PHOTON ECHO IN A GAS IN THE PRESENCE OF A MAGNETIC FIELD

A. I. ALEKSEEV and I. V. EVSEEV

Moscow Engineering Physics Institute

Submitted June 10, 1969

Zh. Eksp. Teor. Fiz. 57, 1735–1744 (November, 1969)

Some features of photon echo on atomic transitions involving total momentum changes $1/2 \rightleftarrows 3/2$, $1 \to 1$ and $1 \rightleftarrows 2$ in the presence of an external magnetic field **H** are investigated. The intensity and polarization of a photon echo produced in a gas by the passage of two linearly polarized light pulses are determined. It is shown that a magnetic field affects the photon-echo intensity and leads to a specific rotation of its polarization. This rotation of polarization differs from Faraday rotation since the angle of rotation does not depend on the distance travelled by the light but on H and on the time between the last two pulses. The angle of rotation of polarization of the photon echo also depends on the type of the atomic transition. It is suggested that the g-factors of the energy levels be measured on basis of the polarization rotation angle and variation of photon echo intensity. The specific polarization properties of the photon echo can be used for experimental identification of atomic transitions.

THE photon-echo method is a very sensitive tool for the experimental investigation of relaxation processes in solids[1-3] and in gases[4,5]. The method consists of successively passing through the investigated medium two exciting light pulses, separated in time by an amount τ. The photon echo is produced in the medium at the time 2τ following the passage of the first pulse. The relaxation time is determined from the attenuation of the photon-echo intensity as a function of τ.

This, however, does not exhaust all the possibilities of the photon-echo method. An experimental investigation of the polarization effects of the photon echo in a gas makes it also possible to determine other physical characteristics of the gas molecules. The polarization properties of the photon echo are of particular interest in the presence of an external magnetic field[6].

The rotation of the polarization of the photon echo in a magnetic field was recently observed experimentally without detailed quantitative measurements[5]. The observed rotation of the polarization was interpreted in[5] as ordinary Faraday rotation. Such an interpretation cannot be regarded as valid. The Faraday effect is due to the interaction between the transmitted wave and a medium placed in a magnetic field. Therefore the rotation angle in the case of Faraday rotation is proportional to the excess population of the working levels and to the path traversed by the light[7]. In a gas, the Faraday angles of the transmitted pulses and of the photon echo are relatively small. This is due to the small value of the overpopulation, since in a gas both working levels are usually excited and their excess population is due to the Boltzmann distribution (see the Appendix).

If the Faraday effect is neglected, then there is no rotation of the polarization of the photon echo at all as the pulse passes through the medium. The photon echo in a gas is induced in the presence of a magnetic field at a definite instant when the polarization is already rotated, even if the polarizations of both excited pulses coincide. The direction of this rotation opposes the Faraday rotation in a monochromatic wave passing through the same medium. The angle of rotation of the photon-echo polarization relative to the polarization of the transmitted pulses does not depend on the excess level population and on the linear dimensions of the medium, but is determined by the quantities τ and H, and by the gyromagnetic factors of the levels and the type of atomic transition. The physical cause of the indicated rotation is the precession of the polarization current around H after the passage of the exciting light pulses. It follows from the foregoing that when there is no Faraday rotation, the rotation of the photon-echo polarization vector in a magnetic field relative to the polarization of the transmitted pulses is a new physical effect. This effect was predicted by one of the authors in[6], where photon echo in the atomic transitions $1 \rightleftarrows 0$ and $1/2 \to 1/2$ was observed in the presence of a magnetic field.

In the general case, it is necessary to add to the aforementioned specific rotation of the photon-echo polarization also the contribution from the Faraday rotation.

In the absence of an external magnetic field, the polarization properties of the photon echo in different atomic transitions are quite distinct. If the exciting pulses move in the same direction and are polarized at an angle ψ to each other, then the polarization of the photon echo in the atomic transitions $1/2 \rightleftarrows 3/2$ at $H = 0$ makes an angle larger than ψ with the polarization of the first pulse. At the same time, the polarization of the photon echo in the atomic transitions $1 \rightleftarrows 2$ does not obey this rule and depends on a very complicated manner on ψ and on the other parameters of the problem.

Since the polarization of electromagnetic waves is characterized by a vector, the polarization features of the photon echo must be described with allowance for the degeneracy of the resident energy levels. We assume henceforth that the degeneracy of the levels is due to the different orientations of the total angular momentum.

1. FUNDAMENTAL EQUATIONS

We consider an atom (molecule) in a homogeneous magnetic field $\mathbf{H}$. The Zeeman splitting of the degenerate level, characterized by the aggregrate of quantum numbers n and the total angular momentum J, is given in the approximation linear in H by

$$\Delta E_{nJM} = \mu_0 g_{nJ} HM, \tag{1}$$

where μ_0 is the Bohr magneton and M is the projection of the total angular momentum. In the case of an arbitrary type of coupling, the gyromagnetic factor g_{nJ} is a certain coefficient (g-factor) characterizing the given term.

