

# Coherent emission by atoms induced by a traveling or standing wave

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We consider induced coherent emission in the form of a forward and a backward echo, which appear in a gas following the action of two ultrashort exciting pulses of a traveling and a standing wave separated by a time interval  $\tau$  and linearly polarized in planes with angle  $\Psi$  between them. The polarization properties and the damping laws of the direct and backward echo are determined as functions of the delay time  $\tau$ , the angle  $\Psi$ , the type of the resonant transition, and the relaxation. It is shown that a characteristic narrow resonance of width  $1/2\tau$  can be obtained by adding the waves of the forward and backward echoes. By using the general formulas and the experimental results on the behavior of the forward and backward echoes, it is possible to determine the shift and the broadening of the spectral line with account taken of the elastic atomic collisions, to identify the atomic and molecular transitions, as well as to study the characteristics of the depolarizing atomic collisions for any degree of degeneracy of the resonant energy levels.

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In the investigation of the optical characteristics of atomic transitions and of the relaxation of quantum states, extensive use is made of the photon echo produced in a resonant medium by the action of two ultrashort light-wave pulses separated by a time interval  $\tau$  (see, e.g., the reviews by Alekseev and Evseev<sup>1</sup> and by Samartsev<sup>2</sup> and the book by Allen and Eberly<sup>3</sup>). Depending on the exciting pulses used, the echo phenomenon evolves differently. The exhaustive theoretical and experimental investigations performed to date dealt with cases in which both exciting pulses were traveling<sup>1–3</sup> or standing<sup>4–7</sup> waves. The possibility of producing echo phenomena in nondegenerate systems by using a traveling-wave pulse and a standing-wave pulse were discussed by Shiren,<sup>8</sup> but his results and the method he used to solve the equations will not do in the case of standing waves in gaseous media. Photon echo induced by pulses of a standing wave and a traveling wave were recently observed in ruby<sup>9</sup> and indicate that this pulse sequence offers certain advantages over those in Refs. 1–7.

We investigate here theoretically, for the first time ever, photon echo produced in a gas by a sequence of a traveling wave and standing wave, with an arbitrary angle  $\psi$  between their polarization planes. The investigation is carried out in general form, with account taken of the thermal motion of the atoms, of the level degeneracy, of radiative decays, and of elastic depolarizing atomic collisions. The final formulas are valid for any level-degeneracy multiplicity and for an arbitrary width ratio of the spectral line and of the frequency spectrum of the exciting pulses. In contrast to the previously investigated photon echoes,<sup>1–7</sup> the coherent radiation by atoms following the action of a traveling- and standing-wave pulse sequence constitutes a superposition of two oppositely directed traveling waves with different properties. Coherent radiation in the direction of the exciting traveling-wave pulse (forward echo) is similar in many respects to the photon echo produced by traveling waves.<sup>1–3</sup> The oppositely directed coherent radiation (backward echo), however, has properties that differ substantially from those reported in Refs. 1–3. For example, at  $\psi = \pi/2$  there is no forward echo on atomic

transitions with total angular momentum change  $l \neq 0$  and  $l \neq 1$ , while the backward echo is linearly polarized in the plane of the exciting traveling-wave pulse. The forward and backward echoes depend on the parameters of the resonant atomic transitions, on the angle  $\psi$ , and on the relaxation processes. This makes it possible to identify the atomic transitions, to determine the shift and broadening of the spectral line, as well as to study the characteristics of other atomic collisions. In particular, for the  $\frac{1}{2} \rightarrow \frac{1}{2}$  atomic transition, the obtained distinguishing features of the coherent emission by the atoms agree, under certain conditions, with experiment<sup>9</sup>.

As indicated in Refs. 4–7, the intensity of the photon echo produced by two pulses of standing waves of frequency  $\omega$  contains a characteristic narrow resonance of width  $\frac{1}{2}\tau$  which appears when the frequency  $\omega$  is scanned. No such resonance appears in the forward and backward echoes if the atoms are excited by the traveling + standing wave pulse sequence. This narrow resonance can be obtained in experiment, however, if the light waves of the forward and backward echoes are combined by reflecting mirrors and made to propagate in the same direction. The narrow resonance obtained in this manner makes it possible to determine the collisional shift of the spectral line.

The obtained properties of the coherent radiation of the atoms following the action of the traveling and of the standing waves can stimulate the undertaking of new experiments aimed at the study of quantum transitions and atomic collisions in a gas, all the more since analogous properties will be possessed also by stationary coherent radiation induced in a gas by the action of a traveling and standing monochromatic waves separated by a certain distance in space (the method of separated fields in optics<sup>4</sup>).

