

PHOTON ECHO POLARIZATION IN A GAS MEDIUM

A. I. ALEKSEEV and I. V. EVSEEV

Moscow Engineering-physics Institute

Submitted January 22, 1969

Zh. Eksp. Teor. Fiz. 56, 2118-2128 (June, 1969)

The amplitude and polarization of a photon echo in a gaseous medium are determined for various atomic transitions and for the case of linear or circular polarization of the light pulses. It is proved that for atomic transitions involving a change of the total momentum $0 \rightleftharpoons 1$ and $1 \rightarrow 1$, the photon echo has the polarization of the second pulse. If the light pulses are linearly polarized the photon-echo amplitude is proportional to $\cos \psi$, where ψ is the angle between the polarization vectors of the pulses. The photon-echo intensity for the $J_2 = \frac{1}{2} \rightarrow J_1 = \frac{1}{2}$ atomic transition is independent of angle ψ and the polarization vector of the photon echo makes an angle 2ψ with the polarization direction of the first pulse.

PHOTO echo was observed recently in a gas medium by transmitting two pulses of light with wavelength $\lambda = 10.6 \mu$ from a CO_2 laser through SF_6 gas.^[1] The quantum-mechanical transition that is resonant to the 10.6μ wavelength is $J_2 = J + 1 \rightarrow J_1 = J$ between two excited rotational levels of the SF_6 molecules with angular momenta J_2 and J_1 , respectively. The numerical value of J was not determined exactly. Owing to the Boltzmann factor, only 1/300th of the total number of the SF_6 molecules, whose density was $5 \times 10^{14} \text{ cm}^{-3}$, took part in the echo formation. The obtained phenomenon was used by the authors to determine experimentally the collision width of the excited level as a function of the pressure and of the presence of impurity gases. It was established that, unlike the photon echo in ruby,^[2,3] the polarization of photon echo in gas coincides with the polarization of the second signal, and the echo intensity depends on the angle ψ between the polarization vectors of the transmitted light pulses approximately like $\cos^2 \psi$.

In the present article we present a theoretical investigation of the polarization effects of the photon echo in a gas. It is shown that for quantum-mechanical transitions with total angular-momentum change $0 \rightleftharpoons 1$ and $1 \rightarrow 1$, the polarization and the amplitude of the photon echo agree with the experimental data of^[1]. If the first and second signals are respectively linearly and circularly polarized (or vice versa), then the photon-echo polarization coincides with the polarization of the second signal. If the two transmitted signals are circularly polarized, then the photon echo is produced only when the directions of rotation of the electric field vectors of both signals are the same. Thus, our investigation leads to the conclusion that for the indicated atomic transitions the photon echo in a gas medium has the same polarization as the second signal.

The foregoing rule does not hold for photon echo on the atomic transition $J_2 = \frac{1}{2} \rightarrow J_1 = \frac{1}{2}$. A distinguishing feature of this transition leads to the fact that the resonant linearly-polarized wave breaks up into right- and left-polarized circular waves, which propagate independently of each other. At the same time, any two linearly polarized waves are mutually dependent in such a medium. As a result, a linearly polarized pulse following a circular pulse produces a photon echo with circular polarization, a factor of importance for an experimental

identification of the echo.

An analogous singularity of the propagation of right- and left-polarized waves is observed also in ruby placed in a magnetic field. At low temperature, the atomic transitions with change of projection M of the total angular momentum

$${}^4A_2(M = -\frac{1}{2}) \rightarrow 2E(\bar{E})(M = -\frac{1}{2}), {}^4A_2(M = \frac{1}{2}) \rightarrow 2E(\bar{E})(M = \frac{1}{2}),$$

form in ruby two independent two-level subsystems without degeneracy; the first of them interacts only with the right-polarized wave, and the second with the left-polarized wave.^[3] The independence of the propagation of the circular waves in ruby leads to a specific rotation of the photon-echo polarization by an angle 2ψ relative to the polarization of the first pulse. Unlike in ruby, the energy levels with total angular momentum $\frac{1}{2}$ in a gas are doubly degenerate. Nonetheless, the photon-echo polarization in the gas is also rotated through an angle 2ψ in the atomic transition $\frac{1}{2} \rightarrow \frac{1}{2}$. If the gas is placed, like the ruby, in a magnetic field, then the polarization effects of the photon echo will be quite different in these two media.

In photon-echo polarization in a gas, allowance for the level degeneracy is a fundamentally essential factor, for in the case of dipole transitions 1 or 2 of the working levels are degenerate. Therefore, the methodological approach to the investigation of photon echo, proposed in^[3] for two-level systems without degeneracy, cannot be applied to gases without any stipulation. In particular, the intuitive interpretation is lost. The concepts of 90 and 180° pulses, which are borrowed from spin echo,^[4] lose their distinct meaning in the case of photon echo in a gas medium, if the time of reversible relaxation is small compared with the time duration of the light pulses, as was the case in the experiment of^[1].

