Photoinduced magnetization of an atomic gas in a magnetic field

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The nonlinear magnetization induced in an atomic gas by an elliptically or circularly polarized light wave whose amplitude is an arbitrary function of time is calculated in the resonance approximation using perturbation theory. The limiting cases of a monochromatic traveling wave and a short pulse are studied. The nonlinear magnetization is studied as a function of the type of atomic transition, detuning from resonance, Doppler broadening, gas temperature, and relaxation constants. In the case of a short pulse a method is proposed for determining the widths of the resonance levels. The fundamental properties and the precession of the nonlinear magnetization in the presence of a magnetic field are found.

The magnetization of an atomic gas during the passage of an extended light wave or ultrashort light pulse has been observed in a number of experiments.[1–5] The results of a series of experiments performed with potassium or rubidium vapor both with and without a constant magnetic field **H** are reported in Refs. 1 and 4. In the case of resonance excitation, optical pumping redistributes the populations of the Zeeman sublevels of the gas atoms. This redistribution ultimately results in an uncompensated magnetic moment per unit volume of the atomic gas, and it is this uncompensated magnetic moment that is the photoinduced magnetization. In Refs. 1–5 a detecting coil, wound around a cell containing the experimental gas, was used to record this magnetization. The alternating photoinduced magnetization excited a current in the winding of this coil and produced a voltage at the terminals of the coil. The behavior of the photoinduced magnetization could be judged from this current and voltage. In addition to fundamental problems of the interaction of laser radiation with matter, this magnetooptic phenomenon has also been employed for observing the relaxation rates of the excited states of an atom.

In Refs. 1–5 the photoinduced magnetization was explained with the help of a phenomenological representation of the interaction of a light wave with an atomic gas. With the help of this representation it is possible to consider only separate aspects of the observed magneto-optic phenomenon, and many questions remained open. In this connection, as well as in view of the high sensitivity of the experimental method, as noted in Ref. 2, it is of interest to construct a detailed theory of photoinduced magnetization in order to both explain existing experimental data and formulate new experiments.

Nonlinear magnetization of an atomic gas upon resonance excitation of atoms by an arbitrarily polarized light wave was studied in the present work on the basis of a quantum-mechanical equation for the density matrix, taking into account the splitting of the levels in a constant magnetic field **H** and the flow of atoms into the lower level due to spontaneous emission in the upper level. Such a general approach to this problem made it possible to determine the basic laws of photoinduced magnetization and also to investigate its vector properties together with its quantitative characteristics.

It was established that without the magnetic field **H** a light wave induces magnetization only in the case of elliptic or circular polarization. In a magnetic field, however, a linearly polarized wave propagating in the direction of the field **H** also induces at the transition frequency ω_{ba} in the absence of resonance, $\omega \neq \omega_{ba}$, magnetization whose sign is revered by the substitution $\mathbf{H} \rightarrow -\mathbf{H}$ grows with increasing magnetic field in accordance with the experiment of Ref. 1, and decreases in sufficiently strong fields **H**. This photoinduced magnetization intensifies the magnetic field **H** for one sign of the detuning from resonance $\omega - \omega_{ba}$ and decreases the field for the opposite sign of $\omega - \omega_{ba}$, while at resonance $\omega = \omega_{ba}$ the photoinduced magnetization vanishes, owing to the symmetry of the Zeeman sublevels with positive and negative projections of the angular momentum.

The magnetization induced by an arbitrarily polarized light wave whose wave vector **k** is parallel to **H** consists of two parts, one an even function of **H** and of the detuning from resonance $\omega - \omega_{ba}$, while the other is an odd function of the same quantities. The direction of the odd part of the light-induced magnetization is determined by the direction of the vector **H**, while the direction of the even part depends on the direction of the wave vector **k** and the direction of rotation of the electric field **E** of the exciting light wave. In the case of resonance at the transition frequency, $\omega = \omega_{ba}$, replacement of right-hand elliptic or circular polarization with left-hand polarization changes the direction of the photoinduced magnetization, while near resonances with the Zeeman sublevels, in a sufficiently strong magnetic field **H** switching from right- to left-hand circular polarization does not change the direction of the photoinduced magnetization, in agreement with the experiment of Ref. 1.

In the case when an ultrashort light pulse propagates at an angle to the direction of the magnetic field the photoinduced magnetization precesses around **H** as a sum of separate magnetizations.