## 1. CALCULATION METHOD AND BASIC RELATIONS

Qualitatively new effects, compared with the known cases<sup>1–7</sup> arise when the resonant gas medium is successively excited by pulses of a traveling and a standing

wave:

$$\mathbf{E}_1 = (1_x \cos \psi + 1_z \sin \psi) a_1 \cos(\omega t - ky + \Phi_1), \quad 0 \leq t - y/c \leq \tau_1, \quad (1)$$

$$\mathbf{E}_2 = 1_x a_2 \cos(ky + \varphi) \cos(\omega t + \Phi_2), \quad \tau + \tau_1 \leq t \leq \tau + \tau_1 + \tau_2. \quad (2)$$

The amplitudes  $a_1$  and  $a_2$  of the external electric field and the phase shifts  $\varphi$ ,  $\Phi_1$ , and  $\Phi_2$  are all constant real quantities,  $1_x$  and  $1_z$  are the unit vectors of the Cartesian axes  $X$  and  $Z$ , and  $\psi$  is the arbitrary angle between the polarization planes of waves (1) and (2). The delay time  $L/c$  of the electro-magnetic signal between the points of entrance in ( $y = 0$ ) and exit from ( $y = L$ ) the investigated gas is small compared with the duration  $\tau_2$  of the second pulse (2) and the characteristic times of the problem. It can therefore be assumed that the standing-wave pulse (2) is produced simultaneously over the entire extent of the gas medium.

The electric field  $\mathbf{E}$ , with account taken of the reaction of the gas medium, is described with the aid of the d'Alembert equation for  $\mathbf{E}$  and the quantum-mechanical equation for the density matrix; we shall express them in the two-level approximation in the convenient notation

$$\left( \frac{\partial^2}{\partial y^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \mathbf{E} = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \int \sum_{\mu m} (\rho_{\mu m} \mathbf{d}_{\mu m} + \mathbf{d}_{\mu m} \rho_{\mu m}) dV, \quad (3)$$

$$\left( \frac{\partial}{\partial t} + v_y \frac{\partial}{\partial y} + i\omega_0 + \frac{\gamma_a + \gamma_b}{2} \right) \rho_{\mu m} = \frac{i\mathbf{E}}{\hbar} \left( \sum_{m'} \mathbf{d}_{\mu m'} \rho_{m' m} - \sum_{\mu'} \rho_{\mu' m} \mathbf{d}_{\mu' m} \right) - \sum_{\mu' m' q} (-1)^{2j_b + \mu + \mu'} \times (2\kappa + 1) \Gamma^{(\kappa)} \begin{pmatrix} j_b & j_a & \kappa \\ \mu & -m & q \end{pmatrix} \begin{pmatrix} j_b & j_a & \kappa \\ \mu' & -m' & q \end{pmatrix} \rho_{\mu' m'}, \quad (4)$$

$$\left( \frac{\partial}{\partial t} + v_y \frac{\partial}{\partial y} + \gamma_b \right) \rho_{\mu m} = \frac{i\mathbf{E}}{\hbar} \sum_m (\mathbf{d}_{\mu m} \rho_{\mu m} - \rho_{\mu m} \mathbf{d}_{\mu m}) + \frac{\gamma_b N_b}{2j_b + 1} f(v) \delta_{\mu m} - \sum_{\mu' m' q} (-1)^{2j_b + \mu + \mu'} \times (2\kappa + 1) \Gamma_b^{(\kappa)} \begin{pmatrix} j_b & j_b & \kappa \\ \mu & -\mu' & q \end{pmatrix} \begin{pmatrix} j_b & j_b & \kappa \\ \mu_1 & -\mu_1' & q \end{pmatrix} \rho_{\mu' m'}, \quad (5)$$

while the equation for  $\rho_{\mu m}$  is obtained from (5) by the simultaneous interchange of the indices  $\mu \rightarrow m$ ,  $m \rightarrow \mu$ , and  $b \rightarrow a$ . The density matrices  $\rho_{\mu \mu'}$  and  $\rho_{m m'}$  characterize here the atom respectively on the upper and lower excited levels with energies  $E_b$  and  $E_a$ , and also with total angular momenta  $j_b$  and  $j_a$ . The density matrix  $\rho_{\mu m}$  describes the transitions between the indicated levels, the degeneracy of which is connected with the projections  $\mu$  and  $m$  of the total angular momenta. The frequency  $\omega_0 = (E_b - E_a)/\hbar$  of the atomic transition  $j_b - j_a$  is close to the frequency  $\omega = kc$  of the electromagnetic pulses (1) and (2). Next,  $\mathbf{d}_{\mu m}$  is the matrix element of the operator of the electric dipole moment  $\mathbf{d}$ ,  $N_b$  and  $N_a$  are the stationary densities of the atoms on the upper and lower levels in the absence of the external fields (1) and (2),  $v_y$  is the projection of the thermal velocity  $\mathbf{v}$  on the Cartesian  $Y$  axis,  $u$  is the most probable velocity of the resonant atoms, and

$$f(v) = (\pi^{-1/2} u)^{-3} \exp(-v^2/u^2)$$

is the Maxwellian distribution. The constants  $\hbar\gamma_b$  and  $\hbar\gamma_a$  are the partial widths of the upper and lower levels and are due to the radiative decay and inelastic gaskinetic collisions, while the quantities  $\hbar\Gamma_b^{(\kappa)}$  and  $\hbar\Gamma_a^{(\kappa)}$  char-