1. FUNDAMENTAL EQUATIONS

As the fundamental equations we take

$$\square A_\alpha = -\frac{4\pi}{c} \int dV \text{Sp } I_\alpha \quad (1)$$

and the quantum-mechanical equation for the density matrix, in terms of which the polarization current I_α is expressed. The vector potential A_α and the current I_α are written in the form

$$A_\alpha = a_\alpha \exp[i(\mathbf{k}\mathbf{r} - \omega t + \Phi)], \quad (2)$$

$$I_\alpha = j_\alpha \exp[i(\mathbf{k}\mathbf{r} - \omega t + \Phi)], \quad (3)$$

where a_α and j_α are slowly-varying amplitudes, and the phase Φ is real and constant. An important role is played in the problem only by the phase difference between A_α and I_α , which we refer to j_α . The equations for the amplitudes without the relaxation terms are of the form¹⁾

$$\left(\frac{\partial}{\partial t} + c \frac{\partial}{\partial z}\right) a_\alpha = i2\pi\lambda \int dv \text{Sp } j_\alpha, \quad (4)$$

$$\left(\frac{\partial}{\partial t} + i\mathbf{k}\mathbf{v}\right) j_\alpha + i \frac{3}{4}(2J_2 + 1)\gamma c\lambda N_{\alpha\beta} a_\beta = 0, \quad (5)$$

$$\begin{aligned} \frac{\partial}{\partial t} N_{\alpha\beta} + \frac{i}{\hbar c} (a_\sigma^* j_\sigma - j_\sigma^+ a_\sigma) T_{\alpha\beta} \\ + \frac{i}{\hbar c} (a_\sigma^* T_{\sigma\beta} j_\alpha - j_\beta^+ T_{\alpha\sigma} a_\sigma) = 0, \end{aligned} \quad (6)$$

where

$$\begin{aligned} j_\alpha &= -i\omega_0 R_{\mu m} d_{m\mu}^\alpha \exp[i(\omega t - \mathbf{k}\mathbf{r} - \Phi)], \\ N_{\alpha\beta} &= \rho_2 T_{\alpha\beta} - \rho_1 \alpha^\beta, \quad T_{\alpha\beta} = d_{\mu m}^\beta d_{m\mu'}^\alpha / |d_{J_1 J_2}|^2, \\ \rho_2 &= \rho_{\mu\mu'}, \quad \rho_1 \alpha^\beta = d_{\mu m}^\beta \rho_{mm'} d_{m'\mu'}^\alpha / |d_{J_1 J_2}|^2, \\ \gamma &= 4 |d_{J_1 J_2}|^2 / 3(2J_2 + 1)\hbar\lambda^3. \end{aligned}$$

Here $\rho_{\mu\mu'}$ and $\rho_{mm'}$ are density matrices describing the quantum-mechanical state of the atom on the upper and lower degenerate levels with total angular momenta J_2 and J_1 , respectively; $R_{\mu m}$ is the density matrix describing the transitions between the indicated working levels, and $d_{\mu m}^\alpha$, $d_{J_1 J_2}^\alpha$ are the dipole and the reduced dipole moments of this transition; γ is the probability of spontaneous emission of a quantum $\hbar\omega_0$ per unit time for a separate isolated atom. The term $\mathbf{k} \cdot \mathbf{v}$ takes into account the Doppler variation of the frequency when the atom moves with velocity \mathbf{v} . The frequency ω of the electromagnetic field coincides with the frequency ω_0 of the atomic transition $\lambda = c/\omega$. For the different atomic transitions we obtain

a) for $J_2 = J \rightarrow J_1 = J$

$$T_{\alpha\beta} = \frac{\hat{J}_\beta \hat{J}_\alpha}{J(J+1)(2J+1)\hbar^2},$$

b) for $J_2 = J \rightarrow J_1 = J+1$

$$T_{\alpha\beta} = \frac{i(2J+3)\hbar e_{\alpha\beta\gamma} \hat{J}_\gamma + 2(J+1)^2 \hbar^2 \delta_{\alpha\beta} - (\hat{J}_\alpha \hat{J}_\beta + \hat{J}_\beta \hat{J}_\alpha)}{2(J+1)(2J+1)(2J+3)\hbar^2},$$

c) for $J_2 = J+1 \rightarrow J_1 = J$

$$T_{\alpha\beta} = \frac{-i(2J+1)\hbar e_{\alpha\beta\gamma} \hat{J}_\gamma + 2(J+1)^2 \hbar^2 \delta_{\alpha\beta} - (\hat{J}_\alpha \hat{J}_\beta + \hat{J}_\beta \hat{J}_\alpha)}{2(J+1)(2J+1)(2J+3)\hbar^2}$$

We write out the initial conditions for Eqs. (4)–(6):

$$a_\alpha(\mathbf{r}, 0) = j_\alpha(\mathbf{r}, 0) = 0, \quad (7)$$

$$N_{\alpha\beta}(\mathbf{r}, 0) = [n_2 / (2J_2 + 1) - n_1 / (2J_1 + 1)] f T_{\alpha\beta}, \quad (8)$$

where n_2 and n_1 is the total number of atoms at the upper and lower levels respectively at the initial time $t = 0$, and

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

¹⁾Summation over repeated vector and matrix indices is implied. The matrix index is chosen to be the projection of the total angular momentum of the upper level.

is the Maxwell distribution function; u is the thermal velocity of the atom.

