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Translated by P. Millard

Effect of depolarizing collisions on the photon echo in a magnetic field

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(Submitted 2 October 1978)

Zh. Eksp. Teor. Fiz. 76, 1212-1225 (April 1979)

A connection between the different relaxation characteristics, which describe the change of the optical-coherence matrix under the influence of elastic depolarizing collisions, is established for the first time ever. The calculation is performed for a Van der Waals interaction on a transition with level angular momenta $j_a = j_b = 1$. It is found that the relative difference between the relaxation characteristics ranges from 5 to 20% for the considered values of the interaction parameters. The possibility is demonstrated of experimentally measuring the characteristics that describe the relaxation of the optical-coherence matrix on the transitions $j \rightarrow j$ ($j > 1$) and $j \rightarrow j + 1$ ($j \geq 1/2$) by the photon-echo method in a gas situated in a longitudinal magnetic field. It is found for the $1 \rightarrow 1$ and $1/2 \rightarrow 3/2$ transitions that the magnetic field intensity can be chosen such that the polarization echo vector component perpendicular to the polarization plane of the exciting pulses is due entirely only to depolarizing collisions.

PACS numbers: 42.65.Gb, 42.50.+q

Recent years have seen progress in nonlinear laser spectroscopy, which permits the study of quantum-transition structures obscured by the Doppler broadening of spectral lines. An extensive bibliography on this topic is contained in the monograph of Letokhov and Chebotayev.¹ In a gas medium, the resonance levels of moving atoms (molecules) are usually degenerate. A consistent calculation of the elastic collisions shows therefore that the relaxation of that density-matrix component which describes optical coherence (transitions between the considered levels) is determined not by a single quantity but by an aggregate $\mathcal{F}^{(\kappa)}$, where $|j_a - j_b| \leq \kappa \leq j_a + j_b$, and j_a and j_b are the angular momenta of the levels of the transition. The aggregate of the quantities $\mathcal{F}^{(\kappa)}$ determines in essence the properties of the resonant electromagnetic radiation that passes through a gas medium. In particular, the gain of a weak probing wave on a transition with level angular momenta $j_a = 1$ and $j_b = 2$, in a medium saturated by a strong field, depends essentially on the ratio of the relaxation characteristics of the dipole polarization $\mathcal{F}^{(1)}$ and of the quadrupole polarization $\mathcal{F}^{(2)}$ of the medium. So far, however, there are no concrete theoretical or experimental results concerning the ratios of the different $\mathcal{F}^{(\kappa)}$.

In the first part of this paper we calculate all the relaxation characteristics $\mathcal{F}^{(\kappa)}$ for the transition $j_a = j_b = 1$ in the case of a Van der Waals interaction of the

colliding atoms. The connection between the relaxation characteristics having different values of κ are established here for the first time. The results allow us to check the correctness the assumption, used in many papers, that all the $\mathcal{F}^{(\kappa)}$ are approximately equal.

In the second part of the paper it is shown on the basis of a theoretical calculation that the differences $\mathcal{F}^{(\kappa)} - \mathcal{F}^{(1)}$ ($\kappa \neq 1$) can be determined by direct experiment using photon echo in a gas medium placed in a longitudinal magnetic field.

The photon echo method has been coming into ever increasing use for the study of gas media.²⁻¹⁶ It is employed to determine successfully the relaxation characteristics of resonant transitions, as well as to identify atomic and molecular transitions. In particular, Wang⁶ has obtained theoretically the influence of the difference $\mathcal{F}^{(\kappa)} - \mathcal{F}^{(1)}$ ($\kappa \neq 1$) on the polarization of photon echo on the transitions $j \rightarrow j$ ($j > 1$) and $j \rightarrow j + 1$ ($j \geq 1$). On the remaining transitions, the echo amplitude depended only on the quantity $\mathcal{F}^{(1)}$, and its polarization was not affected by the depolarizing collisions.

Application of a longitudinal magnetic field on a gas medium in which a photon echo is produced extends substantially the capabilities of the photon echo. Thus, for example, a specific rotation of the photon-echo polarization vector is observed, different from the Faraday rotation. This effect was predicted by Aleks-

sev,⁴ was investigated in detail in Ref. 5, and has by now been experimentally verified.¹⁵

The dipole polarization of a gas medium placed in a magnetic field, for the transitions $j - j (j \geq 1)$ and $j \neq j + 1 (j \geq \frac{1}{2})$, is connected with the other multipole polarization moments that are induced in the medium together with the dipole polarization by the exciting light pulses. Therefore, in contrast to Ref. 6, we have observed that the photon echo polarization is altered by the depolarizing collisions both on the $1 \rightarrow 1$ and on the $\frac{1}{2} \rightarrow \frac{3}{2}$ transitions. In addition, the dependence of the angle of the specific rotation of the polarization-echo angle on the magnetic field H makes it possible to choose H such that the entire difference between the echo polarization and the polarizations of the exciting light pulses is determined by the depolarizing collisions. The separation of the echo electric-field intensity component perpendicular to the polarization plane of the exciting pulses makes it possible then to obtain directly the experimental information on the differences $\mathcal{F}^{(\kappa)} - \mathcal{F}^{(\lambda)} (\kappa \neq \lambda)$.

1. CALCULATION OF THE RELAXATION CHARACTERISTICS $\mathcal{F}^{(\kappa)}$

We consider a transition between excited levels a and b of an atom (molecule) with respective angular momenta j_a and j_b . Assuming the concentration of the excited atoms to be small enough, we take into account the elastic collisions of these atoms with the unexcited ones, the ground state of which is characterized by an angular momentum $j=0$. In the approximation of the depolarizing atomic collisions, the density-matrix component $\rho_{\mu, m}^{(b, a)}$ describing the transitions between the considered levels satisfies the equation

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \nabla + \frac{\gamma_a^{(0)} + \gamma_b^{(0)}}{2} - i\omega_0 \right) \rho_{\mu m}^{(b, a)} = - \sum_{\mu', m'} \mathcal{F}^{(\nu)} \rho_{\mu' m'}^{(b, a)}, \quad (1)$$

where ν is the velocity of the excited atom; the indices μ and m number respectively the projections of the angular momenta j_b and j_a ; $1/\gamma_a^{(0)}$ and $1/\gamma_b^{(0)}$ are the times of relaxation of the excited states by gaskinetic inelastic collisions and radiative decay; ω_0 is the frequency of the transition between levels a and b .