acterize the contributions of the elastic depolarizing atomic collisions to the widths of the upper and lower levels. The level widths  $\hbar(\gamma_b + \Gamma_b^{(\kappa)})$  and  $\hbar(\gamma_a + \Gamma_a^{(\kappa)})$  are large compared with the partial width  $\hbar\gamma$  of the upper level and connected with the probability  $\gamma$  of spontaneous emission of a quantum  $\hbar\omega_0$  by an isolated atom:

$$\gamma = 4|d|^2 \omega_0^3 / 3(2j_b + 1) \hbar c^2,$$

where  $d$  is the reduced dipole moment.<sup>10</sup> The real and imaginary parts of  $\hbar\Gamma^{(\kappa)}$  describe respectively the broadening and shift of the spectral line as a result of elastic depolarizing atomic collisions. The quantities  $\Gamma^{(\kappa)}$  with  $\kappa > 1$  characterize the relaxation of the multipole moments of the atom. The notation for the  $3j$  symbols is universal.<sup>10</sup>

It is assumed in (4) and (5) that the density  $N_a + N_b$  of the atoms that populate the excited resonant levels is small compared with the density of the unexcited atoms of the host and impurity gases. If resonant exchange of excitation between identical atoms is possible in the collision, then the contribution made to (4) and (5) by the produced dipole-dipole interaction should be small compared with the contribution from the van der Waals interaction with the impurity atoms. The cross section for the elastic collision should then be large compared with the gaskinetic cross section. Under these assumptions, the relaxation due to gaskinetic collision and the radiative decay are taken into account in (4) and (5) with the aid of the usual constants  $\gamma_a$  and  $\gamma_b$ , whereas the intense elastic collisions are described in greater detail by the method of Refs. 11-13.

To simplify the right-hand sides of (4) and (5) we neglect the changes of the atom velocities in the collision, but take into account the reorientation of the angular momentum of the excited atom, i.e., we take into consideration transitions between Zeeman sublevels of one and the same level (the model of elastic depolarizing collisions). Moreover, the collision matrices in (4) and (5) are averaged over the direction of the velocity of the excited atom. Therefore the quantities  $\Gamma^{(\kappa)}$ ,  $\Gamma_a^{(\kappa)}$ , and  $\Gamma_b^{(\kappa)}$  are functions of the modulus  $v$  of the velocity of the excited atom. Since elastic collisions do not alter the level populations, the relation  $\Gamma_a^{(0)} = \Gamma_b^{(0)} = 0$  is satisfied.

For an interaction of the van der Waals type, calculations of the quantities  $\Gamma^{(\kappa)}$ ,  $\Gamma_a^{(\kappa)}$ , and  $\Gamma_b^{(\kappa)}$  for the atomic transitions  $1 \rightleftharpoons 0$ ,  $1 - 1$ ,  $2 \rightleftharpoons 1$ ,  $\frac{1}{2} \rightarrow \frac{1}{2}$ , and  $\frac{3}{2} \rightleftharpoons \frac{1}{2}$  were carried through to conclusion and are given in Refs. 13-15.

Prior to the appearance of the electromagnetic pulse (1), the gas medium was in an equilibrium state described by the density matrices

$$\rho_{\mu m}|_{t=0} = 0, \quad \rho_{\mu \mu'}|_{t=0} = \frac{N_b f(v)}{2j_b + 1} \delta_{\mu \mu'}, \quad (6)$$

$$\rho_{m m'}|_{t=0} = \frac{N_a f(v)}{2j_a + 1} \delta_{m m'}.$$

These relations are the initial conditions for Eqs. (4) and (5), and determine the normalization of the density matrices.

We seek the solution of (4) and (5) in the given-field approximation, neglecting the reaction of the resonant

atoms to this external field (optically thin medium). The durations  $\tau_1$  and  $\tau_2$  of the pulses (1) and (2) are assumed to be small:

$$(\gamma_a + \gamma_b + \Gamma^{(a)})\tau_n \ll 1, \quad |\Gamma^{(a)''}|\tau_n \ll 1,$$

$$\Gamma_a^{(a)}\tau_n \ll 1, \quad \Gamma_b^{(a)}\tau_n \ll 1, \quad |\omega - \omega_0|\tau_n \ll 1, \quad n=1, 2,$$

where the single and double primes mark the real and imaginary parts of  $\Gamma^{(a)}$ . Consequently, during the time of action of the pulses (1) and (2) we can neglect the irreversible relaxation and the detuning  $\omega - \omega_0$  in (4) and (5). However,  $\tau \gg \tau_1$  and  $\tau \gg \tau_2$ , therefore in the time interval between the actions of the pulses (1) and (2) and after this action it is necessary to take into account in (4) and (5) the irreversible relaxation and the detuning.