2. ECHO ON ATOMIC TRANSITION WITH ONE DEGENERATE LEVEL

Let us consider the simplest variant

$$J_2 = 0 \rightarrow J_1 = 1, \quad T_{\alpha\beta} = \delta_{\alpha\beta} / 3,$$

for which all the investigated formulas are quite simple. Let the transmitted pulses propagate along the Z axis. The first pulse, which is linearly polarized along the X axis, is incident on the boundary $z = 0$ of the medium at the instant $t = 0$. The reaction of the resonant medium on the transmitted pulses will be neglected. We then have from (5)–(8) in the region $0 \leq t - z/c \leq T_1$

$$\begin{aligned} j_1\left(t - \frac{z}{c}\right) &= -\frac{\gamma c \lambda a N_{0f}}{4\Omega_1} \left\{ \frac{\mathbf{k}\mathbf{v}}{\Omega_1} \left[1 - \cos \Omega_1 \left(t - \frac{z}{c} \right) \right] \right. \\ &\quad \left. + i \sin \Omega_1 \left(t - \frac{z}{c} \right) \right\}, \end{aligned} \quad (9)$$

$$j_2(t - z/c) = 0, \quad N_0 = n_2 - n_1 / 3,$$

$$N_{11}\left(t - \frac{z}{c}\right) = \frac{N_{0f}}{3} \left\{ 1 - \frac{\gamma \lambda a^2}{\hbar \Omega_1^2} \left[1 - \cos \Omega_1 \left(t - \frac{z}{c} \right) \right] \right\}, \quad (10)$$

$$N_{22}\left(t - \frac{z}{c}\right) = \frac{N_{0f}}{3} \left\{ 1 - \frac{\gamma \lambda a^2}{2\hbar \Omega_1^2} \left[1 - \cos \Omega_1 \left(t - \frac{z}{c} \right) \right] \right\}, \quad (11)$$

$$N_{33} = N_{22}, \quad \Omega_1^2 = (\mathbf{k}\mathbf{v})^2 + \gamma \lambda a^2 / \hbar. \quad (12)$$

where T_1 is the time duration of the first pulse, and the remaining components of the tensor $N_{\alpha\beta}$ vanish.

According to (4), the polarization current (9) induces a field \mathbf{a}_p , which assumes the largest value near the boundary $z = l$ of the gas medium:

$$\begin{aligned} \mathbf{a}_p &= \frac{\pi^{1/2}}{2} N_0 a \lambda^2 l \gamma T_0 \int_0^\infty \frac{e^{-\xi^2} \sin[T_1(\xi^2 + \varepsilon^2)^{1/2}/T_0]}{(\xi^2 + \varepsilon^2)^{1/2}} d\xi, \\ T_0 &= 1/kv, \quad t - z/c = T_1, \quad \varepsilon^2 = \gamma \lambda a^2 T_0^2 / \hbar. \end{aligned} \quad (13)$$

In order to be able to neglect the reaction of the medium on the transmitted pulse, it is necessary to have

$$a_p / a \ll 1. \quad (14)$$

For gaseous media, this inequality is readily satisfied as a result of the small excess population N_0 , since usually the two working levels are excited and their excess population is due to the Boltzmann distribution of the particles over the levels. For example, for the parameters of the experiments of^[1] we obtain

$$N_0 \lambda^2 l \gamma T_0 \sim 10^{-3},$$

where $\gamma = 10^{-1} \text{ sec}^{-1}$ is taken from^[5].

The solution of (5) and (6) in the region $T_1 \leq t - z/c$, after the passage of the first pulse, obtained by putting $a_\alpha = 0$. As a result we find that the current (3) is directed along the X axis

$$I_1(t - T_1 - z/c) = j_1(T_1) \exp\{i[\omega(z/c - t) - \mathbf{k}\mathbf{v}(t - T_1 - z/c) + \Phi_1]\}, \quad (15)$$

and the tensor $N_{\alpha\beta}$ is given by

$$N_{\alpha\beta} = N_{\alpha\beta}(T_1), \quad (16)$$

where $j_1(T_1)$ and $N_{\alpha\beta}(T_1)$ are the functions (9)–(12) taken at the instant of time $t = T_1 + z/c$. Here Φ_1 is the constant phase of the first pulse in the form (2).

