}f(v)g^*(\tau_s)g(t-\tau_s-T_1-T_2-z/c)$$
$$\times\exp[ikv_z(t-2\tau_s-T_1-T_2-z/c)]. \tag{24}$$

In the case of a narrow spectral line, the duration of the signal echo (22) is equal to the time T_0 of Doppler relaxation, i.e., it exceeds T_1 and T_2. The photon echo is elliptically polarized and propagates with a carrier frequency ω_0. The elastic collisions affect the rotation of the axes of the polarization ellipse and the deformation of its shape. At $\psi=\pi/2$, the polarization of the echo becomes linear, coinciding with the polarization of the exciting pulse. At the same time, in the absence of elastic depolarizing collisions, the polarization of the echo is always identical with the linear polarization of the second exciting pulse, and at $\psi=\pi/2$ there is no echo at all.[4]

The elastic collisions shift additionally the moment of time t_0 at which the maximum of the echo amplitude is reached, and the shift $\varepsilon=t_e-2\tau_s-T_1-T_2$ for the amplitudes (23) and (24) is equal to

$$\varepsilon^x=\frac{d}{d\tau_s}\int[2|h(\tau_s)|^2+|g(\tau_s)|^2]f(v)d\mathbf{v}$$
$$\times\left\{2\int(\mathbf{kv})^2[2|h(\tau_s)|^2+|g(\tau_s)|^2]f(v)d\mathbf{v}\right\}^{-1}, \tag{25}$$
$$\varepsilon^y=\frac{d}{d\tau_s}\int|g(\tau_s)|^2f(v)d\mathbf{v}\left\{2\int(\mathbf{kv})^2|g(\tau_s)|^2f(v)d\mathbf{v}\right\}^{-1},$$

respectively, where we have assumed $\varepsilon^x/T_0\ll1$ and $\varepsilon^y/T_0\ll1$. If these inequalities are not satisfied, then ε^x and ε^y are obtained by solution of transcendental equations.

The quantities (23) and (24) at the instant of the maximum are

$$e_x^e=\frac{8\pi\cos\psi}{15}\int_0^\infty\exp\{-2(\gamma+\Gamma_1')\tau_s\}(8+3e^{-2\Gamma\tau_s}+4e^{-\Gamma\tau_s}\cos\Delta\tau_s)v^2f(v)dv, \tag{26}$$

$$e_y^e=\frac{8\pi\sin\pi}{15}\int_0^\infty\exp\{-2(\gamma+\Gamma_1')\tau_s\}(1+e^{-2\Gamma\tau_s}-2e^{-\Gamma\tau_s}\cos\Delta\tau_s)v^2f(v)dv, \tag{27}$$

where $\Gamma=\Gamma_0'-\Gamma_1'$ and $\Delta=\Gamma_0''-\Gamma_1''$.

In contrast to the collisionless photon echo,[4] the plane of polarization of the echo (26) and (27) at the instant of achieving maximum is not identical with the polarization of the second exciting pulse, but lies between the vectors $\mathbf{l}_1$ and $\mathbf{l}_2$, making the angle θ_e with the vector $\mathbf{l}_2$, and $\tan\theta_e=e_y^e/e_x^e$.

By measuring the quantities θ_e, ε^x, ε^y and the intensity of the echo as a function of the parameter τ_s we can verify the validity of any particular atomic-collision model that leads to the concrete expressions (6a) and (6b) for $\Gamma_1=\Gamma_1(v)$ and $\Gamma_0=\Gamma_0(v)$. In Figs. 1 and 2, this corresponds to the region $\alpha^2\gg1$.

The results are especially interesting in those cases in which Γ_1 and Γ_0 are functions of v that are quite smooth in comparison with $v^2f(v)$ in the vicinity of the mean thermal velocity of the atoms. For quasiresonant collisions, this corresponds to the inequality $\alpha^2\ll1$ (see Figs. 1 and 2). Then the quantities Γ_1 and Γ_0 in formulas (22)–(27) can be assumed to be constants: $\Gamma_1\approx\Gamma_1(u)$ and $\Gamma_0\approx\Gamma_0(u)$. If we also take into account the fact that usually $|\Gamma|\ll\gamma+\Gamma_1'$ and $|\Delta|\ll\gamma+\Gamma_1'$, then the obtained expressions are simplitifed:

$$e_x^e=2\cos\psi\exp[-2(\gamma+\Gamma_1')\tau_s], \tag{28}$$
$$e_y^e={}^2/_{15}\sin\psi(\Gamma^2+\Delta^2)\tau_s^2\exp[-2(\gamma+\Gamma_1')\tau_s], \tag{29}$$
$$\varepsilon^x=-2T_0^2\{\gamma+\Gamma_1'+{}^1/_3\Gamma[1-{}^4/_5(\gamma+\Gamma_1')\tau_s]\}\quad\text{at}\quad(\gamma+\Gamma_1')^2T_0\ll\Gamma, \tag{30}$$
$$\varepsilon^y={}^{14}/_3T_0^2(1/\tau_s-\gamma-\Gamma_1')\quad\text{at}\quad T_0\ll\tau_s, \tag{31}$$
$$15\,\mathrm{tg}\,\theta_e=(\Gamma^2+\Delta^2)\tau_s^2\,\mathrm{tg}\,\psi. \tag{32}$$