According to the adopted calculation method, the field  $\mathbf{E}$  in (4) and (5) coincides with (1) in the region  $0 \leq t - y/c \leq \tau_1$ . We therefore substitute  $\rho_{\mu m} = Q_{\mu m} \exp[i(ky - \omega t - \Phi_1)]$  in (4) and (5) and discard terms with multiple frequencies (resonance approximation). We change over in the equations obtained for the slow functions  $Q_{\mu m}$ ,  $\rho_{\mu \mu'}$ , and  $\rho_{m m'}$  to new independent variables  $\xi = y - v_y t$  and  $t' = t$ , after which the solution under the initial conditions (6) is determined just as in Ref. 16. After the passage of the first pulse (1) we put  $\mathbf{E} = 0$  in (4) and (5), and obtain the solution of the initial equations with allowance for the detuning  $\omega - \omega_0$  and the collision integrals by expanding in the irreducible tensor operator, in analogy with the procedure in Ref. 11.

During the time interval  $\tau + \tau_1 \leq t \leq \tau + \tau_1 + \tau_2$  the field  $\mathbf{E}$  in (4) and (5) is the standing wave (2), and this indicates that it is advantageous to substitute  $\rho_{\mu m} = R_{\mu m} \exp[-i(\omega t + \Phi_2)]$  in these equations and introduce the notation  $\rho_{\mu \mu'} \equiv R_{\mu \mu'}$  and  $\rho_{m m'} \equiv R_{m m'}$ . In the resonance approximation and neglecting the irreversible relaxation and the detuning, we obtain for the matrix  $R$ , in terms of the variables and  $t'$ , the following operator equation

$$\frac{\partial R}{\partial t'} = \mathbf{d}_s(\mathbf{d}R - R\mathbf{d}) - \frac{a_2}{2\hbar} \cos(k\xi + kv_y t' + \varphi).$$

Its symbolic solution is

$$R = e^{iA} R_0 e^{-iA}, \quad (7)$$

$$A = \frac{a_2}{2\hbar} \mathbf{1}_d \int_{t_0}^{t'} \cos(k\xi + kv_y t + \varphi) dt.$$

Here  $R_0$  is the value of the matrix  $R$  at the initial instant of time  $t'_0 = \tau + \tau_1$ , and the nonzero matrix elements of the operators  $\cos A$  and  $\sin A$  in (7) take the form

$$(\cos A)_{\mu\mu'} = \delta_{\mu\mu'} \cos(F\Theta_\mu), \quad (\cos A)_{m m'} = \delta_{m m'} \cos(F\Theta_m),$$

$$(\sin A)_{\mu m} = (\sin A)_{m\mu} = (-1)^{j_b - \mu} \frac{d}{|d|} \delta_{\mu m} \sin(F\Theta_\mu),$$

where the quantization axis is chosen along the vector  $\mathbf{l}_s$  and we denote

$$F = \frac{a_2 |d|}{\hbar kv_y} \sin \frac{kv_y(t' - t'_0)}{2} \cos \left( k\xi + \varphi + kv_y \frac{t' + t'_0}{2} \right),$$

$$\Theta_\nu = \begin{pmatrix} j_b & 1 & j_a \\ -\nu & 0 & \nu \end{pmatrix}. \quad (8)$$

The described procedure has made it possible to find for the first time ever an analytic solution of the equations (4) and (5) for the density matrix in the field of a standing wave of arbitrary intensity, and for any mul-

tiplicity of level degeneracy, but in the absence of irreversible relaxation and detuning. The solution obtained for  $R$  at  $t' = \tau + \tau_1 + \tau_2$  serves as the initial condition for the determination of the density matrix after the action of the second pulse (2), when we have  $\mathbf{E} = 0$  in (4) and (5) and take into account and the irreversible relaxation.

From the density matrix  $\rho_{\mu m}$  obtained in this manner we determine the coherent radiation induced by the excited medium, by solving Eq. (3) by the method of Ref. 7. The electric field  $\mathbf{E}_k$  of this coherent radiation takes after emerging from the gas medium takes the form

$$\mathbf{E}_k = \mathbf{e}^{(+)}(t - y/c) e^{i(ky - \omega t - \Phi_2)} + \mathbf{e}^{(-)}(t + y/c) e^{-i(ky + \omega t + \Phi_2)} + c.c.,$$

where the amplitudes of the forward  $\mathbf{e}^{(+)}(t)$  and backward  $\mathbf{e}^{(-)}(t)$  waves are expressed in terms of the obtained matrix  $R_{\mu m}$  in the following manner:

$$\mathbf{e}^{(\pm)}(t) = iL \frac{\omega^2}{c^2} \int d\mathbf{v} e^{\mp i k v_y t'} \int_{-n c/\omega}^{n c/\omega} \sum_{\mu m} R_{\mu m} \mathbf{d}_{\mu m} e^{\mp i k \xi} d\xi. \quad (9)$$