After passage of the first pulse, the polarization cur-

rent (15) of the group of molecules moving with velocity v differs from zero in all the subsequent instants of time. Yet the average polarization current, in the right side of Eq. (1), attenuates exponentially as a result of the Doppler dephasing.

The polarization current (15) induces an electromagnetic field whose order of magnitude is given by (13). This field, however, attenuates exponentially in the region $T_1 < t - z/c$,

$$\exp[-(t - T_1 - z/c)^2 / 4T_0^2].$$

Thus, the initial rectangular electromagnetic pulse with a time duration T_1 is followed by an exponentially damped "tail" of duration T_0 (usually $T_0 \ll T_1$). This "tail" produces in turn, in the region

$$T_1 \leq t - z/c \leq T_1 + T_0$$

an additional polarization current, which is much smaller than the main current (15), if the inequality (14) is satisfied.

At the instant $t = \tau$, a second pulse with amplitude b and time duration T_2 is seen along the Z axis; its polarization vector makes an angle ψ with the polarization of the first pulse. The vector potential and the current I in the region $\tau \leq t - z/c$ will be represented in the form

$$A_x = b_x \exp[i(kr - \omega t + \Phi_2)], \quad (17)$$

$$I_x = j_x \exp[i(kr - \omega t + \Phi_2)]. \quad (18)$$

We rotate the coordinate system around the Z axis so that the X axis coincides with the polarization vector of the second pulse. In the rotated coordinate system, Eqs. (4)–(6) remain unchanged, and the initial conditions are obtained from (15) and (16) by means of the rotation transformation at the instant of time $t = \tau + z/c$. As a result we get

$$j_1\left(t - \tau - \frac{z}{c}\right) = j_1^*(T_1) \cos \psi \frac{\gamma \lambda b^2}{2\hbar \Omega_1^2} \left[1 - \cos \Omega_2 \left(t - \tau - \frac{z}{c} \right) \right] \times \exp \{ i [k v (\tau - T_1) - \Phi_1 + \Phi_2] \}. \quad (19)$$

$$j_2\left(t - \tau - \frac{z}{c}\right) = j_1(T_1) \sin \psi \left[\cos \Omega_2 \left(t - \tau - \frac{z}{c} \right) - i \frac{k v}{\Omega_2} \sin \Omega_2 \left(t - \tau - \frac{z}{c} \right) \right] \exp \left\{ i \left[\Phi_1 - \Phi_2 - \frac{1}{2} k v \left(t + \tau - 2T_1 - \frac{z}{c} \right) \right] \right\} + i \frac{3\gamma^2 c \lambda^2 a^2 b}{8\hbar \Omega_1^2 \Omega_2} \times N_{of} \sin 2\psi (1 - \cos \Omega_1 T_1) \sin \Omega_2 \left(t - \tau - \frac{z}{c} \right), \quad (20)$$

$$\Omega_2^2 = (k v)^2 + \gamma \lambda b^2 / \hbar, \quad \tau \leq t - z/c \leq \tau + T_2,$$

where $j_1^*(T_1)$ is the complex-conjugate current (9) at the instant $t = T_1 + z/c$. Expression (20) is exact, and in (19) we have retained only the terms that contribute to the echo. In the region $\tau + T_2 \leq t - z/c$, the solution of (5) with $a_\alpha = 0$ is given by

$$j_\alpha(t - \tau - T_2 - z/c) = j_\alpha(T_2) \exp \{ i [\Phi_2 - \omega(\tau + T_2) - k v (t - \tau - T_2 - z/c)] \}, \quad (21)$$

where $j_\alpha(T_2)$ is the current of polarization of (19) and (20) at $t = \tau + T_2 + z/c$.

As seen from (21), (19), and (4), a photon echo having the polarization of the second pulse is produced and its amplitude of the echo is proportional to $\cos \psi$. We write out the polarization current of the right side of (1), which is responsible for the echo:

$$\int dv I = - \frac{\gamma^2 \lambda^2 c a b^2 N_0}{8\hbar} \cos \psi \int dv f \frac{1 - \cos \Omega_2 T_2}{\Omega_1 \Omega_2^2} \left[\frac{k v}{\Omega_1} (1 - \cos \Omega_1 T_1) - i \sin \Omega_1 T_1 \right] \exp \left\{ i \left[\omega \left(\frac{z}{c} - t \right) - k v \left(t - 2\tau + T_1 - T_2 - \frac{z}{c} \right) - \Phi_1 + 2\Phi_2 \right] \right\}. \quad (22)$$