The matrix $\mathcal{F}^{(\nu)}$, which describes the relaxation of $\rho_{\mu, m}^{(b, a)}$ under the influence of the elastic depolarizing collisions, is given by

$$\mathcal{F}^{(\nu)} \rho_{\mu m}^{(b, a)} = n_0 \int (\delta_{\mu\mu'} \delta_{mm'} - S_{\mu\mu'}^{(b)} S_{mm'}^{(a)}) |\mathbf{v} - \mathbf{v}_0| f(\nu_0) d\nu_0 d\rho. \quad (2)$$

Here n_0 is the density of the unexcited atoms, $f(\nu_0)$ is the Maxwellian distribution in the velocities \mathbf{v}_0 of the unexcited atoms; $d\rho$ stands for integration with respect to the impact parameter ρ in a plane perpendicular to the relative velocity $\mathbf{v} - \mathbf{v}_0$; $S_{\mu\mu'}^{(b)}$ and $S_{mm'}^{(a)}$ are the matrix elements of the S -matrix of the elastic scattering of the excited atom in the states b and a by the unexcited one. We note that in the approximation of the depolarizing collisions $S_{\mu\mu}^{(b)}$ satisfies the equation¹⁷

$$i\hbar(\mathbf{v} - \mathbf{v}_0) \nabla S_{\mu\mu}^{(b)} = \sum_{\mu''} V_{\mu\mu''}(\mathbf{R}) S_{\mu''\mu}^{(b)}, \quad (3)$$

with the additional condition $S_{\mu\mu_0}^{(b)}(-\infty) = \delta_{\mu\mu_0}$, where the index μ_0 characterizes the state of the atom prior to the collision. $V_{\mu\mu'}(\mathbf{R})$ is the matrix element of the operator

of the interaction of the excited and unexcited atoms, and \mathbf{R} is the vector that joins them. A similar equation holds also for $S_{mm'}^{(a)}$.

It is customary to neglect the dependence of $\mathcal{F}^{(\nu)} \rho_{\mu' m'}$ on the velocity \mathbf{v} of the excited atom in investigations of the influence of depolarizing collisions on the interaction between electromagnetic radiation and a gas; this is equivalent to averaging $\mathcal{F}^{(\nu)}$ with over \mathbf{v} with a Maxwellian distribution. In the present paper this averaging is carried out in two steps. In the first, the averaging is over the direction of \mathbf{v} , and the dependence on the modulus of \mathbf{v} is retained in $\mathcal{F}^{(\nu)}$.

It is convenient to continue the investigation by expanding the matrices $\rho_{\mu m}^{(b, a)}$ and $\mathcal{F}^{(\nu)} \rho_{\mu' m'}$ in irreducible tensor operators,¹⁸ e.g.,

$$\rho_{\mu m}^{(b, a)} = (-1)^{j_b - \mu} (2j_b + 1)^{-1/2} \sum_{\kappa, q} (2\kappa + 1) \begin{pmatrix} j_b & j_a & \kappa \\ \mu & -m & q \end{pmatrix} \Psi_q^{(\kappa)}. \quad (4)$$

We then obtain from (1) the following equations for the components $\Psi_q^{(\kappa)}$:

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \nabla + \frac{\gamma_a^{(0)} + \gamma_b^{(0)}}{2} - i\omega_0 \right) \Psi_q^{(\kappa)} = -\mathcal{F}^{(\nu)} \Psi_q^{(\kappa)}. \quad (5)$$

Here $\mathcal{F}^{(\nu)} \Psi_q^{(\kappa)}$ are complex quantities connected with $\mathcal{F}^{(\nu)} \rho_{\mu' m'}$ by a relation analogous to (4). We note that independent relaxation of each of the components $\Psi_q^{(\kappa)}$ is the consequence of the averaging over the direction of the velocity of the excited atom. The quantities $\mathcal{F}^{(\nu)} \Psi_q^{(\kappa)}$ are best determined in a coordinate frame with a Z axis directed along the relative velocity $\mathbf{v} - \mathbf{v}_0$, and with an X axis chosen along the vector ρ . In this system, the $\mathcal{F}^{(\nu)} \Psi_q^{(\kappa)}$ are connected with the scattering matrices $\bar{S}^{(b)}$ and $\bar{S}^{(a)}$ by the relation

$$\mathcal{F}^{(\nu)} \Psi_q^{(\kappa)} = n_0 \sum_{m, m', \mu, \mu', l} (-1)^{2j_b + \mu + \mu'} \begin{pmatrix} j_b & j_a & \kappa \\ \mu & -m & l \end{pmatrix} \begin{pmatrix} j_b & j_a & \kappa \\ \mu' & -m' & l \end{pmatrix} \times \int (\delta_{\mu\mu'} \delta_{mm'} - \bar{S}_{\mu\mu'}^{(b)} \bar{S}_{mm'}^{(a)}) |\mathbf{v} - \mathbf{v}_0| f(\nu_0) d\nu_0 d\rho, \quad (6)$$

where $\bar{S}_{\mu\mu'}^{(b)}$ and $\bar{S}_{mm'}^{(a)}$ are the scattering matrices of the atom in the states b and a , calculated in the indicated coordinate frame.

For the subsequent calculation we must specify the form of the interaction potential and the values of the angular momenta of the levels. All the calculations were performed for the case $j_a = j_b = 1$ and for a Van der Waals potential. As shown in Ref. 19, a Van der Waals interaction potential is characterized by two independent constants, we shall designate by $c_0^{(a)}$ and $c_1^{(a)}$ for the atom in state a and by $c_0^{(b)}$ and $c_1^{(b)}$ for the atom in state b .