The coherent radiation (9) breaks up into two groups of terms. Those in the first attenuate rapidly during the time of the reversible Doppler relaxation  $T_0 = 1/kv_y$ , which is small compared with  $\tau$  and with the time of the irreversible relaxation due to radiative decay and atomic collisions. On the other hand, the second group also relaxes first, but then increases again in the form of a photon echo that is described at  $\tau + \tau_1 + \tau_2 \leq t$  by the expression

$$\mathbf{e}^{(\pm)}(t) = \mathbf{e}_0 \exp[i(\Delta t + \Phi^{(\pm)})] \int d\mathbf{v} f(\mathbf{v}) (1, W_0^{(\pm)} + 2^{1/2} W_1^{(\pm)}), \quad (10)$$

where we have introduced the notation ( $p = 0, 1$ )

$$W_p^{(\pm)} = \sum_{\mu} \exp[-\gamma^{(1)}(t - \tau) - \gamma^{(2)'} \tau \pm i\gamma^{(2)''} \tau] G_{\mu p} H_{\mu p}^{(\pm)},$$

$$G_{\mu p} = (2\kappa + 1)^{1/2} Y_{\mu p}(\psi, 0) \sum_{\nu} \Theta_{\nu} \begin{pmatrix} j_b & \kappa & j_a \\ -\mu & 0 & \mu \end{pmatrix} \frac{\Lambda_1}{\Omega_{\mu}}$$

$$\times \left[ \sin \Omega_{\mu} \tau, \cos kv_y t_0 + \frac{kv_y}{\Omega_{\mu}} (1 - \cos \Omega_{\mu} \tau) \sin kv_y t_0 \right],$$

$$H_{\mu p}^{(+)} = \sum_{m} \begin{pmatrix} j_b & 1 & j_a \\ -\mu & p & m \end{pmatrix} \begin{pmatrix} j_b & \kappa & j_a \\ -\mu & p & m \end{pmatrix} \cdot [J_2(F_0 \Theta_{\mu} + F_0 \Theta_m) - J_2(F_0 \Theta_{\mu} - F_0 \Theta_m)],$$

$$H_{\mu p}^{(-)} = (-1)^p \sum_{m} \begin{pmatrix} j_b & 1 & j_a \\ -\mu & p & m \end{pmatrix} \begin{pmatrix} j_b & \kappa & j_a \\ -\mu & p & m \end{pmatrix}$$

$$\times [J_2(F_0 \Theta_{\mu} + F_0 \Theta_m) + J_2(F_0 \Theta_{\mu} - F_0 \Theta_m)],$$

$$e_0 = \pi^{1/2} L |d| \left( \frac{N_a}{2j_a + 1} - \frac{N_b}{2j_b + 1} \right) \frac{\omega}{c}, \quad F_0 = \frac{\Lambda_2}{kv_y} \sin \frac{kv_y \tau_2}{2},$$

$$\gamma^{(a)} = (\gamma_a + \gamma_b)/2 + \Gamma^{(a)} = \gamma^{(a)'} + i\gamma^{(a)''}, \quad t_0 = t - 2\tau - \tau_1 - \tau_2,$$

$$\Phi^{(-)} = -2\varphi + \Phi_2 - \Phi_1, \quad \Phi^{(+)} = -\Phi^{(-)} - 2\Delta\tau, \quad \Delta = \omega - \omega_0,$$

$$\Omega_{\mu} = [(kv_y)^2 + (\Lambda_1 \Theta_{\mu})^2]^{1/2}, \quad \Lambda_n = a_n |d|/\hbar, \quad n=1, 2.$$

Here  $J_2(s)$  are Bessel function of second order and of argument  $s$ ,  $Y_{\mu p}(\psi, 0)$  is the spherical function  $Y_{\mu p}(\psi, \chi)$  (Ref. 10) at  $\chi = 0$ , while  $\Theta_{\mu}$  and  $\Theta_m$  are given by expression (8) with  $\nu = \mu$  and  $\nu = m$ . According to the formula for  $G_{\mu p}$ , the only nonzero terms of  $W_p^{(\pm)}$  are those with odd  $\kappa$ .

The foregoing reasoning are valid also for resonant molecular transitions between quantum states whose degeneracy is due to differences in the angular-momentum orientations.