From the experimentally measured laws (28) and (29) of the decay of the echo amplitudes as a function of τ_s, and also from the shifts (30) and (31) of the maxima of these amplitudes, it is not difficult to calculate Γ, Δ and $\gamma+\Gamma_1'$ separately. For the calculation of the quantity $\Gamma^2+\Delta^2$, we can use Eq. (32) independently for the different values of θ_e, ψ and τ_s. If the gas-kinetic constant γ is known or determined from another experiment, then the photon-echo method makes it possible to calculate the quantities Γ_1', Γ and Δ from the experimental data, which in turn allows us to determine the fundamental parameters of the elastic collisions $\Gamma_1=\Gamma_1'+i\Gamma_1''$ and $\Gamma_0=\Gamma_0'+i\Gamma_0''$.

4. CASE OF A BROAD SPECTRAL LINE

In most of the experiments that have been carried out, the photon echo in the gas was formed on a broad spectral line ($1/T_0\gg1/T_i$, $i=1,2$). The method set forth above of solution of the problem is also applicable in this case. However, the formulas become very complicated. The electric field intensity of the photon echo on a broad spectral line is given by

$$\mathbf{E}^e=\mathbf{e}^e\frac{\pi|d|\omega LN_0}{4\sqrt{3}c}\exp[i(\omega t-kz+2\Phi_2-\Phi_1)], \tag{33}$$

where the following notation has been introduced:

$$e_x^e=\cos\psi\int d\mathbf{v}F(\mathbf{v})[2h^*(\tau_s)h(t-\tau_s-T_1-T_2-z/c)$$
$$+g^*(\tau_s)g(t-\tau_s-T_1-T_2-z/c)], \tag{34}$$

$$e_y^e=\sin\psi\int d\mathbf{v}F(\mathbf{v})g^*(\tau_s)g(t-\tau_s-T_1-T_2-z/c), \tag{35}$$

$$F(\mathbf{v})=\frac{8|d|^3e_1e_2^2}{3\sqrt{3}\hbar^3\Omega_1\Omega_2^2}f(v)(1-\cos\Omega_2T_2)\left[i\frac{kv_z-\Delta\omega}{\Omega_1}(1-\cos\Omega_1T_1)-\sin\Omega_1T_1\right]$$
$$\times\exp[i(kv_z-\Delta\omega)(t-2\tau_s-T_1-T_2-z/c)],$$
$$\Omega_n=[(kv_z-\Delta\omega)^2+4|d|^2e_n^2/3\hbar^2]^{1/2},\quad n=1,2.$$

The integral $\int F(\mathbf{v})d\mathbf{v}$ describes the profile of the photon echo amplitude in the absence of collisions.[9,10]

The treatment of the experimental results with the aim of determination of $\Gamma_1(v)$ and $\Gamma_0(v)$ is possible here only by numerical methods. However, in the particular case of strict resonance $\Delta\omega=0$, under the condition that $\Gamma_1(v)$ and $\Gamma_0(v)$ are smooth functions of v in the neighborhood of the mean thermal velocity $\Gamma_1\approx\Gamma_1(u)$ and $\Gamma_0\approx\Gamma_0(u)$, the amplitudes (34) and (35) take the simple form

$$e_x^e={}^1/_4\cos\psi(3+3e^{-2\Gamma\tau_s}+2e^{-\Gamma\tau_s}\cos\Delta\tau_s)\exp[-2(\gamma+\Gamma_1')\tau_s]\int F(\mathrm{v})d\mathrm{v}, \quad (36)$$

$$e_y^e={}^1/_4\sin\psi(1+e^{-2\Gamma\tau_s}-2e^{-\Gamma\tau_s}\cos\Delta\tau_s)\exp[-2(\gamma+\Gamma_1')\tau_s]\int F(\mathrm{v})d\mathrm{v}. \quad (37)$$

Use has been made here of the fact that $F(v)$ is a very sharp function of the variable v_z near the value $v_z=\Delta\omega/k=0$,[9] if $1/T_0\gg1/T_i$, $i=1, 2$.

The duration of the echo signal (33) on the broad spectral line is approximately the same as the duration of the longer exciting pulse and the experimentally observed quantity $\tan\theta_e=e_y^e/e_x^e$ is connected with Γ and Δ in simple fashion.

Calculation of the relaxation parameters $\Gamma_1=\Gamma_1'+i\Gamma_1''$ and $\Gamma_0=\Gamma_0'+i\Gamma_0''$ according to (36) and (37) is carried out as in the case of the narrow spectral line. The final results are valid also for the atomic transition $j_b=1\to j_a=0$ $(E_b>E_a)$.

If the exciting pulses are circularly polarized then the photon echo on the atomic transitions $1\rightleftharpoons0$ arises only for identical rotation of the vectors of the electric field of these pulses, regardless of the intensity of the depolarizing collisions.

1)Repeated matrix and tensor indices imply summation.

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