We introduce $\Gamma^{(\kappa)}(\nu)$ and $\Delta^{(\kappa)}(\nu)$ defined by

$$\mathcal{F}^{(\nu)} \Psi_q^{(\kappa)} = \Gamma^{(\kappa)} \Psi_q^{(\kappa)} - i\Delta^{(\kappa)} \Psi_q^{(\kappa)}.$$

After cumbersome but straightforward calculations, we obtain the following expressions for $\Gamma^{(\kappa)}$ and $\Delta^{(\kappa)}$:

$$\Gamma^{(\kappa)} \Psi_q^{(\kappa)} - i\epsilon (c_1^{(b)} - c_1^{(a)}) \Delta^{(\kappa)} \Psi_q^{(\kappa)} = \gamma(\nu) F^{(\kappa)}(s, t) = \gamma(\nu) [F_1^{(\kappa)}(s, t) + iF_2^{(\kappa)}(s, t)], \quad (7)$$

where

$$\gamma(\nu) = \pi n_0 |c_1^{(b)} - c_1^{(a)}|^{3/2} \left(\frac{2T}{M_1} \right)^{3/2} \alpha^{-1/2} e^{-\alpha^2 s^2} \Phi \left(\frac{9}{5}, \frac{3}{2}, \alpha^2 x^2 \right), \quad (8)$$

$$s = (c_1^{(b)} + c_0^{(b)} - c_1^{(a)} - c_0^{(a)}) / (c_1^{(b)} - c_0^{(b)}), \quad t = (c_1^{(a)} - c_0^{(a)}) / (c_1^{(b)} - c_0^{(b)}),$$

$$\alpha = \sqrt{M_2/M_1}, \quad x = v\sqrt{M_1/2T}, \quad \varepsilon(x) = \begin{cases} 1, & x > 0 \\ -1, & x < 0 \end{cases} \quad (9)$$

Here M_1 is the mass of the excited atom and M_2 is the mass of the unexcited impurity (or host) gas: $\Phi(\alpha, \beta, z)$ is a confluent hypergeometric function. All the dimensionless real functions $F_i^{(\kappa)}(s, t)$ ($i=1, 2$; $\kappa=0, 1, 2$) are expressed in terms of single integrals with respect to the quantity u connected with the impact parameter by the relation

$$u = 2\rho^2 |v - v_0| / |c_1^{(b)} - c_0^{(b)}|.$$

To shorten the notation we present expressions for the complex functions $F^{(\kappa)}(s, t)$:

$$F^{(0)}(s, t) = B \int_0^{\infty} \frac{du}{u^{3/2}} \left\{ 3 - \left[2 \operatorname{Re} w(u) w^* \left(\frac{u}{t} \right) + 2v(u) v^* \left(\frac{u}{t} \right) + \exp \left(-\frac{3\pi i}{8u} (1-t) \right) \right] \exp \left(-\frac{3\pi i}{8u} s \right) \right\}, \quad (10)$$

$$F^{(1)}(s, t) = B \int_0^{\infty} \frac{du}{u^{3/2}} \left\{ 3 - \left[\operatorname{Re} w(u) w \left(\frac{u}{t} \right) + v(u) v^* \left(\frac{u}{t} \right) + \exp \left(\frac{3\pi i}{8u} t \right) \operatorname{Re} w(u) + \exp \left(-\frac{3\pi i}{8u} \right) \operatorname{Re} w \left(\frac{u}{t} \right) \right] \exp \left(-\frac{3\pi i}{8u} s \right) \right\}, \quad (11)$$

$$F^{(2)}(s, t) = \frac{2}{5} F^{(0)}(s, t) + \frac{3}{5} F^{(1)}(s, t) + \frac{12}{5} B \int_0^{\infty} \frac{du}{u^{3/2}} v(u) v^* \left(\frac{u}{t} \right) \exp \left(-\frac{3\pi i}{8u} s \right), \quad (12)$$

where $B = 2^{9/5} \Gamma(\frac{5}{2}) / 15\sqrt{\pi}$ and $\Gamma(x)$ is the gamma function. The complex functions $v(u)$ and $w(u)$ satisfy the system of equations

$$iu \frac{dw}{d\xi} = -(1-\xi^2)^{3/2} [(1-2\xi^2)w + 2\xi(1-\xi^2)^{3/2}v],$$

$$iu \frac{dv}{d\xi} = -(1-\xi^2)^{3/2} [2\xi(1-\xi^2)^{3/2}w - (1-2\xi^2)v] \quad (13)$$

with initial conditions

$$w(\xi=-1)=1, \quad v(\xi=-1)=0.$$

The system (13) is a consequence of Eq. (3) and of the analogous equation for $S_{\mu\mu'}^{(b)}$. The values of w and v in (10)–(12) are the solutions of the system (13) and are taken at the point $\xi=1$.

The calculation of the integrals (10)–(12) and the solution of the system (13) were carried out numerically with a computer, with the exception of the values $u \ll 1$ and $u \gg 1$. The contribution of these regions to the integrals were estimated from an approximate analytic solution of the system (13).

It can be shown that the following relation holds:

$$F^{(\kappa)}(-s/t, 1/t) = t^{-\kappa} F^{(\kappa)}(s, t), \quad t > 0,$$

the calculation was therefore performed for two values of the parameter t , 0.1 and 1. The parameter s took on in this case the values 0, ± 0.2 , ± 0.4 , ± 0.6 , ± 1 , ± 2 , ..., ± 10 . As a check on the accuracy of the calculations, we used the results at $t=1$ and $s=0$. These values of the

parameters correspond to the case when the interaction potentials of the two excited levels a and b are equal, and the problem reduces to the calculation of the polarization characteristics of the level with angular momentum $j=1$.¹⁹

Figure 1 shows, for $t=1$, the dependence of the dimensionless widths

$$F_i^{(\kappa)} = \Gamma^{(\kappa)}(v) / \gamma(v)$$

and of the dimensionless shifts

$$F_2^{(\kappa)} = -\varepsilon(c_0^{(1)} - c_0^{(0)}) \Delta^{(\kappa)}(v) / \gamma(v), \quad \kappa=0, 1,$$

on the parameter s (curves 1–4). We note that the following relations hold for the given value $t=1$:

$$F_1^{(\kappa)}(s) = F_1^{(\kappa)}(-s), \quad F_2^{(\kappa)}(s) = -F_2^{(\kappa)}(-s).$$

It follows from (10)–(12) that there is no such symmetry for $t \neq 1$. The difference between $F_1^{(0)}$ and $F_1^{(1)}$ is significant only at $|s| \lesssim 1.5$. At $|s| \gtrsim 1.5$ the difference between these quantities does not exceed the calculation error, and the two curves therefore coalesce in this region. A similar remark holds also for the behavior of the shifts $F_2^{(0)}$ and $F_2^{(1)}$, where the coalescence regions begin with $|s| \gtrsim 3$. Since the important role in what follows is played by the behavior of the differences $F_1^{(2)} - F_1^{(1)}$ and $F_2^{(2)} - F_2^{(1)}$, these quantities were calculated separately with increased accuracy. The results of these calculations are represented by curves 5 and 6.