In the case of a narrow spectral line, when the inhom-

geneous width  $1/T_0 = k$  of the Doppler contour is small compared with the width of the spectral distribution of each exciting pulse

$$1/T_0 \ll 1/\tau_1, \quad 1/T_0 \ll 1/\tau_2, \quad (11)$$

the basic formula (10) takes the simple form

$$\varepsilon^{(\pm)}(t) = \varepsilon_0 \exp[i(\Delta t + \Phi^{(\pm)}) - (t - 2\tau)^2/4T_0^2] (1_s w_0^{(\pm)} + 2^{1/2} 1_s w_1^{(\pm)}), \quad (12)$$

$$w_p^{(\pm)} = \sum_{\mu} \exp[-\bar{\gamma}^{(\pm)}(t-\tau) - \bar{\gamma}^{(\pm)'} \tau \pm i\bar{\gamma}^{(\pm)''} \tau] g_{\mu p} h_{\mu p}^{(\pm)},$$

$$g_{\mu p} = (2\kappa + 1)^{1/2} Y_{\mu p}(\psi, 0) \sum_{\mu} \begin{pmatrix} j_b & \kappa & j_a \\ -\mu & 0 & \mu \end{pmatrix} \sin(\Lambda_1 \tau, \Theta_\mu),$$

$$h_{\mu p}^{(+)} = \sum_{\mu m} \begin{pmatrix} j_b & 1 & j_a \\ -\mu & p & m \end{pmatrix} \begin{pmatrix} j_a & \kappa & j_b \\ -\mu & p & m \end{pmatrix}$$

$$\times \left[ J_2 \left( \Lambda_2 \tau_2 \frac{\Theta_\mu + \Theta_m}{2} \right) - J_2 \left( \Lambda_2 \tau_2 \frac{\Theta_\mu - \Theta_m}{2} \right) \right],$$

$$h_{\mu p}^{(-)} = (-1)^p \sum_{\mu m} \begin{pmatrix} j_b & 1 & j_a \\ -\mu & p & m \end{pmatrix} \begin{pmatrix} j_b & \kappa & j_a \\ -\mu & p & m \end{pmatrix}$$

$$\times \left[ J_2 \left( \Lambda_2 \tau_2 \frac{\Theta_\mu + \Theta_m}{2} \right) + J_2 \left( \Lambda_2 \tau_2 \frac{\Theta_\mu - \Theta_m}{2} \right) \right].$$

In the derivation of (12) it was assumed that the quantity  $\gamma^{(\kappa)}$  as a function of the velocity  $v$  varies slowly in the effective region  $v \sim u$ . It was therefore replaced by its value at  $v = u$  and represented in the form of a sum of its real and imaginary parts:

$$\bar{\gamma}^{(\kappa)} = \gamma^{(\kappa)}|_{v=u} = \bar{\gamma}^{(\kappa)'} + i\bar{\gamma}^{(\kappa)''}.$$

This replacement is valid for collisions of resonant atoms  $M$  with impurity atoms  $M_0$  whose masses satisfy the condition  $M \gg M_0$ .

## 2. PROPERTIES OF FORWARD AND BACKWARD ECHOES

By comparing formulas (10) and (12) with the experimental results we can extract extensive spectroscopic information on the relaxation processes and energy levels, as is clearly seen from an assessment of the obtained general relations and from an analysis of simple particular cases.

For the  $\frac{1}{2} \rightarrow \frac{1}{2}$  atomic transition we have from (12)

$$\varepsilon^{(\pm)}(t) = (1_s \cos \psi \mp 1_s \sin \psi) \times e_0(t) \exp[i(\Delta t + \Phi^{(\pm)} - \bar{\gamma}^{(\pm)'}(t-\tau) \pm \bar{\gamma}^{(\pm)''} \tau)], \quad (13)$$

$$e_0(t) = \frac{\varepsilon_0}{(6\pi)^{1/2}} \sin\left(\frac{\Lambda_1 \tau_1}{6^{1/2}}\right) J_2\left(\frac{\Lambda_2 \tau_2}{6^{1/2}}\right) \exp\left[-\left(\frac{t-2\tau}{2T_0}\right)^2 - \bar{\gamma}^{(\pm)'} t\right].$$

According to (13) the forward echo  $\varepsilon^{(+)}(t)$  has the same polarization as the photon echo from two traveling waves,<sup>17,18</sup> while the backward echo  $\varepsilon^{(-)}(t)$  has the polarization of the first exciting pulse. At  $\psi = \pi/2$  the polarization planes of the forward and backward echoes coincide, in agreement with the experimental results.<sup>9</sup>

The intensity of the forward and backward echoes at the maximum depend on the delay time  $\tau$  via the factor  $\exp(-4\bar{\gamma}^{(\pm)'} \tau)$ . By performing a set of echo-intensity measurements at the maximum for different values of  $t$ , it is easy to determine the width  $\hbar\bar{\gamma}^{(\pm)'}$  of the spectral line. Moreover, if reflecting mirrors are used to superimpose the forward and backward echo signals (13), then the intensity of the obtained superposition is proportional at  $\psi = \pi/2$  to the factor

$$\sin^2[(\Delta - \bar{\gamma}^{(\pm)''} \tau + \Phi_2 - \Phi_1 - 2\varphi) \exp(-2\bar{\gamma}^{(\pm)'} t)],$$

which makes it possible to determine the collision shift  $\hbar\bar{\gamma}^{(\pm)'}$  of the spectral line from the results of measurements made for different values of  $\tau$  with the other parameters unchanged. This factor gives rise to a characteristic narrow resonance of width  $1/2\tau$  when the frequency  $\omega$  of the exciting pulses is scanned. We emphasize that the forward and backward echo signals taken separately do not contain this resonance, which appears only when both exciting pulses are standing waves.<sup>4-7</sup>