We write the equation for the density matrix $\rho_{nJm,n'J'M'}$ of a group of atoms moving with velocity v in a form in which definite values are possessed by the quantum numbers n, J, and M as well as by the energy E_{nJM}:

$$i\hbar\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right)\rho_{nJM,\,n'J'M'} = (E_{nJM} - E_{n'J'M'})\,\rho_{nJM,\,n'J'M'}$$
$$+ \frac{1}{c}\,(\rho_{nJM,\,n''J''M''}\,\mathbf{d}_{n''J''M'',\,n'J'M'}$$
$$- \mathbf{d}_{nJM,\,n''J''M''}\,\rho_{n''J''M'',\,n'J'M'})\,(\mathbf{A} + \mathbf{A}^*),$$
$$E_{nJM} = E_{nJ} + \mu_0 g_{nJ} HM,$$

where $d_{nJM,n'J'M'}$ is the dipole moment of the transition, and A is the vector potential of the electromagnetic pulse[1)].

We consider only two groups of sublevels with energies $E_{n_0J_2M}$ and $E_{n_0J_1M}$. In the absence of a magnetic field, they merge to form two degenerate levels $E_{n_0J_2}$ and $E_{n_0J_1}$, which are in resonance with the frequency of the transmitted electromagnetic pulses:

$$E_{n_0J_2} - E_{n_0J_1} = \hbar\omega.$$

Here J_2 and J_1 are the total angular momenta of the upper and lower degenerate levels, respectively.

For simplicity, we assume that the Zeeman splitting (1) is small compared with $\hbar\omega$. We denote by

$$\rho_{n_0J_2M,\,n_0J_2M'} \equiv \rho_{\mu\mu'}$$

the density matrix describing the transitions between the Zeeman sublevels of the upper level. Analogously,

$$\rho_{n_0J_1M,\,n_0J_1M'} \equiv \rho_{mm'}$$

is the density matrix of the Zeeman sublevels of the lower level. The transitions between the Zeeman sublevels of the upper and lower levels are described by the density matrix

$$\rho_{n_0J_2M,\,n_0J_1M'} \equiv R_{\mu m}.$$

The vector potential

$$A_\alpha = a_\alpha \exp\left[i(\mathbf{kr} - \omega t + \Phi)\right] \tag{2}$$

of the transmitted electromagnetic pulse satisfies the d'Alembert equation

$$\Box A_\alpha = -\frac{4\pi}{c}\int d\mathbf{v}\,\mathrm{Sp}\,I_\alpha, \tag{3}$$

in which the polarization current I_α is connected with the density matrix $R_{\mu m}$ in the following manner:

$$I_\alpha = R_{\mu m}\,\dot{d}^{\alpha}_{m\mu'}, \tag{4}$$

where the dot over the dipole moment of the transition $\mathbf{d}_{m\mu}$ denotes the operator of the derivative of this quantity. It is convenient to separate in the current (4) the slowly-varying amplitude

$$I_\alpha = j_\alpha \exp\left[i(\mathbf{kr} - \omega t + \Phi)\right]. \tag{5}$$

In (2) and (5), the phase Φ is real and constant. The alternating phase difference between the current and the vector potential, which appears in the problem, will be assigned to j_α. Then the equations for the slow functions take the form

$$\left(\frac{\partial}{\partial t} + c\nabla\right)a_\alpha = i\cdot 2\pi\lambdabar\int d\mathbf{v}\,\mathrm{Sp}\,j_\alpha, \tag{6}$$

$$\left(\frac{\partial}{\partial t} + i\mathbf{kv}\right)j_\alpha + i\frac{\varepsilon_2}{\hbar}(\mathbf{n}\hat{\mathbf{J}}_2)\,j_\alpha - i\varepsilon_1 j_\sigma Q_{\sigma\alpha}$$
$$+ {}^3/_4 i\,(2J_2 + 1)\,\gamma c\lambdabar\,(\rho_2 T_{\alpha\beta} - \rho_1^{\alpha\beta})\,a_\beta = 0, \tag{7}$$

$$\frac{\partial}{\partial t}\rho_2 - i\frac{\varepsilon_2}{\hbar}[\rho_2(\mathbf{n}\hat{\mathbf{J}}_2) - (\mathbf{n}\hat{\mathbf{J}}_2)\rho_2] + i\frac{1}{\hbar c}(a_\sigma^* j_\sigma - a_\sigma j_\sigma^+) = 0, \tag{8}$$

$$\frac{\partial}{\partial t}\rho_1^{\alpha\beta} - i\varepsilon_1(\rho_1^{\sigma\beta}Q_{\sigma\alpha} - Q_{\beta\sigma}\rho_1^{\alpha\sigma}) - i\frac{1}{\hbar c}(a_\sigma^* T_{\sigma\beta} j_\alpha - j_\beta^+ T_{\alpha\sigma} a_\sigma) = 0, \tag{9}$$