Figure 2 shows the quantities $F_1^{(\kappa)}$, $F_2^{(\kappa)}$ ($\kappa=0, 1, 2$) and the difference $F_1^{(2)} - F_1^{(1)}$ at $t=0.1$ as functions of the parameter s . Within the limits of the calculation accuracy, the quantities $F_1^{(0)}$, $F_1^{(1)}$, and $F_1^{(2)}$ are equal. This statement pertains also to $F_2^{(0)}$, $F_2^{(1)}$, and $F_2^{(2)}$. A comparison of the results at $t=0.1$ and $t=1$ shows that in the region $|s| \gtrsim 1$ all the quantities depend weakly on t , whereas at $|s| \lesssim 1$ their dependence is strong. Just as at $t=1$, the calculation of the differences $F_1^{(2)} - F_1^{(1)}$ and $F_2^{(2)} - F_2^{(1)}$ was carried out with increased accuracy. The results of the calculation of $F_1^{(2)} - F_1^{(1)}$ are shown by curve 3. The quantity $F_2^{(2)} - F_2^{(1)}$ is not shown, since the absolute

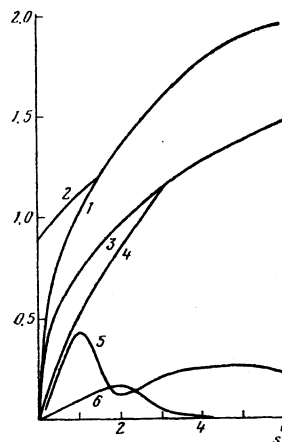


FIG. 1. Plots of $F_1^{(0)}$, $F_1^{(1)}$, $F_2^{(0)}$, $F_2^{(1)}$, $2(F_1^{(1)} - F_1^{(2)})$ and $2(F_2^{(2)} - F_2^{(1)})$ against s at $t=1$ —curves 1, 2, 3, 4, 5, and 6, respectively.

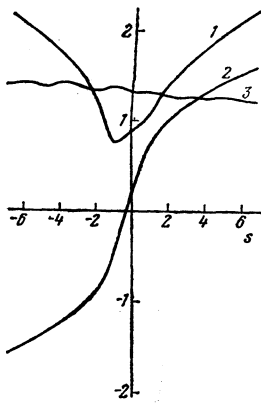


FIG. 2. Plots of $F_1^{(0)}$, $F_2^{(0)}$, and $10(F_1^{(1)} - F_1^{(2)})$ against s at $t = 0.01$ —curves 1, 2, and 3, respectively.

value of this difference does not exceed 0.05. We note that for most calculated variants we have

$$|F_1^{(2)} - F_1^{(1)}| \gg |F_2^{(2)} - F_2^{(1)}|.$$

We can thus conclude that the relative difference between the quantities $\Gamma^{(0)}(v)$, $\Gamma^{(1)}(v)$, and $\Gamma^{(2)}(v)$ ranges from 5 to 20%. In the next part of the paper we predict an effect that permits an experimental determination of the difference between these quantities.

2. CALCULATION OF THE PHOTON ECHO ELECTRIC FIELD INTENSITY

We consider the formation of a photon echo in a gas medium situated in a homogeneous magnetic field H , when the time interval τ_s between the two exciting light pulses can be comparable with the irreversible relaxation times. In a magnetic field the upper degenerate level a and the lower one b , with energies E_a and E_b and with total angular momenta j_a and j_b , at resonance with the frequency ω of the exciting light pulses, are converted into two groups of Zeeman sublevels with energies $E_{a\mu}$ and $E_{b\mu}$. In the approximations linear in H , the quantities $E_{a\mu}$ and $E_{b\mu}$ are given by

$$E_{a\mu} = E_a + \mu_0 g_a H m, \quad E_{b\mu} = E_b + \mu_0 g_b H \mu,$$

where μ_0 is the Bohr magneton, and the subscripts m and μ characterize the projections of the total angular momenta of the upper and lower resonance levels with respective g -factors g_a and g_b . We assume for simplicity that the Zeeman splitting is small compared with $\hbar\omega$.

To determine the electric field intensity of the photon echo, we use the d'Alembert equation

$$\square E = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \int P dv \quad (14)$$

and the quantum-mechanical equation for the density matrix, with account taken of the interaction of the atoms (molecules) of the gas with the electromagnetic field, with the external constant field H , the irreversible relaxation, as well as the radiative transition to the lower resonant level on account of spontaneous emission on the upper one. The medium polarization vector P in (14) pertains to a group of atoms moving with ve-

locity v , and is connected with the density-matrix component $\rho_{\mu\mu'}^{(b,a)}$ by the relation

$$P = \sum_{\mu, \mu'} \rho_{\mu\mu'}^{(b,a)} d_{\mu\mu'} + \text{c.c.},$$

where $d_{\mu\mu'}$ are the matrix elements of the dipole moment of the atom and correspond to the transition between states a and b .