Another way of obtaining the characteristic narrow resonance and determining the collision shift  $\hbar\bar{\gamma}^{(\pm)'}$  of the spectral line is to add the backward echo signal to a monochromatic wave

$$E_0 = \varepsilon_0 \cos[\omega'(t+y/c) + \Phi_0] \quad (14)$$

of a heterodyne laser with  $|\omega' - \omega_0| \ll \omega_0$ . It is convenient here to use only the projection of the amplitude  $\varepsilon^{(-)}(t+y/c)$  of the reverse echo on the  $X$  axis. The backward-echo signal at  $\psi \neq 0$  is then completely separated from the excited pulses, thereby increasing substantially the sensitivity of the measurement. The intensity  $I$  of the obtained summary field undergoes modulation oscillations:

$$I = \frac{c\varepsilon_0^2}{8\pi} + \frac{c\varepsilon_0 e_0(t+y/c)}{2\pi} \sin \psi \cos[(\omega' - \omega_0 - \bar{\gamma}^{(\pm)'}) (t+y/c) + \Phi_0 - \Phi_1 - 2\varphi],$$

from which the sought information can be extracted.

It follows from the general formulas (10) and (12) that elastic atomic collisions do not influence the polarizations  $\varepsilon^{(+)} t$  and  $\varepsilon^{(-)} t$  of the direct and reverse echoes in the atomic transitions  $1 \rightleftharpoons 0$ ,  $1 \rightarrow 1$ ,  $\frac{1}{2} \rightarrow \frac{1}{2}$ , and  $\frac{3}{2} \rightleftharpoons \frac{1}{2}$ . In other transitions, however, the atomic collisions do change the echo polarization, and this can be used to check on the validity of the chosen model of atomic collisions, and to determine the relations between the relaxation constants  $\Gamma^{(\kappa)}$  and various values of  $\kappa$ .

We illustrate the foregoing using as an example the atomic transitions  $2 \rightleftharpoons 1$  and a narrow spectral line (11), when the amplitude (12) for the backward echo is of the form

$$\varepsilon^{(-)}(t) = \{1_s \cos \psi [b_1 + b_2(5 \cos^2 \psi - 3)e^{i\tau}] + 1_s \sin \psi [b_3 + b_4(5 \cos^2 \psi - 1)e^{i\tau}]\} e_1(t) \exp[i(\Delta t + \Phi^{(-)} - \bar{\gamma}^{(-)'} t)], \quad (15)$$

$$e_1(t) = \frac{\varepsilon_0}{(250\pi)^{1/2}} \exp\left[-\left(\frac{t-2\tau}{2T_0}\right)^2 - \bar{\gamma}^{(-)'} t\right],$$

$$b_1 = (3 \sin \delta_1 + 3^{1/2} \sin \beta_1) [J_2(\delta_2) + {}^{+2}/J_2(\beta_2)],$$

$$b_2 = \left(\sin \delta_1 - \frac{3^{1/2}}{2} \sin \beta_1\right) [J_2(\delta_2) - J_2(\beta_2)],$$

$$b_3 = 2(3 \sin \delta_1 + 3^{1/2} \sin \beta_1) \left\{ J_2\left(\frac{\delta_2}{2}\right) + \frac{1}{3} \left[ J_2\left(\frac{\delta_2 + \beta_2}{2}\right) + J_2\left(\frac{\delta_2 - \beta_2}{2}\right) \right] \right\},$$

$$b_4 = \left(\sin \delta_1 - \frac{3^{1/2}}{2} \sin \beta_1\right) \left\{ J_2\left(\frac{\delta_2}{2}\right) - \frac{1}{2} \left[ J_2\left(\frac{\delta_2 + \beta_2}{2}\right) + J_2\left(\frac{\delta_2 - \beta_2}{2}\right) \right] \right\}$$

$$\gamma_{13} = \bar{\gamma}^{(-)} - \bar{\gamma}^{(+)}, \quad \delta_n = 10^{-1/2} \Lambda_n \tau_n,$$

$$\beta_n = ({}^{2}/1_s)^{1/2} \Lambda_n \tau_n, \quad n = 1, 2.$$

In the experiment it is convenient to investigate only the projection of the amplitude (15) on the  $X$  axis, corresponding to a radiation intensity

$$I_e = \frac{c[e_1(t+y/c)]^2}{2\pi} \sin^2 \psi [b_3^2 + b_4^2 (5 \cos^2 \psi - 1)^2 e^{2\gamma_{13} \tau} + 2b_3 b_4 (5 \cos^2 \psi - 1) e^{\gamma_{13} \tau} \cos \gamma_{13} \tau], \quad (16)$$

where we put  $\gamma^{13} = \gamma^{13'} + i\gamma^{13''}$ .