where

$$Q_{\alpha\beta} = (2J_1 + 1)\,d^{\alpha}_{\mu m}(\mathbf{n}\hat{\mathbf{J}}_1)_{mm'}\,d^{\beta}_{m'\mu'}/\hbar\,|d^{J_2}_{J_1}|^2,$$
$$T_{\alpha\beta} = d_{\mu m}{}^{\beta} d_{m\mu'}{}^{\alpha} / |d_{J_1}{}^{J_2}|^2, \tag{10}$$

$$\rho_1^{\alpha\beta} = d^{\beta}_{\mu m}\,\rho_{mm'}\,d^{\alpha}_{m'\mu'}/|d_{J_1}{}^{J_2}|^2,$$
$$\rho_2 = \rho_{\mu\mu'}, \quad \mathbf{n} = \mathbf{H}/H, \quad \lambdabar = c/\omega = 1/k,$$
$$\varepsilon_1 = \mu_0 g_1 H/\hbar, \quad \varepsilon_2 = \mu_0 g_2 H/\hbar, \quad \gamma = 4|d_{J_1}{}^{J_2}|^2/3(2J_2 + 1)\hbar\lambdabar^3. \tag{11}$$

Here $d^{J_2}_{J_1}$ is the reduced dipole moment of the transition, γ is the probability of spontaneous emission, and $\hat{\mathbf{J}}_1$ and g_1 are respectively the operator of the total angular momentum and the gyromagnetic factor of the lower level. $\hat{\mathbf{J}}_2$ and g_2 are the analogous quantities for the upper level. The term $\mathbf{k}\cdot\mathbf{v}$ takes into account the Doppler frequency shift occurring when an atom moves with velocity v. The derivatives $\mathbf{v}\nabla$ of the slow functions have been omitted from Eqs. (7)–(9), since in the approximation considered below they make a negligibly small contribution, of the order of v/c.

For the different atomic transitions we obtain:

for $J_2 = J \to J_1 = J$

$$Q_{\alpha\beta} = \hat{J}_\alpha(\mathbf{n}\hat{\mathbf{J}})\hat{J}_\beta / J(J+1)\hbar^3, \quad T_{\alpha\beta} = \hat{J}_\beta\hat{J}_\alpha / J(J+1)(2J+1)\hbar^2;$$

for $J_2 = J \to J_1 = J + 1$

$$Q_{\alpha\beta} = \{2(J+1)^2(\mathbf{n}\hat{\mathbf{J}})\delta_{\alpha\beta} + (J+1)\,[(\hat{J}_\alpha\hat{J}_\beta + \hat{J}_\beta\hat{J}_\alpha)(\mathbf{n}\hat{\mathbf{J}}) +$$
$$+ (\mathbf{n}\hat{\mathbf{J}})(\hat{J}_\alpha\hat{J}_\beta + \hat{J}_\beta\hat{J}_\alpha)] - (2J+3)\,[\hat{J}_\alpha(\mathbf{n}\hat{\mathbf{J}})\hat{J}_\beta + \hat{J}_\beta(\mathbf{n}\hat{\mathbf{J}})\hat{J}_\alpha] -$$
$$- i(J+2)\,[(J+1)e_{\alpha\beta\gamma}n_\gamma + e_{\alpha\beta\gamma}(\hat{J}_\gamma\mathbf{n}\hat{\mathbf{J}} + \mathbf{n}\hat{\mathbf{J}}\hat{J}_\gamma)]\} / 2(J+1)(2J+1)\hbar^3,$$
$$T_{\alpha\beta} = \frac{i(2J+3)\hbar e_{\alpha\beta\gamma}\hat{J}_\gamma + 2(J+1)^2\hbar^2\delta_{\alpha\beta} - \hat{J}_\alpha\hat{J}_\beta - \hat{J}_\beta\hat{J}_\alpha}{2(J+1)(2J+1)(2J+3)\hbar^2}.$$

The transformation (4), (10), and (11) effects the transition to the matrix indices of the projection operator of the total angular momentum of the upper level. This is a very convenient factor in the analysis of the atomic transitions $J_2 = J \to J_1 = J + 1$ and also $J_2 = J \to J_1 = J$. In the case $J_2 = J + 1 \to J_1 = J$, to the contrary, it is convenient to change over to the matrix indices of the projected operator of the total angular momentum of the lower level, introducing the notation

$$j_\alpha = R_{m\mu}\,\dot{d}^{\alpha}_{\mu m'}\exp[i(\mathbf{kr} - \omega t + \Phi)],$$

$$\rho_1 = \rho_{mm'}, \quad \rho_2^{\alpha\beta} = d^{\beta}_{m\mu}\,\rho_{\mu\mu'}\,d^{\alpha}_{\mu'm'}/|d_{J_1}{}^{J_2}|^2.$$