It is convenient to solve the system of equations for the components of the density matrix by expanding the matrix in renormalized tensor operators. In this case $\rho_{\mu\mu'}^{(b,a)}$ is connected with $\Psi_q^{(x)}$ by relation (4), and the connection of $\rho_{\mu\mu'}^{(a,a)}$ with $f_q^{(x)}$ and of $\rho_{\mu\mu'}^{(b,b)}$ with $\varphi_q^{(x)}$ takes the form

$$\rho_{mm'}^{(a,a)} = (-1)^{j_a - m} (2j_a + 1)^{-1/2} \sum_{x,q} (2x+1) \begin{pmatrix} j_a & j_a & x \\ m & -m' & q \end{pmatrix} f_q^{(x)},$$

$$\rho_{\mu\mu'}^{(b,b)} = (-1)^{j_b - \mu} (2j_b + 1)^{-1/2} \sum_{x,q} (2x+1) \begin{pmatrix} j_b & j_b & x \\ \mu & -\mu' & q \end{pmatrix} \varphi_q^{(x)}.$$

Let the exciting light pulses propagate along the Z axis and let them be linearly polarized along the X axis. Then the electric field intensity of the exciting pulses are given by

$$E_1 = I_x e^{(1)} \exp[i(\omega t - kz + \Phi_1)] + \text{c.c.}, \quad 0 \leq t - z/c \leq T_1, \quad (15)$$

$$E_2 = I_x e^{(2)} \exp[i(\omega t - kz + \Phi_2)] + \text{c.c.}, \quad \tau_s + T_1 \leq t - z/c \leq \tau_s + T_1 + T_2, \quad (16)$$

where T_1 and T_2 are the pulse durations; the amplitudes $e^{(1)}$ and $e^{(2)}$, as well as the phase shifts Φ_1 and Φ_2 , are constant; I_x is the unit vector of the Cartesian axis X .

We separate in the sought functions the rapidly oscillating particles

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

$$\Psi_q^{(x)} = \psi_q^{(x)} \exp[i(\omega t - kz + \Phi)],$$

where e and $\psi_q^{(x)}$ are slowly varying amplitudes, and Φ is the constant phase shift. In the resonant approximation we get from (14) and from the equations for $\Psi_q^{(x)} f_q^{(x)}$ and $\varphi_q^{(x)}$ the following equations for the slow functions:

$$\left(\frac{\partial}{\partial t} + c \frac{\partial}{\partial z} \right) e_q = -2i\pi\omega \int p_q dv, \quad (17)$$

$$\begin{aligned} \partial \Psi_q^{(x)} + i \frac{E_b - E_a}{2} \sum_{x'} Q_q^{xx'} \Psi_q^{(x')} \\ = \frac{i}{\hbar} (2j_a + 1)^{-1/2} d' \sum_{x', q', q_1} e_{-q_1} [S_{qq', q_1}^{xx'} f_{q'}^{(x')} + (-1)^{x+x'} R_{qq', q_1}^{xx'} \varphi_{q'}^{(x')}], \end{aligned} \quad (18)$$

$$\partial_a f_q^{(x)} = \frac{i}{\hbar} (2j_b + 1)^{-1/2} \sum_{x', q', q_1} C_{qq', q_1}^{xx'} T_{q'}^{xx'}, \quad (19)$$

$$\partial_b \varphi_q^{(x)} = \frac{i}{\hbar} (2j_b + 1)^{-1/2} \sum_{x', q', q_1} (-1)^{x+x'} B_{qq', q_1}^{xx'} T_{q'}^{xx'} + \Gamma_q f_q^{(x)}, \quad (20)$$

where we have introduced the abbreviated symbols for the operators:

$$\partial = \frac{\partial}{\partial t} + \gamma^{(x)} + i(\Delta\omega - kv_z - \Delta^{(x)} - qe_b),$$

$$\partial_{a,b} = \frac{\partial}{\partial t} + \gamma_{a,b}^{(x)} + \Gamma_{a,b}^{(x)} - iqe_{a,b}.$$

The detuning of the field frequency ω from the central

frequency ω_0 is designated $\Delta\omega = \omega - \omega_0$, and the frequencies of the transitions between the neighboring Zeeman sublevels of the considered levels take the form

$$\varepsilon_{a,b} = \mu_0 g_{a,b} H / \hbar.$$

The matrix operator

$$Q_i^{m'} = (-1)^{j_a + j_b + \sigma} [2j_a(2j_a + 1)(2j_a + 2)]^{1/2} (2\kappa' + 1) \begin{pmatrix} 1 & \kappa & \kappa' \\ 0 & -q & q \end{pmatrix} \begin{Bmatrix} 1 & \kappa & \kappa' \\ j_b & j_a & j_a \end{Bmatrix}$$

in Eq. (18) determines the degree of connection between the different components of the optical-coherence matrix due to the inequality of the g -factors of the levels. In (19) and (20), the quantity

$$T_{q'q_i}^{m'} = d\psi_{q'}^{(m')} e_{-q_i}^* + (-1)^{\sigma' + \sigma + \sigma'} e_{-q_i} d' \psi_{-q'}^{(m')*}$$

describes the change of the components of the density matrix of the level because of the interaction with the electromagnetic field. The term $\Gamma_{\kappa} f_q^{(x)}$ in (20) takes into account the arrival at the lower level because of spontaneous emission from the upper. The quantity S in (18) is obtained from

$$C_{\kappa'q_i}^{m'} = (-1)^{j_b + \sigma'} (2j_a + 1)^{1/2} (2\kappa' + 1) \begin{pmatrix} \kappa' & 1 & \kappa \\ -q' & q_i & q \end{pmatrix} \begin{Bmatrix} \kappa' & 1 & \kappa \\ j_a & j_a & j_b \end{Bmatrix}$$

by the substitution $j_a \rightleftharpoons j_b$ everywhere, except in the second column of the $6j$ -number. In turn, the quantities B and R are obtained respectively from C and S by the transformation $j_a \rightleftharpoons j_b$. The circular components of the vector p are connected with $\psi_q^{(1)}$ by the relation

$$p_q = (-1)^{j_a - j_b + \sigma} (2j_b + 1)^{-1/2} d\psi_q^{(1)};$$

e_q is the circular component of the vector e ; d is the reduced matrix element of the dipole-moment operator of the transition $j_a \rightarrow j_b$, and the quantities $\gamma^{(x)}$ are given by

$$\gamma^{(x)} = (\gamma_a^{(x)} + \gamma_b^{(x)}) / 2 + \Gamma^{(x)}.$$

Next, $\Gamma_a^{(x)}$ and $\Gamma_b^{(x)}$ describe the collision relaxations of the upper and lower levels, due to the transitions between the Zeeman sublevels of the excited atom. They are connected with the S matrix by relations such as (6), and since we are dealing with elastic collisions that do not alter the total level population of the levels, it follows that $\Gamma_a^{(0)} = \Gamma_b^{(0)} = 0$. Finally, $\Gamma^{(x)}$ and $\Delta^{(x)}$ are obtained from (6) by averaging over the modulus of the velocity

$$\Gamma^{(x)} - i\Delta^{(x)} = \int v^2 dv f(v) \mathcal{F}^{(x)}(v),$$

where $f(v)$ describes the Maxwellian distribution of the excited atoms in velocity.