With the aid of (1) and (22) we obtain the vector potential $A(z, t)$ of the photon echo outside a gaseous medium of dimension l :

$$A(z, t) = i 2\pi \lambda l \int dv I. \quad (23)$$

The complexity of the expression for the current I does not make it possible to evaluate (22) in analytic form. To calculate this integral, it is necessary to use the parameters of a concrete experiment. In the particular case when

$$T_0^{-2} = (k v)^2 \ll \gamma \lambda a^2 / \hbar, \quad (k v)^2 \ll \gamma \lambda b^2 / \hbar \quad (24)$$

the expression (23) for the vector potential of the photon echo simplifies to:²⁾

$$A(z, t) = - \frac{\pi}{4} (\hbar \lambda \gamma)^{1/2} \lambda N_0 \cos \psi \sin \Omega_1 T_1 (1 - \cos \Omega_2 T_2) \times \exp \left\{ - \frac{(t - 2\tau + T_1 - T_2 - z/c)^2}{4T_0^2} + i \left[\omega \left(\frac{z}{c} - t \right) - \Phi_1 + 2\Phi_2 \right] \right\} \quad (25)$$

As follows from (22) and (25), the concepts of 90° and 180° pulses have a definite meaning in a gaseous medium only in the particular case (24), when the quantity $k \cdot v$ in the frequencies Ω_1 and Ω_2 can be neglected. If the pulse duration is larger than or equal to the time of the reversible relaxation $T_0 = 1/kv$, then $\Omega_1 T_1$ and $\Omega_2 T_2$ depends significantly on the velocities of the atoms, and the concepts of 90° and 180° pulses become arbitrary.

The vector potential of the photon echo (23) or (25) gives rise to an increment of the polarization current (22). This increment can be estimated from formula (9), assuming approximately that the photon echo is a rectangular pulse with a time duration T_0 . It turns out that the addition caused by the photon-echo field is smaller than the main current (22) if $T_0 \ll T_1$. However, conservation of this addition leads to the occurrence of a second echo.

It is easy to show that the polarizations of the second signal and of the photon echo will be the same, as before, also in the case when the inequality (14) is not satisfied and it is necessary to take into account the reaction of the medium on the transmitted electromagnetic signals. However, this results in a complicated nonlinear equation for the echo amplitude.

All the final results for the atomic transition $J_2 = 1 \rightarrow J_1 = 0$ are obtained from the preceding formulas (22)–(25) by making the substitutions

$$\gamma \rightarrow 3\gamma, \quad N_0 \rightarrow n_2 / 3 - n_1.$$

3. ECHO ON ATOMIC TRANSITION WITH TWO DEGENERATE LEVELS

In view of the complexity of the formulas, we shall solve Eqs. (4)–(6) for the particular case $J_2 = 1 \rightarrow J_1$

²⁾The characteristic exponential factor (25) of the photon echo was obtained in a different manner in [6,7], without allowance for the degeneracy of the working levels. Allowance for the degeneracy changes the numerical values of Ω_1 and Ω_2 and of the pre-exponential factor.

= 1. In this approximation we obtain in lieu of (9)–(12)

$$i = -\frac{\gamma c \lambda a (n_2 - n_1) f}{8 \Omega_1 \hbar^2} \left\{ \frac{\mathbf{k}\mathbf{v}}{\Omega_1} \left[t - \cos \Omega_1 \left(t - \frac{z}{c} \right) \right] + i \sin \Omega_1 \left(t - \frac{z}{c} \right) \right\} J_1 J_\alpha \quad (26)$$

$$N_{\alpha 1} = \frac{(n_2 - n_1) f}{18 \hbar^2} \left\{ 1 - \frac{3 \gamma \lambda a^2}{2 \hbar \Omega_1^2} \left[1 - \cos \Omega_1 \left(t - \frac{z}{c} \right) \right] \right\} J_1 J_\alpha, \quad (27)$$

$$N_{\sigma 2} = \frac{(n_2 - n_1) f}{18 \hbar^2} \left\{ 1 - \frac{3 \gamma \lambda a^2}{4 \hbar \Omega_1^2} \left[1 - \cos \Omega_1 \left(t - \frac{z}{c} \right) \right] \right\} J_2 J_\sigma, \quad (28)$$

$$\sigma = 1, 2; \quad 0 \leq t - z/c \leq T_1; \quad \Omega_1^2 = (\mathbf{k}\mathbf{v})^2 + 3 \gamma \lambda a^2 / 2 \hbar,$$

where we use the relation $\hat{J}_\alpha^3 = \hat{J}_\alpha$, and all the physical quantities have been defined above. The other components of $N_{\alpha\beta}$ are not needed in explicit form.