The equation for these quantities and for a_α^* are ob-

[1)] Summation over the repeated matrix and tensor indices is implied throughout.

tained from (6)—(9) by renumbering the indices of the density matrices ($1 \rightleftarrows 2$), the g factors, and the total angular momentum operators. The amplitude $\mathbf{a}$ is replaced everywhere by its complex conjugate $\mathbf{a}^*$, and vice-versa: $\mathbf{a} \rightleftarrows \mathbf{a}^*$, and minus signs appear in the right side of (6) and in front of the wave vector $\mathbf{k}$ in (7). The quantities $Q_{\alpha\beta}$ and $T_{\alpha\beta}$ for the atomic transition $J_2 = J + 1 \rightarrow J_1 = J$ coincide formally with the expressions obtained above for the atomic transition $J_2 = J \rightarrow J_1 = J + 1$.

We note that the photon echo in a gas is formed most frequently in atomic transitions between two excited levels, whose total angular momenta can assume arbitrary values. The transition to the matrices of the smaller angular momentum is particularly useful in the consideration of the atomic transitions $1/2 \rightleftarrows 3/2$ and $1 \rightleftarrows 2$, since the basic matrices in the space of 2×2 and 3×3 matrices are quite simple.

The initial conditions for Eqs. (6)—(9) are written in the following form:

$$a_\alpha(\mathbf{r}, 0) = j_\alpha(\mathbf{r}, 0) = 0, \quad \rho_2(\mathbf{r}, 0) = \frac{n_2 f}{2J_2 + 1}, \quad \rho_1^{\alpha\beta}(\mathbf{r}, 0) = \frac{n_1 f T_{\alpha\beta}}{2J_1 + 1} \qquad (12)$$

where n_2 and n_1 are the densities of the atoms at the sublevels of the upper and lower levels, respectively, at the initial instant $t = 0$. The numerical value of n_2 or n_1 is determined by the Boltzmann distribution of the atoms over the levels. The function f describes the velocity distribution of the atoms:

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

where u is the thermal velocity of the atom. We have neglected Boltzmann distribution of the atoms over the Zeeman sublevels at the initial instant $t = 0$, assuming the distribution over the sublevels in (12) to be equally probable. In all formulas, the unit matrix is not written out in explicit form.

2. PHOTON ECHO IN THE ATOMIC TRANSITION $1/2 \rightarrow 3/2$

By virtue of the specific properties of the Pauli matrices σ_α, Eqs. (6)—(9) for the atomic transition $1/2 \rightarrow 3/2$ simplify noticeably, inasmuch as the quantities $Q_{\alpha\beta}$ and $T_{\alpha\beta}$ assume a relatively simple form:

$$Q_{\alpha\beta} = [4(\mathbf{n}\sigma)\delta_{\alpha\beta} - n_\alpha\sigma_\beta - \sigma_\alpha n_\beta - 5ie_{\alpha\beta\gamma}n_\gamma] / 6,$$
$$T_{\alpha\beta} = (\delta_{\alpha\beta} + \sigma_\alpha\sigma_\beta) / 12.$$

Let the exciting pulses propagate in the direction of the magnetic field $\mathbf{H}$, which we choose to be the Z axis. The first pulse, which is linearly polarized along the X axis, falls on the boundary of the medium $z = 0$ at the instant of time $t = 0$.

We shall henceforth assume that the time interval τ between two transmitted pulses is large compared with the durations of the pulses themselves and of the photon echo. Let the Faraday rotation be insignificant and let the influence of the magnetic field during the time of passage of the light pulses be negligible. We seek an approximate solution of the problem, neglecting the reaction of the medium on the transmitted light pulse. To this end it is necessary to have[8]

$$N_0 \lambda\!\!\!{}^{-2} l \gamma T_0 \ll 1, \qquad (13)$$

where N_0 is the excess population of the working levels, l the linear dimension of the medium, and $\hbar/T_0$ the Doppler width of the spectral line.

We emphasize that the polarization effects of the photon echo in a magnetic field do not depend on the inequality (13) and do not change when the reaction of the medium on the transmitted light pulses is taken rigorously into account. This is connected with the fact that the polarization current induced by the passing pulse serves only as an initial condition for the subsequent solution of Eq. (7) with $\mathbf{a} \equiv 0$ in the time interval following the passage of the indicated pulse. The photon echo is the result of phase synchronization of individual emitters precisely in the time interval when there is no electromagnetic field ($\mathbf{a} \equiv 0$), and the equation for the current with allowance for $\mathbf{H}$ can be solved exactly.