If we choose the angle  $\psi$  to satisfy the condition  $5 \cos^2 \psi = 1$ , then the intensity (16) contains only one relaxation constant  $\bar{\gamma}^{(1)}$ . It can be easily determined, just as in the case of the atomic transition  $\frac{1}{2} \rightarrow \frac{1}{2}$ . At other values of the angle  $\psi$ , the intensity (16) acquires a characteristic dependence on the parameters  $\gamma^{13}$  and  $\gamma^{13''}$ , and this can be used to determine the latter.

To find the remaining constant  $\bar{\gamma}^{(1)n}$  we must add the  $x$ -projection of the amplitude (15) to the monochromatic wave (14) of the heterodyne laser and investigate the intensity  $I$  of the combined wave outside the gas medium:

$$I = \frac{ca_0^2}{8\pi} + \frac{ca_0 e_i (t+y/c)}{2\pi} \sin \psi \{ b_s \cos [(\omega' - \omega_0 - \bar{\gamma}^{(1)'}) (t+y/c) + \Phi] + b_e \exp(\gamma_{13} \tau) (5 \cos^2 \psi - 1) \cos [(\omega' - \omega_0 - \bar{\gamma}^{(1)'}) (t+y/c) + \Phi + \gamma_{13} \tau] \},$$

$$\Phi = \Phi_0 - \Phi_1 - 2\varphi.$$

Investigation of the reverse echo yields thus the characteristics  $\Gamma^{(1)}$  and  $\Gamma^{(2)}$  of elastic atomic collisions, which enter in the expressions  $\gamma^{(1)} = (\gamma_a + \gamma_b)/2 + \Gamma^{(1)}$  and  $\gamma^{(2)} = (\gamma_a + \gamma_b)/2 + \Gamma^{(2)}$ . The real term  $(\gamma_a + \gamma_b)/2$  can be determined independently by the same method at low gas pressures, when the atomic collisions are negligible.

If the elastic collisions are characterized by a large set of relaxation constants  $\gamma^{(\kappa)}$  with  $\kappa = 1, 3, \dots$ , then the broadening  $\bar{\kappa} \gamma^{(1)}$ , and the shift  $\bar{\kappa} \gamma^{(1)n}$  of the spectral line must be determined by using a traveling-wave pulse (1) having a small area  $\Lambda_1 \tau_1 \ll 1$ . The only one term, with  $\kappa = 1$ , is significant in the general formulas (10) and (12), while the remaining terms are negligibly small.

The polarization properties  $\epsilon^{(+)}(t)$  and  $\epsilon^{(-)}(t)$  of the forward and backward echoes in the atomic transitions  $j \rightarrow j$  and  $j \rightleftharpoons j+1$ , with  $j \gg 1$ , are much clearer in the limiting case of small areas  $\Lambda_1 \tau_1 \ll 1$  and  $\Lambda_2 \tau_2 \ll 1$  of the exciting pulses (1) and (2), when the expression (12) for the amplitude becomes much simpler:

$$\epsilon^{(\pm)}(t) = e^{(\pm)}(t) (I_1 \cos \psi + i I_2 \sin \psi) \quad \text{for } j \rightarrow j;$$

$$\epsilon^{(\pm)}(t) = i e^{(\pm)}(t) (I_1 \cos \psi + a^{(\pm)} I_2 \sin \psi) \quad \text{for } j \rightleftharpoons j+1;$$

$$e^{(\pm)}(t) = \epsilon_0 \frac{\Lambda_1 \tau_1 (\Lambda_2 \tau_2)^2}{160 j \pi^{3/2}} \exp \left\{ - \left( \frac{t-2\tau}{2T_0} \right)^2 - \bar{\gamma}^{(1)' } t + i [\Delta t + \Phi^{(\pm)} - \bar{\gamma}^{(1)' } (t-\tau) \pm \bar{\gamma}^{(1)' } \tau] \right\},$$

$$a^{(+)} = -1/2, \quad a^{(-)} = 1/4.$$

It is seen that the polarization plane of the backward echo lies, for all types of atomic transitions, within the acute angle made by the polarization planes of the exciting pulses. At the same time, the polarization plane of the direct echo for the atomic transitions  $j \rightleftharpoons j+1$  lies inside the obtuse angle made by the polarization planes of the exciting pulses, and inside an acute angle

for the  $j \rightarrow j$  transition. This feature of the forward echo coincides with the properties of the photon echo produced by two traveling-wave pulses.<sup>18</sup>

A detailed analysis shows that the obtained dependence of the polarization of the direct echo on the type of the atomic transition is valid also in a wider range  $\Lambda_n \tau_n \lesssim 1$ ,  $n = 1, 2$ , independently of the condition (11); this makes it possible to distinguish in experiment between the  $j \rightarrow j$  transition and  $j \rightleftharpoons j+1$  transitions. An investigation of the functional dependence of the intensities of the forward and backward echo on the angle  $\psi$  will also make it possible to identify atomic and molecular transitions.<sup>19</sup>

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