The solution in the region $T_1 \leq t - z/c$ will be obtained in analogy with (15) and (16) by using (26)–(28) as the initial conditions, taken at the instant of time $t = T_1 + z/c$. After turning on the second pulse (17), it is convenient to change over to a coordinate system with the X axis along the polarization vector of this pulse. In this case, besides transforming the tensors, it is necessary to re-write the angular-momentum operators in a new representation, connected with the rotated coordinate system. As a result we obtain

$$\left(\frac{\partial}{\partial t} + i \mathbf{k}\mathbf{v} \right) j_1 + i \frac{9}{4} \gamma c \lambda b N_{11} = 0, \quad (29)$$

$$\frac{\partial}{\partial t} N_{11} + i \frac{b}{6 \hbar^2 c} (j_1 J_1^2 + J_1^2 j_1 - 2 j_1 J_1^2) = 0, \quad (30)$$

$$j_1(0) = j_0 e^{-i\varphi} (\hat{J}_1^2 \cos \psi + \hat{J}_2 \hat{J}_1 \sin \psi),$$

$$N_{11}(0) = B_1 \hat{J}_1^2 + B_2 \hat{J}_2 \hat{J}_1, \quad \varphi = \Phi_2 - \Phi_1 + \mathbf{k}\mathbf{v}(\tau - T_1),$$

where j_0 is the factor of the matrix (26) taken at the instant $t = T_1 + z/c$, and B_1 and B_2 are certain constants which are of no importance for the formation of the photon echo. Unlike the preceding gaseous medium with one degenerate level, the equation for $j_2(t - \tau - z/c)$ contains in this case the inhomogeneity

$$\left(\frac{\partial}{\partial t} + i \mathbf{k}\mathbf{v} \right) j_2 + i \frac{9}{4} \gamma c \lambda b N_{21} = 0, \quad (31)$$

$$\frac{\partial}{\partial t} N_{21} + i \frac{b}{6 \hbar^2 c} J_1^2 j_2 = i \frac{b}{6 \hbar^2 c} (2 j_1^+ - j_1) J_1 J_2, \quad (32)$$

$$j_2(0) = j_0 e^{-i\varphi} (\hat{J}_2^2 \sin \psi + \hat{J}_1 \hat{J}_2 \cos \psi), \quad N_{21}(0) = D_1 \hat{J}_2^2 + D_2 \hat{J}_1 \hat{J}_2,$$

where D_1 and D_2 are inessential constants. According to these equations, the diagonal matrix elements of the current j_2 are connected with the off-diagonal elements of the current j_1 . In other words, the diagonal elements of j_1 and j_2 are independent. Therefore the trace of j_2 is given by

$$\text{Sp } j_2 = c_1 + c_2 \exp(-i\varphi),$$

where c_1 and c_2 do not contain φ . Consequently, the current j_2 makes no contribution to the photon echo, which therefore acquires the polarization of the second pulse. The final form of the polarization current producing the photon echo is given by (3):

$$I_1 \left(t - \tau - T_2 - \frac{z}{c} \right) = j_1^+(T_1) \cos \psi \frac{3 \gamma \lambda b^2}{4 \hbar \Omega_2^2} (1 - \cos \Omega_2 T_2) \times \exp \left\{ i \left[\omega \left(\frac{z}{c} - t \right) - \mathbf{k}\mathbf{v} \left(t - 2\tau + T_1 - T_2 - \frac{z}{c} \right) - \Phi_1 + 2\Phi_2 \right] \right\},$$

$$\Omega_2^2 = (\mathbf{k}\mathbf{v})^2 + 3 \gamma \lambda b^2 / 2 \hbar,$$

where $j_1^+(T_1)$ is the Hermitian conjugate of the current (26), taken at the instant $t = \tau + T_2$ for $\alpha = 1$. A physically meaningful quantity is

$$\int d\mathbf{v} \text{Sp } I_1(t - \tau - T_2 - z/c). \quad (33)$$

It is easy to see that expression (33) coincides with formula (22), if we make in the latter the substitutions

$$\gamma \rightarrow 3\gamma/2, \quad N_0 \rightarrow 2(n_2 - n_1)/3.$$

4. PHOTON ECHO WITH CIRCULAR POLARIZATION

For concreteness, we consider the atomic transition $J_2 = 1 \rightarrow J_1 = 0$. We represent the current j_α in the form of a superposition of right- and left-polarized components

$$j_\alpha = l_+ a j_+ + l_- a j_-, \quad l_+ + l_- = 1.$$

Let the first signal have a right-circular polarization

$$A_\alpha = l_+ a \exp[i(kz - \omega t + \Phi_1)].$$

We then obtain from (5)–(8)

$$\left(\frac{\partial}{\partial t} + i \mathbf{k}\mathbf{v} \right) j_+ + i \frac{3}{4} \gamma c \lambda a N_{-+} = 0,$$

$$\frac{\partial}{\partial t} N_{-+} + i \frac{2a}{\hbar c} (j_+ - j_+^*) = 0,$$

$$j_+(z, 0) = 0, \quad N_{-+}(z, 0) = (n_2/3 - n_1) f \equiv N_{0f}, \quad (34)$$