The solution of Eqs. (7)—(9) during the time of passage of the first pulse of duration T_1 is written in the form

$$j_\alpha = (\delta_{1\alpha} + \sigma_\alpha\sigma_1) j,$$
$$j\left(t - \frac{z}{c}\right) = -\frac{\gamma c \lambda\!\!\!{}^{-} a}{8\Omega_1} N_0 f \left\{\frac{\mathbf{kv}}{\Omega_1}\left[1 - \cos\Omega_1\left(t - \frac{z}{c}\right)\right] + i\sin\Omega_1\left(t - \frac{z}{c}\right)\right\},$$
$$\Omega_1^2 = (\mathbf{kv})^2 + \gamma\lambda\!\!\!{}^{-} a^2/\hbar, \qquad N_0 = (n_2 - n_1/2)/2. \qquad (14)$$

We need no explicit expression for the density matrices.

After the passage of the first pulse $T_1 \leq t - z/c$, the polarization current (5) takes the form

$$I_\alpha = I_\alpha^a j(T_1) \exp\{i[\omega(z/c - t) - \mathbf{kv}(t - T_1 - z/c) + \Phi_1]\}, \quad (15)$$

where $j(T_1)$ is the current (14) taken at the instant $t = T_1 + z/c$; Φ_1 is the constant phase of the first pulse of the type (2), and I_α^a is the matrix vector:

$$I_1^a = \frac{1}{2}\begin{pmatrix} 3e^{i\alpha} + e^{-i\beta} & 0 \\ 0 & 3e^{-i\alpha} + e^{i\beta} \end{pmatrix},$$

$$I_2^a = \frac{i}{2}\begin{pmatrix} e^{-i\beta} - 3e^{i\alpha} & 0 \\ 0 & 3e^{-i\alpha} - e^{i\beta} \end{pmatrix}, \quad I_3^a = \begin{pmatrix} 0 & e^{-i\beta} \\ -e^{i\beta} & 0 \end{pmatrix},$$

$$\alpha = (3\varepsilon_1 - \varepsilon_2)(t - T_1 - z/c)/2, \quad \beta = (\varepsilon_1 + \varepsilon_2)(t - T_1 - z/c)/2.$$

It is easy to see that the trace of I_α^a is equal to the sum of two vectors, which rotate at each point z with angular velocities $(3\epsilon_1 - \epsilon_2)/2$ and $(\epsilon_1 + \epsilon_2)/2$. If the g-factors of the levels are the same, $\epsilon_1 = \epsilon_2 = \epsilon$, then the aforementioned two vectors merge into a single vector rotating with angular velocity ϵ.

Assume that at the instant $t = \tau$, there is sent along the Z axis a second pulse with amplitude b, time duration T_2, and a polarization vector that makes an angle ψ with the polarization of the first pulse:

$$A_\alpha = b_\alpha \exp[i(kz - \omega t + \Phi_2)], \quad \Phi_2 = \text{const}.$$

We rotate the coordinate system in such a way that X the axis coincides with the direction of polarization of the second pulse. This does not change Eqs. (6)—(9). The initial conditions for the current

$$I_\alpha = j_\alpha \exp[i(kz - \omega t + \Phi_2)] \qquad (16)$$

are determined from (15) by transforming the rotation to the instant $t = \tau + z/c$. All the formulas in the region $\tau \leq t - z/c$ will be sought henceforth in the rotated coordinate system with the X axis along the polarization vector of the second pulse.

In the region

$$\tau \leqslant t - z/c \leqslant \tau + T_2 \quad (17)$$

we are interested only in that part of the current which makes a contribution to the photon echo. The terms of the current that are proportional to the initial values of the density matrices ρ_2 and $\rho_1^{\alpha\beta}$, taken at the instant $t = \tau + z/c$, make no contribution to the photon echo. Therefore Eqs. (6)–(9) must be solved in the region (17) with zero initial conditions for the density matrices. The final expression of the sought part of the current (16), making a contribution to the photon echo, takes the form

$$I_\alpha = 2^{-3} j^*(T_1)[1 - \cos\Omega_2(t - \tau - z/c)] I_\alpha^b \times \exp\{i[\omega(z/c - t) + \mathbf{kv}(\tau - T_1) - \Phi_1 + 2\Phi_2]\},$$

$$I_1^b = (2x + iy\sigma_z), \quad I_2^b = y - ix\sigma_z, \quad I_3^b = i(y\sigma_x + x\sigma_y),$$

$$x = \cos(\varphi_1 + \psi) + 3\cos(\varphi_2 + \psi), \quad y = \sin(\varphi_1 + \psi) - 3\sin(\varphi_2 + \psi),$$
$$\varphi_1 = (\varepsilon_1 + \varepsilon_2)\tau/2, \quad \varphi_2 = (3\varepsilon_1 - \varepsilon_2)\tau/2, \quad \Omega_2^2 = \gamma\lambda b^2/\hbar. \quad (18)$$

In order to simplify formula (18), we have carried out an expansion in terms of the parameter

$$1/(\Omega_2 T_0)^2 \ll 1, \quad (19)$$

retaining the first term of the expansion.