At the initial instant of time $t - z/c = 0$ the quantities

$$\psi_q^{(m)} = \psi_q^{(m)}(v, t - z/c), \quad f_q^{(m)} = f_q^{(m)}(v, t - z/c), \quad \varphi_q^{(m)} = \varphi_q^{(m)}(v, t - z/c)$$

take the form

$$\begin{aligned} \psi_q^{(m)}(v, 0) &= 0, \quad f_q^{(m)}(v, 0) = (2j_a + 1) n_a f(v) \delta_{m,0} \delta_{q,0}, \\ \varphi_q^{(m)}(v, 0) &= (2j_b + 1) n_b f(v) \delta_{m,0} \delta_{q,0}, \end{aligned} \quad (21)$$

where $(2j_a + 1)n_a$ and $(2j_b + 1)n_b$ are the densities of the atoms on the upper and lower levels at $t - z/c = 0$. The

Boltzmann distribution of the atoms over the Zeeman sublevels at the initial instant $t - z/c = 0$ are neglected under the assumption of equal probability of the distribution over the sublevels in (21). In the calculations that follow we neglect the effect of the magnetic field during the time that the exciting light pulses pass through the gas medium, assuming them to be short enough:

$$\varepsilon_a T_i \ll 1, \quad \varepsilon_b T_i \ll 1, \quad i = 1, 2.$$

In addition, we assume that the durations T_1 and T_2 of the exciting pulses are small compared with the intermediate time τ_s between them and with the times of the irreversible relaxations. This assumption, which is usually well satisfied in experiment, allows us to disregard the irreversible relaxation during the time of passage of the exciting pulses.

When solving Eqs. (17)–(20) we neglect the reaction of the medium on the exciting light pulses. These equations then become linear and make it possible to determine the intensity of the electric field of the photon echo in the given-field approximation (15) and (16). To linearize Eqs. (17)–(20) the following condition must be satisfied

$$\omega |d|^2 L |N_0| T / (2j_a + 1) \hbar c \ll 1,$$

where T is the smallest of the times T_1 , T_2 , and T_0 ; $N_0 = n_a - n_b$ is the density of the excess population of the Zeeman sublevels, L is the dimension of the gas medium, and the time T_0 of the irreversible Doppler relaxation is expressed in terms of the average thermal velocity u of the gas atoms: $T_0 = 1/ku$.

We consider in this section the formation of photon echo on the $j_a = j_b = 1$ transition by exciting pulses that are linearly polarized in one plane. Solving Eqs. (17)–(20) with account taken of the initial conditions (21), we obtain $\Psi_q^{(x)}(t - z/c)$ at the instant of time $t = T_1 + z/c$, when the first exciting pulse leaves the point z of the gas medium, in the form

$$\Psi_q^{(x)}(T_1) = -\frac{\sqrt{3} p^{(1)}}{d} \delta_{m,1} (\delta_{q,1} - \delta_{q,-1}) \exp[i(\omega T_1 + \Phi_1)], \quad (22)$$

where

$$\begin{aligned} p^{(1)} &= -i \frac{|d|^2}{3\sqrt{2}\hbar\Omega_1} e^{(1)} N_0 f(v) \left[\sin \Omega_1 T_1 + i \frac{kv_z - \Delta\omega}{\Omega_1} (1 - \cos \Omega_1 T_1) \right], \\ \Omega_1^2 &= (kv_z - \Delta\omega)^2 + 2|d|^2 e^{(1)2} / 3\hbar^2. \end{aligned}$$

It follows from (22) that the polarization vector P of a group of atoms moving at a velocity v is directed at the instant of time $t = T_1 + z/c$ along the polarization vector of the exciting pulse, this being a consequence of the given-field approximation.

In the region $T_1 \leq t - z/c \leq \tau_s + T_1$, after the passage of the first exciting pulse, we obtain from (18) with allowance for (22)

$$\begin{aligned} \Psi_q^{(x)} \left(t - T_1 - \frac{z}{c} \right) &= -\frac{\sqrt{3} p^{(1)}}{d} L^{(x)} \left(t - T_1 - \frac{z}{c} \right) (\delta_{q,1} - \delta_{q,-1}) \\ &\times \exp \left[i(\omega_0 + kv_z) \left(t - T_1 - \frac{z}{c} \right) + i(\omega T_1 + \Phi_1) \right] \\ &\times \exp \left[\frac{i}{2} q (\varepsilon_a + \varepsilon_b) \left(t - T_1 - \frac{z}{c} \right) \right], \end{aligned} \quad (23)$$

where

$$L^{(1)}(t) = \left\{ \text{ch } \tau t - \frac{1}{2r} [\gamma^{(1)} - \gamma^{(2)} - i(\Delta^{(1)} - \Delta^{(2)})] \text{sh } \tau t \right\} R(t),$$

$$L^{(2)}(t) = i \sqrt{\frac{3}{5}} \frac{\varepsilon_a - \varepsilon_b}{2r} \text{sh } \tau t R(t),$$

$$R(t) = \exp \left\{ -\frac{1}{2} [\gamma^{(1)} + \gamma^{(2)} - i(\Delta^{(1)} + \Delta^{(2)})] t \right\},$$

$$r^2 = \frac{1}{4} \{ [\gamma^{(1)} - \gamma^{(2)} - i(\Delta^{(1)} - \Delta^{(2)})]^2 - (\varepsilon_a - \varepsilon_b)^2 \}.$$

Analyzing (23) we can verify that the vector \mathbf{P} rotates around the direction of the magnetic field. By the instant of passage of the second exciting pulse, it is rotated through a certain angle whose value depends on H , on the g -factors of the levels, and on τ_s . The damping of the vector \mathbf{P} is greatly influenced by the depolarizing collisions.