$$\left(\frac{\partial}{\partial t} + i \mathbf{k}\mathbf{v} \right) j_- + i \frac{3}{4} \gamma c \lambda a N_{++} = 0,$$

$$\frac{\partial}{\partial t} N_{++} + i \frac{a}{\hbar c} j_- = 0,$$

$$j_-(z, 0) = 0, \quad N_{++}(z, 0) = 0. \quad (35)$$

We write out immediately the solution in the region

$$T_1 \leq t - z/c, \quad (ku)^2 \ll 3 \gamma \lambda a^2 / \hbar \equiv \Omega_1^2$$

after the passage of the first pulse

$$I_+ = -i^{1/4} (3 \hbar \gamma \lambda)^{1/2} c f N_0 \sin \Omega_1 T_1 \exp \{ i [\omega (z/c - t) - \mathbf{k}\mathbf{v} (t - T_1 - z/c) + \Phi_1] \},$$

$$N_{-+} = N_{0f} \cos \Omega_1 T_1, \quad N_{+-} = -N_{0f} \sin^2(\Omega_1 T_1 / 2),$$

$$I_- = N_- = N_{++} = 0.$$

If the second pulse has the same polarization

$$A_\alpha = l_+ a b \exp[i(kz - \omega t + \Phi_2)],$$

then the equations and initial conditions of (35) remain in force, and in lieu of (34) we get

$$N_{-+}(0) = N_{0f} \cos \Omega_1 T_1,$$

$$j_+(0) = -i^{1/4} (3 \hbar \gamma \lambda)^{1/2} c f N_0 \sin \Omega_1 T_1 e^{-i\varphi}. \quad (36)$$

We see that in the region

$$\tau \leq t - z/c \leq \tau + T_2 \quad (37)$$

the polarization current j_+ is expressed in the form of a linear combination of $\sin \varphi$ and $\cos \varphi$. This means that the current j_+ will contain terms proportional both to $e^{-i\varphi}$ and to $e^{i\varphi}$. The latter term leads to the occurrence of the photon echo.

If the first signal is right-polarized and the second is left-polarized, then the term with the phase factor $e^{i\varphi}$ can occur, in principle only in a current j_+ satisfying in the region (37) the equations

$$\left(\frac{\partial}{\partial t} + ikv\right)j_+ + i\frac{3}{4}\gamma c\lambda bN_{--} = 0,$$

$$\frac{\partial}{\partial t}N_{--} + i\frac{b}{\hbar c}j_+ = 0,$$

with initial condition $N_{--}(0) = 0$ and (36). In fact, we obtain

$$j_+(t - \tau - z/c) \sim e^{-i\varphi}.$$

Consequently, left-polarized second signal produces no photon echo.

Other variants, for example pulses with linear and circular polarizations, can be investigated analogously. In each case, the photon echo on the atomic transitions $0 \rightleftharpoons 1$ and $1 \rightarrow 1$ has the polarization of the second signal.

5. ECHO ON ATOMIC TRANSITION $J_2 = 1/2 \rightarrow J_1 = 1/2$

The atomic transition $J_2 = 1/2 \rightarrow J_1 = 1/2$ is distinguished from all others in that it leads to a resolution of a linearly polarized wave into independent circular waves with right and left polarizations. Therefore it is convenient to solve the photon-echo problem in this case in terms of circular waves. The amplitude a_α of a linearly polarized first pulse (2) and the current j_α will be written in the form

$$a_\alpha = l_{+\alpha}a_+ + l_{-\alpha}a_-,$$

$$j_\alpha = l_{+\alpha}j_+ + l_{-\alpha}j_-,$$

$$a_+ = a_- = \frac{a}{\sqrt{2}} \quad j_+ = \begin{pmatrix} j_{++} & 0 \\ j_{+-} & 0 \end{pmatrix}, \quad j_- = \begin{pmatrix} 0 & j_{+-} \\ 0 & j_{--} \end{pmatrix}.$$

The matrix-element indices plus and minus denote upper-level momentum projections $+1/2$ and $-1/2$, respectively. It follows from (5) and (6) that the off-diagonal matrix elements of the currents j_+ and j_- comprise a system of mutually coupled equations. At the same time, the diagonal matrix elements form two independent systems of equations for the right-circularly polarized wave:

$$\left(\frac{\partial}{\partial t} + ikv\right)j_{++} + i\frac{3}{2}\gamma c\lambda a_+N_{++} = 0, \tag{38}$$

$$\frac{\partial}{\partial t}N_{++} + i\frac{2}{3\hbar c}(a_+^*j_{++} - a_+j_{++}^*) = 0 \tag{39}$$

and for the left-circularly polarized wave:

$$\left(\frac{\partial}{\partial t} + ikv\right)j_{--} + i\frac{3}{2}\gamma c\lambda a_-N_{--} = 0, \tag{40}$$