The value of the current (18) at the instant $t = \tau + T_2 + z/c$ is the initial condition in the solution of (7) with $a \equiv 0$ in the region after the passage of the second pulse:

$$\tau + T_2 \leqslant t - z/c. \quad (20)$$

We write out immediately the trace of the current (4) that enters in equation (3):

$$\mathrm{Sp}\, I_\alpha(t - \tau - T_2 - z/c) = 2^{-3} j^*(T_1)(1 - \cos\Omega_2 T_2) I_\alpha^e \times \exp\{i[\omega(z/c - t) - \mathbf{kv}(t - 2\tau + T_1 - T_2 - z/c) - \Phi_1 + 2\Phi_2]\}, \quad (21)$$
$$I_1^e = \cos\psi_1 + 3\cos\psi_2 + 3\cos\psi_3 + 9\cos\psi_4,$$
$$I_2^e = -\sin\psi_1 + 3\sin\psi_2 + 3\sin\psi_3 - 9\sin\psi_4,$$
$$I_3^e = 0,$$
$$\psi_1 = \varphi_1 + \psi - (\varepsilon_1 + \varepsilon_2)(t - \tau - T_2 - z/c)/2,$$
$$\psi_2 = \varphi_1 + \psi + (3\varepsilon_1 - \varepsilon_2)(t - \tau - T_2 - z/c)/2,$$
$$\psi_3 = \varphi_2 + \psi + (\varepsilon_1 + \varepsilon_2)(t - \tau - T_2 - z/c)/2,$$
$$\psi_4 = \varphi_2 + \psi - (3\varepsilon_1 - \varepsilon_2)(t - \tau - T_2 - z/c)/2.$$

Owing to the Maxwellian distribution function that enters in $j(T_1)$, the integration of (21) with respect to the velocity causes the photon echo in the medium to occur at the instant $t = 2\tau - T_1 + T_2 - z/c$. The terms T_1, T_2, and z/c can usually be neglected in comparison with 2τ, so that the instant of occurrence of the photon echo is approximately equal to $t = 2\tau$. The order of magnitude of the duration of the photon echo pulse is $T_0 \equiv 1/ku$. The direction of the polarization current (21) at the instant of occurrence of the photon echo, $t = 2\tau - T_1 + T_2 + z/c$, is determined by the vector $I_\alpha^e(\tau)$:

$$I_1^e(\tau) = 5\cos\psi + 3\cos(\psi + 2\varepsilon_1\tau), \quad (22)$$

$$I_2^e(\tau) = -5\sin\psi + 3\sin(\psi + 2\varepsilon_1\tau), \quad I_3^e(\tau) = 0, \quad (23)$$

where we have neglected the term T_1 compared with τ.

We write out the vector potential of the photon echo in the particular case

$$1/(\Omega_1 T_0)^2 \ll 1 \quad (24)$$

with simultaneous satisfaction of the inequality (19):

$$A_\alpha(z, t) = -\frac{\pi}{16} I_\alpha^e \lambda l(\hbar\lambda\gamma)^{1/2} N_0 \sin\Omega_1 T_1(1 - \cos\Omega_2 T_2) \times \exp\left\{-\frac{(t - 2\tau + T_1 - T_2 - z/c)^2}{4T_0^2} + i\left[\omega\left(\frac{z}{c} - t\right) - \Phi_1 + 2\Phi_2\right]\right\}. \quad (25)$$

The photon-echo polarization vector at the point of the maximum intensity coincides with $I_\alpha^e(\tau)$. In the absence of the magnetic field it makes an angle larger than ψ with the polarization vector of the first pulse. The presence of a magnetic field leads to rotation of the polarization of the photon echo in a clockwise direction, when viewed along the pulse propagation direction. It is easy to determine the g-factor of the lower level from the angle of this rotation.

As seen from (22) and (23), the intensity of the photon echo also depends on the angle of rotation of polarization of the photon echo in the magnetic field. For the particular case $\psi = 0$, the intensity of the photon echo in a magnetic field is proportional to the factor $17 + 15\cos 2\epsilon_1\tau$. Consequently, it is also possible to determine the gyromagnetic factor of the lower level from the change of intensity of the proton echo as a function of τ and H.

So far we have ignored the attenuation of the photon-echo amplitude as the result of the irreversible relaxation. The latter is taken into account in (6) and (7), as usual, by introducing relaxation terms. As a result, the intensity of the photon echo acquires the characteristic factor

$$\exp[-\Gamma(2\tau - T_1 + T_2],$$

where $\hbar\Gamma$ is the sum of the widths of the upper and lower working levels[9].