During the time $\tau_s + T_1 \leq t - z/c \leq \tau_s + T_1 + T_2$ a second exciting pulse (16) passes through the point z of the medium. The linearized equations (17)–(20) are solved in the same manner as in the preceding case of the passage of the first pulse. The initial conditions are chosen to be the solutions of the system (17)–(20) taken at the instant of time $t = \tau_s + T_1 + z/c$. One of the initial conditions is obtained from (23) at $t = \tau_s + T_1 + z/c$. We note that it is precisely this initial condition alone which contributes to the electric field intensity of the photon echo. At the instant $t = \tau_s + T_1 + T_2 + z/c$, when the second pulse (16) leaves the point z of the gas medium, the part of $\Psi_q^{(2)}$ that contributes to the echo is of the form

$$\Psi_q^{(2)}(T_2) = -\frac{\sqrt{3} p^{(1)} p^{(2)}}{2d} M_q^{(2)} \exp \{ i[\omega(\tau_s + T_1 + T_2) + (\Delta\omega - kv_z)\tau_s + 2\Phi_2 - \Phi_1] \},$$

$$M_q^{(1)} = (\delta_{q,1} - \delta_{q,-1}) L^{(1)*}(\tau_s) \cos[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s],$$

$$M_q^{(2)} = (\delta_{q,1} + \delta_{q,-1}) L^{(2)*}(\tau_s) \sin[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s],$$

$$p^{(2)} = 2|d|^2 e^{(2)2} (1 - \cos \Omega_2 T_2) / 3\hbar^2 \Omega_2^2,$$

$$\Omega_2^2 = (kv_z - \Delta\omega)^2 + 2|d|^2 e^{(2)2} / 3\hbar^2.$$

Expression (24) is the initial condition for the solution of Eqs. (17) and (18) in the region $\tau_s + T_1 + T_2 \leq t - z/c$ after the passage of the second exciting pulse. Leaving out the intermediate calculations, we write down the final expression for the electric field intensity of the photon echo:

$$E^e = e^e i \frac{\pi\omega L}{\sqrt{2}c} I \exp \{ i[\omega_0 t - kz + 2\Phi_2 - \Phi_1 + \Delta\omega(2\tau_s + T_1 + T_2)] \} + \text{c.c.},$$

$$e_x^e = 2 \cos[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s] \cos[\frac{1}{2}(\varepsilon_a + \varepsilon_b)t'] L^{(1)*}(\tau_s) L^{(1)}(t') + i^{10} \sin[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s] \sin[\frac{1}{2}(\varepsilon_a + \varepsilon_b)t'] L^{(2)*}(\tau_s) L^{(2)}(t'),$$

$$e_y^e = 2 \cos[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s] \sin[\frac{1}{2}(\varepsilon_a + \varepsilon_b)t'] L^{(1)*}(\tau_s) L^{(1)}(t') - i^{10} \sin[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s] \cos[\frac{1}{2}(\varepsilon_a + \varepsilon_b)t'] L^{(2)*}(\tau_s) L^{(2)}(t'),$$

$$I = \int d\nu p^{(1)*} p^{(2)} \exp[ik\nu_z(t' - \tau_s)],$$

where $t' = t - \tau_s - T_1 - T_2 - z/c$.

As follows from (25)–(28), the photon echo propagates with a carrier frequency ω_0 and is linearly polarized. Owing to the presence in the magnetic field of a specific photon-echo polarization rotation due to the precession of the polarization vector \mathbf{P} around the direction of H ,

the echo polarization plane does not coincide in the general case with the polarization plane of the exciting light pulses.

Investigating (28), we can obtain for each point z of the gas medium the instant of time at which the amplitude of the radiated echo is maximal. In the case of a narrow spectral line $1/T_0 \ll 1/T_i$, $i=1, 2$ the maximum of the echo occurs at the instant $t_e = 2\tau_s + T_1 + T_2 + z/c$. For a broad spectral line ($1/T_0 \gg 1/T_1$), the echo-pulse shape is more complicated and the echo maximum is usually shifted somewhat relative to the instant of time t_e .

3. DISCUSSION OF RESULTS

Expressions (26) and (27) yield the solution of our problem. To illustrate more clearly the influence of the depolarizing collisions on the photon-echo signal, we consider the case of resonance $\Delta\omega = 0$ and put in (26) and (27) $t = 2\tau_s + T_1 + T_2 + z/c$ for both narrow and broad spectral lines. We then obtain

$$e_x^e = 2 \cos^2[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s] |L^{(1)}(\tau_s)|^2 + i^{10} \sin^2[\frac{1}{2}(\varepsilon_a + \varepsilon_b)\tau_s] |L^{(2)}(\tau_s)|^2,$$

$$e_y^e = \sin[(\varepsilon_a + \varepsilon_b)\tau_s] (|L^{(1)}(\tau_s)|^2 - |L^{(2)}(\tau_s)|^2).$$

At low pressures, when the depolarizing collisions are inessential ($\gamma^{(2)} = \gamma^{(1)}$ and $\Delta^{(2)} = \Delta^{(1)} = 0$), expressions (29) and (30) take the form

$$e_x^e = \{1 + \cos[(\varepsilon_a + \varepsilon_b)\tau_s] \cos[(\varepsilon_a - \varepsilon_b)\tau_s]\} \exp(-2\gamma^{(1)}\tau_s),$$

$$e_y^e = \sin[(\varepsilon_a + \varepsilon_b)\tau_s] \cos[(\varepsilon_a - \varepsilon_b)\tau_s] \exp(-2\gamma^{(1)}\tau_s).$$

It follows from (31) and (32) that at a given time interval τ_s between the exciting pulses it is possible to choose the magnetic field intensity such that the echo at the instant of time $t = 2\tau_s + T_1 + T_2 + z/c$ is polarized along the polarization vectors of the exciting pulses. Let, for example, $(\varepsilon_a - \varepsilon_b)\tau_s = \pi/2$. Increasing then the pressure of the impurity or of the host gas, we can observe that the echo electric field intensity acquires at the instant of time $t = 2\tau_s + T_1 + T_2 + z/c$ a component perpendicular to the plane of polarization of the exciting pulses and due to the depolarizing collisions.