$$\frac{\partial}{\partial t}N_{--} + i\frac{2}{3\hbar c}(a_-^*j_{--} - a_-j_{--}^*) = 0 \tag{41}$$

with initial conditions

$$j_{++}(0) = j_{--}(0) = 0$$

$$N_{++}(0) = N_{--}(0) = (n_2 - n_1)f/6.$$

The solution of the equations after the passage of the first pulse is given by

$$I_{++}(t - T_1 - z/c) = I_{--}(t - T_1 - z/c)$$

$$= j_{++}(T_1) \exp\{i[\omega(z/c - t) - kv(t - T_1 - z/c) + \Phi_1]\},$$

$$j_{++}(T_1) = -\frac{\gamma c\lambda a(n_2 - n_1)f}{4\sqrt{2}\Omega_1} \left[\frac{kv}{\Omega_1}(1 - \cos \Omega_1 T_1) + i \sin \Omega_1 T_1 \right],$$

$$N_{++} = N_{--} = \frac{n_2 - n_1}{6} f \left[1 - \frac{\gamma\lambda a^2}{\hbar\Omega_1^2}(1 - \cos \Omega_1 T_1) \right], \tag{42}$$

$$\Omega_1^2 = (kv)^2 + \gamma\lambda a^2/\hbar, \quad T_1 \leq t - z/c.$$

If the second pulse (17) is linearly polarized at an angle relative to the first, then it is necessary to make in (38)–(41) in the region of (37) the following substitutions:

$$a_+ \rightarrow (b/\sqrt{2})e^{i\psi}, \quad a_- \rightarrow (b/\sqrt{2})e^{-i\psi}.$$

As the initial conditions for $N_{++}(0)$ and $N_{--}(0)$ it is necessary to take (42), and for the currents the initial conditions are

$$j_{++}(0) = j_{+-}(T_1)e^{-i\varphi}, \quad j_{--}(0) = j_{+-}(T_1)e^{-i\varphi},$$

where φ was defined above.

We emphasize that the solution of the problem in terms of linear polarization would lead in the region (37) to the system of equations (29)–(32) in which the following substitutions are made:

$$\gamma \rightarrow 2\gamma/3, \quad f \rightarrow 3f/2, \quad \hat{j}_\alpha \rightarrow \hbar\sigma_\alpha.$$

By virtue of the singular properties of the Pauli matrices σ_α , the diagonal elements of the mutually perpendicular polarization currents j_1 and j_2 will now be connected by Eqs. (29)–(32).

We write out the final solution of the equations in the region

$$\tau + T_2 \leq t - z/c$$

after the passage of the second pulse:

$$I_{++}\left(t - \tau - T_2 - \frac{z}{c}\right) = j_{++}^*(T_1) \frac{\gamma\lambda b^2}{2\hbar\Omega_2^2} (1 - \cos \Omega_2 T_2)$$

$$\times \exp\left\{i\left[\omega\left(\frac{z}{c} - t\right) - kv(t - 2\tau + T_1 - T_2) - \Phi_1 + 2(\Phi_2 + \psi)\right]\right\}, \quad \Omega_2^2 = (kv)^2 + \gamma\lambda b^2/\hbar. \tag{43}$$

The solution for the left-polarized current is obtained from (43) by making the substitution $\psi \rightarrow -\psi$. As seen from (43), the left- and right-polarized components of the photon echo on emerging from the gaseous medium add up together to form a pulse which is linearly polarized at an angle 2ψ relative to the initial pulse, and the amplitude of the echo does not depend at all on the angle ψ . A similar phenomenon was observed in ruby.^[2,3]

If the first and second pulses have respectively linear or circular polarizations (or vice versa), then the photon echo on the atomic transition $J_2 = 1/2 \rightarrow J_1 = 1/2$ is always circularly polarized in a direction opposite to that of the other atomic transitions.

The authors are grateful to V. M. Galitskiĭ for continuous interest in the work and useful discussions, and also to A. A. Manykin for a discussion.

¹C. K. N. Patel and R. E. Slusher, Phys. Rev. Lett. **20**, 1087 (1968).

²N. A. Kurnit, I. D. Abella, and S. R. Hartman, Phys. Rev. Lett. **13**, 567 (1964).

³I. D. Abella, N. A. Kurnit, and S. R. Hartman, Phys. Rev. **141**, 391 (1966).

⁴A. N. Oraevskiĭ, Usp. Fiz. Nauk **91**, 181 (1967) [Sov. Phys.-Usp. **10**, 45 (1967/68)].

⁵C. K. N. Patel and R. E. Slusher, Phys. Rev. Lett.

19, 1019 (1967).

⁶E. A. Manykin, ZhETF Pis. Red. 7, 345 (1968)
[JETP Lett. 7, 269 (1968)].

⁷M. Scully, M. J. Stephen, and D. C. Burnham, Phys.

Rev. 171, 213 (1968).

Translated by J. G. Adashko
245