In the case of the atomic transition $J_2 = 3/2 \to J_1 = 1/2$, the vector potential of the photon echo is obtained from (25) by the substitution

$$\gamma \to 2\gamma, \quad N_0 \to (n_2/2 - n_1)/2, \quad \varepsilon_1 \rightleftarrows \varepsilon_2.$$

3. PHOTON ECHO IN THE ATOMIC TRANSITION 1 → 1

After passage of the first pulse, the magnetic field rotates the polarization current. Unlike the atomic transition $1/2 \to 3/2$, the trace of the polarization current in the case of the $1 \to 1$ transition is equal to a sum of two vectors rotating with angular velocities ϵ_1 and ϵ_2.

The subsequent calculation procedure is similar to that considered above, although the calculations are much more complicated on changing over to the unity-angular-momentum matrices. The matrix structure of the polarization current becomes particularly complicated after the passage of the second pulse. However, the trace of the polarization current breaks up, as before, into a sum of two vectors rotating with angular velocities ϵ_1 and ϵ_2. Omitting the rather laborious calculations, we present the final result for the photon-echo vector potential in the approximation (19) and (24), which we express in a coordinate system with the X axis along the polarization vector of the second exciting pulse;

$$A_\alpha(z,t)=\frac{-\pi}{8}I_\alpha^{e}\lambdabar l\left(\frac{2}{3}\hbar\lambdabar\gamma\right)^{1/2}(n_2-n_1)\sin\Omega_1T_1(1-\cos\Omega_2T_2)$$

$$\times\exp\left\{-\frac{(t-2\tau+T_1-T_2-z/c)^2}{4T_0^2}+i\left[\omega\left(\frac{z}{c}-t\right)-\Phi_1+2\Phi_2\right]\right\},$$

$$I_1^e=\cos(\psi+\varepsilon_2\tau)\cos\varepsilon_2(t-\tau-T_2-z/c)+\cos(\psi+\varepsilon_1\tau)\cos\varepsilon_1(t-\tau-T_2-z/c),$$

$$I_2^e=\cos(\psi+\varepsilon_2\tau)\sin\varepsilon_2(t-\tau-T_2-z/c)+\cos(\psi+\varepsilon_1\tau)\sin\varepsilon_1(t-\tau-T_2-z/c),$$

$$I_3^e=0,\quad\Omega_1^2=3\gamma\lambdabar a^2/2\hbar,\quad\Omega_2^2=3\gamma\lambdabar b^2/2\hbar.$$

At the instant of the occurrence of the photon echo, its polarization is parallel to the vector $I_\alpha^e(\tau)$:

$$I_1^e(\tau)=\cos(\psi+\varepsilon_2\tau)\cos\varepsilon_2\tau+\cos(\psi+\varepsilon_1\tau)\cos\varepsilon_1\tau,\qquad(26)$$

$$I_2^e(\tau)=\cos(\psi+\varepsilon_2\tau)\sin\varepsilon_2\tau+\cos(\psi+\varepsilon_1\tau)\sin\varepsilon_1\tau,\qquad(27)$$

$$I_3^e(\tau)=0.$$

An experimental determination of the direction of the vector $I_\alpha^e(\tau)$ makes it possible, with the aid of (26) and (27), to calculate the g factors of the upper and lower levels. The intensity of the photon echo is proportional to the square of the vector $I_\alpha^e(\tau)$ and depends on H in more complicated manner than in the case of the $1/2 \to 3/2$ transition.

4. PHOTON ECHO IN THE ATOMIC TRANSITION $1 \to 2$

The distinguishing feature of the atomic transition $1 \to 2$ becomes manifest already during the time of passage of the first exciting pulse. In particular, the general concept of the 90° pulse loses its meaning even when the inequality (24) is satisfied. In place of one characteristic factor $\sin\Omega_1(t - z/c)$, the amplitude of the polarization current breaks up into a sum of two terms, the first of which is proportional to $\sin\Omega_1(t - z/c)$ and the second to $\sin[2\Omega_1(t - z/c)/\sqrt{3}]$, where $\Omega_1^2 = 9\gamma\lambdabar a^2/10\hbar$.

In view of the complexity of the formulas, we shall not write out the polarization current after the passage of the first pulse. We note only that the trace of the current breaks up into a sum of three vectors rotating with angular velocities ϵ_1, ϵ_2, and $2\epsilon_1 - \epsilon_2$. These rotating vectors remain also after the passage of the second exciting pulse. In the region (20), the trace of the current is not proportional to usual factor $1 - \cos\Omega_2T_2$, which gives rise to the customary concept of the 180° pulse.