The results take the simplest form if the time interval τ_s between the pulses is chosen such that $|\gamma^{(1)} - \gamma^{(2)}|\tau_s \ll 1$. We note that it follows from the results of the calculation of $\gamma^{(1)}$ and $\gamma^{(2)}$ in Sec. I that at this value of τ_s we can have $(\gamma^{(1)} + \gamma^{(2)})\tau_s \lesssim 1$. Satisfaction of the last inequality is of importance for the observation of the photon-echo signal. In addition, we have considered the case $|\gamma^{(1)} - \gamma^{(2)}| \gg |\Delta^{(1)} - \Delta^{(2)}|$, which is realized at definite values of the Van der Waals constants. As a result we obtain for the echo amplitude from (31) and (32) the expressions:

$$e_x^e = \exp[-(\gamma^{(1)} + \gamma^{(2)})\tau_s],$$

$$e_y^e = -\frac{2}{\pi} (\gamma^{(1)} - \gamma^{(2)})\tau_s \sin\left[\frac{\pi(g_a + g_b)}{2(g_a - g_b)}\right] \exp[-(\gamma^{(1)} + \gamma^{(2)})\tau_s].$$

We emphasize that the direction of rotation of the echo polarization depends on the sign of $\gamma^{(1)} - \gamma^{(2)}$. At known g factors of the levels, the measurement of the slope of the photon-echo polarization relative to the polarizations of the exciting light pulses affords a direct experimental possibility of determining $\gamma^{(1)} - \gamma^{(2)}$. We

note that $\gamma^{(1)}$ is determined in the usual manner from the damping of the photon-echo intensity without the presence of a magnetic field, as a function of τ_s . The photon-echo method thus enables us to find $\gamma^{(1)}$ and $\gamma^{(2)}$ separately. This makes it possible to verify the theoretical relations that follow for these quantities from the assumed collision model, and consequently obtain information on the interaction of the atoms in the course of the collision.

The authors thank A. I. Alekseev for a discussion of the results.

APPENDIX

Equations (17)–(20) were used also to consider the formation of photon echo on the transitions $\frac{1}{2} \rightleftharpoons \frac{3}{2}$. We note that for these transitions the collisional relaxation of the components of the density matrix that describes the optical coherence are characterized by only two independent quantities, $\mathcal{J}^{(1)}$ and $\mathcal{J}^{(2)}$. We present the final result for the electric-field intensity of the photon echo in a gas situated in a longitudinal magnetic field. For the case of a narrow spectral line and exact resonance we have

$$E^e = -\frac{\pi}{16\sqrt{6}} \omega_0 \frac{L}{c} dN_0 \sin \Omega_1 T_1 (1 - \cos \Omega_2 T_2) \exp \left[-\frac{(t' - \tau_s)^2}{4T_0^2} \right] \times e^e \exp [i(\omega_0 t - kz + 2\Phi_s - \Phi_1)] + c.c., \quad (33)$$

where

$$\Omega_i^2 = 2|d|^2 e^{(i)2} / 3\hbar^2, \quad i=1, 2.$$

The polarization of the photon echo is determined by the direction of the vector e^e , whose nonzero components are

$$e_x^e = M_1 + M_{-1}, \quad e_y^e = -i(M_1 - M_{-1}). \quad (34)$$

The quantities M_q ($q = \pm 1$) have a complicated dependence on the relaxation characteristics and on the g factors of the considered levels. For the transition $j_a = \frac{3}{2}, j_b = \frac{1}{2}$ they are given by

$$M_q = \exp [iqe_a(t' - \tau_s)] [5L_q^{(1)} L_q^{(1)*} + q\sqrt{5}(L_q^{(1)} L_q^{(2)*} + L_q^{(2)} L_q^{(1)*}) + 5L_q^{(2)} L_q^{(2)*}] - \exp [iqe_a(t' + \tau_s)] [3L_q^{(1)} L_q^{(1)*} + q\sqrt{5}(L_q^{(1)} L_q^{(2)*} - L_q^{(2)} L_q^{(1)*}) + 5L_q^{(2)} L_q^{(2)*}], \quad q = \pm 1, \quad (35)$$

where

$$L_q^{(\kappa)} = L_q^{(\kappa)}(\tau_s), \quad L_q^{(\kappa)} = L_q^{(\kappa)}(t'), \quad \kappa=1, 2.$$

The functions $L_q^{(\kappa)}(t)$ are obtained by solving Eq. (18) following the action of each of the exciting pulses. We emphasize that the appearance of $L_q^{(\kappa)}(t)$ is due entirely to allowance for the difference between the g factors:

$$L_q^{(\kappa)}(t) = -iq \frac{3(\epsilon_b - \epsilon_a)}{4\sqrt{5}r_q} \text{sh}(r_q t) R(t).$$

Here

$$r_q = [\delta_q^2 - 3/16(\epsilon_b - \epsilon_a)^2]^{1/2}, \quad \delta_q = 1/2(\gamma^{(1)} - \gamma^{(2)}) - 1/2i[\Delta^{(1)} - \Delta^{(2)} - 1/2g(\epsilon_b - \epsilon_a)].$$

Conversely, if the g -factors of the levels are close to each other, then the main contribution to the echo polarization is made by the quantity

$$L_q^{(1)}(t) = q \left[\text{ch } r_q t - \frac{\delta_q}{r_q} \text{sh } r_q t \right] R(t).$$

The photon-echo electric-field intensity on the transition $j_a = \frac{1}{2}, j_b = \frac{3}{2}$ is obtained from (33)–(35) by the interchange $a \rightleftharpoons b$.

Expressions (33)–(35) yield information on the depolarizing collisions. In particular, by choosing for the $\frac{3}{2} \rightarrow \frac{1}{2}$ transition the magnetic field from the condition

$$\sin 2e_a \tau_s = 0,$$

we find that the maximum electric echo field component perpendicular to the polarization vectors of the exciting pulses is determined entirely by the depolarizing collisions. We note that it follows from (6) that in the case of a Van der Waals interaction we have $\mathcal{J}^{(1)} = \mathcal{J}^{(2)}$ for the transitions $\frac{1}{2} \rightleftharpoons \frac{3}{2} \mathcal{J}^{(2)}$. This result is the consequence of the matrix structure of the potential of the corresponding interaction.

We emphasize in conclusion that the method developed for the calculation of the photon-echo electric field intensity can be used also for other transitions, $j \rightarrow j$ ($j > 1$) and $j \rightarrow j+1$ ($j > \frac{1}{2}$), and yields information on the relaxation constants $\mathcal{J}^{(\kappa)}$.

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Translated by J. G. Adashko