We leave out the cumbersome intermediate calculations and present the final vector potential of the photon echo at $\psi = 0$, assuming the g-factors of the upper and lower levels to be the same and equal to g:

$$A_\alpha=i\pi I_\alpha^e\frac{l}{\omega}\exp\left\{-\frac{(t-2\tau+T_1-T_2-z/c)^2}{4T_0^2}+i\left[\omega\left(\frac{z}{c}-t\right)-\Phi_1+2\Phi_2\right]\right\},\qquad(28)$$

where

$$I_1^e=u\cos\varepsilon(t-\tau-T_2-z/c)-v\sin\varepsilon(t-\tau-T_2-z/c),\qquad(29)$$

$$I_2^e=u\sin\varepsilon(t-\tau-T_2-z/c)+v\cos\varepsilon(t-\tau-T_2-z/c),\quad I_3^e=0,\qquad(30)$$

$$u=\left\{2x[1-\cos^2\varepsilon\tau\cos\Omega_2T_2-\sin^2\varepsilon\tau\cos(2\Omega_2T_2/\sqrt{3})]+y\left[3\sin^2\varepsilon\tau(1-\cos\Omega_2T_2)+(2-3\sin^2\varepsilon\tau)\left(1-\cos\frac{2\Omega_2T_2}{\sqrt{3}}\right)\right]\right\}\cos\varepsilon\tau,$$

$$v=\frac{1}{\sqrt{3}}(\cos\omega_1T_2-\cos\omega_2T_2)\left[x(1+\cos^2\varepsilon\tau)+\frac{3}{2}y(1-\cos^2\varepsilon\tau)\right]\sin\varepsilon\tau,$$

$$x=i\frac{c\hbar\Omega_1}{4a}N_0\sin\Omega_1T_1,\qquad y=i\frac{c\hbar\Omega_1}{4\sqrt{3}a}N_0\sin\frac{2\Omega_1T_1}{\sqrt{3}},$$

$$\omega_1^2=(7+4\sqrt{3})\Omega_2^2/12,\quad\omega_2^2=(7-4\sqrt{3})\Omega_2^2/12,$$

$$\Omega_2^2=9\gamma\lambdabar b^2/10\hbar,\quad N_0=n_2/3-n_1/5.$$

The remaining quantities have been defined above. If we make in (29) and (30) the formal substitutions

$$\varepsilon(t-\tau-T_2-z/c)\to 0,\quad\varepsilon\tau\to\psi,$$

then formulas (28)—(30) describe the photon echo excited at H = 0 by two light pulses polarized at an angle ψ to each other. We see therefore that in the absence of the magnetic field the polarization of the photon echo in the $1 \to 2$ atomic transition can lie either between the polarizations of the excited pulses or outside these polarizations, depending on the parameters of the experiment.

The vector potential of the photon echo in the $2 \to 1$ atomic transition is obtained from (28) by the substitutions

$$\gamma\to 5\gamma/3,\quad N_0\to n_2/5-n_1/3.$$

The authors are grateful to N. G. Basov and V. M. Galitskiĭ for useful discussions.

APPENDIX

Let us find the Faraday angle of rotation θ of the polarization over a path z in the case of the short pulse that is resonant with the atomic transition $1 \to 1$ with identical $\epsilon_1 = \epsilon_2 = \epsilon$. Since we are interested in the order of magnitude of the rotation angle, we discard the nonlinear terms in the calculation of the polarization current of the gaseous medium. The final result takes the form

$$\theta=\pi(n_2-n_1)\lambdabar^2\gamma T_0ze^{-\eta^2}\int_0^\eta e^{t^2}dt,\qquad(31)$$

where $\eta = \epsilon T_0$ and it is assumed that the collision width of the level is small compared with the Doppler width.

For the parameters of the experiment[4] and for a field intensity H of the order of 1 Oe we have $|\theta| \ll 1$. At the same time, the specific angle of rotation of the polarization of the photon echo, due to the precession of the current around H after the passage of the light pulses, has an order of magnitude larger than unity, and the direction of this rotation is opposite to a Faraday rotation (31) if $n_2 < n_1$.

The experimental[5] specific angle $2\epsilon_2\tau$ of the rotation of the polarization of the photon echo in the atomic transition $3/2 \to 1/2$ is also large compared with the Faraday rotation. However, under the experimental conditions the Faraday angle of rotation can reach a value 2π and must be taken into account.

[1] N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Lett. 13, 567 (1964).
[2] I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. 141, 391 (1966).
[3] A. Compaan, L. Q. Lambert, and I. D. Abella, Phys. Rev. Lett. 20, 1089 (1968).

[4] C. K. N. Patel and R. E. Slusher, Phys. Rev. Lett. **20**, 1087 (1968).
[5] B. Bolger and J. C. Diels, Phys. Lett. **28A**, 401 (1968).
[6] A. I. Alekseev, ZhETF Pis. Red. **9**, 472 (1969) [JETP Lett. **9**, 285 (1969)].
[7] P. S. Pershan, J. P. van der Liel, and L. D. Mainstrom, Phys. Rev. **143**, 574 (1966).
[8] A. I. Alekseev and I. V. Evseev, Zh. Eksp. Teor. Fiz. **56**, 2118 (1969) [Sov. Phys.-JETP **29**, 1139 (1969)].
[9] A. I. Alekseev and I. V. Evseev, ZhETF Pis. Red. **10**, 105 (1969) [JETP Lett. **10**, 67 (1969)].

Translated by J. G. Adashko
206