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1. COMPUTATION METHOD FOR ATOMS WITH SPINLESS NUCLEI

Consider a nonmagnetic magneto-optically inactive atomic gas.[6] Let a plane light wave pass through the gas and have a rotating electric-field vector $\mathbf{E}$

$$\mathbf{E}=\mathbf{l}_{\mathbf{k}\lambda}a\exp[i(\mathbf{kr}-\omega t)]+\text{c.c.}, \tag{1}$$

where $\mathbf{l}_{\mathbf{k}\lambda}$ is a unit complex elliptic-polarization vector

$$(\mathbf{l}_{\mathbf{k}\lambda}\mathbf{l}^*_{\mathbf{k}\lambda}=1, \quad \lambda=\pm 1)$$

and $a=a(t')$ is the complex amplitude, which is a function of $\mathbf{r}$ and t, thanks to the argument $t'=t-t_0-\mathbf{k}(\mathbf{r}-\mathbf{r}_0)/\omega$ and is a slowly varying function compared with $\exp[i(\mathbf{kr}-\omega t)]$. Here t_0 is the initial time at which the light wave (1) enters the atomic gas at the point $\mathbf{r}_0$, with the front of this incident wave in the boundary plane $\mathbf{k}(\mathbf{r}-\mathbf{r}_0)=0$ of some volume containing the given gas. The frequency ω is close to the frequency $\omega_{ba}=(E_b-E_a)/\hbar$ of the atomic transition between two states of an active atom with zero nuclear spin; these states, together with the other quantum numbers, are characterized by the energies E_a and E_b ($E_a<E_b$). The indicated frequency is related to the modulus of the wave vector $\mathbf{k}$ by the dispersion relation $\omega^2\varepsilon(\omega)=k^2c^2$, where c is the velocity of light in vacuum and $\varepsilon(\omega)$ is the real permittivity,[6] which accounts for the effect of both resonance levels of active atoms and nonresonance levels of active and impurity atoms of the given isotropic gas.

In studying the photoinduced magnetization the dependence of the elliptic polarization unit vector $\mathbf{l}_{\mathbf{k}\lambda}$ on $\mathbf{k}$ and on the parameter λ that determines the direction of rotation of $\mathbf{E}$, must be represented explicitly as

$$\mathbf{l}_{\mathbf{k}1}=\mathbf{l}^{(1)}_{\mathbf{k}}\cos\psi+i\mathbf{l}^{(2)}_{\mathbf{k}}\sin\psi \tag{2}$$

for right-hand polarization $\lambda=1$ and as

$$\mathbf{l}_{\mathbf{k},-1}=-\mathbf{l}^{(1)}_{\mathbf{k},\sin\psi}+i\mathbf{l}^{(2)}_{\mathbf{k},\cos\psi} \tag{3}$$

for left-hand polarization $\lambda=-1$. Here the argument ψ assumes the values $0\leqslant\psi\leqslant\pi/2$ and characterizes the semiaxes $\cos\psi(\sin\psi)$ and $\sin\psi(\cos\psi)$ of the polarization ellipses. In particular, for $\psi=\pi/4$ the polarization ellipses become circles and Eqs. (2) and (3) describe the polarization of a circular wave. For $\psi=0$ and $\psi=\pi/2$ the polarization ellipses in Eqs. (2) and (3) degenerate into orthogonal segments and Eqs. (2) and (3) describe two linearly polarized waves with orthogonal polarization planes. The other quantities in Eqs. (2) and (3) satisfy the relations

$$\mathbf{kl}^{(1)}_{\mathbf{k}}=\mathbf{kl}^{(2)}_{\mathbf{k}}=\mathbf{l}^{(1)}_{\mathbf{k}}\mathbf{l}^{(2)}_{\mathbf{k}}=0, \quad \mathbf{l}^{(1)}_{-\mathbf{k}}=\mathbf{l}^{(1)}_{\mathbf{k}}, \quad \mathbf{l}^{(2)}_{-\mathbf{k}}=-\mathbf{l}^{(2)}_{\mathbf{k}},$$

$$\mathbf{l}_{-\mathbf{k}\lambda}=\mathbf{l}^*_{\mathbf{k}\lambda}, \quad \mathbf{l}_{\mathbf{k}\lambda}\mathbf{l}^*_{\mathbf{k}\lambda'}=\delta_{\lambda\lambda'}, \quad [\mathbf{l}^{(1)}_{\mathbf{k}}\mathbf{l}^{(2)}_{\mathbf{k}}]=\beta\mathbf{k}/k,$$

$$[i\mathbf{l}_{\mathbf{k}\lambda}\mathbf{l}^*_{\mathbf{k}\lambda}]=(\mathbf{k}/k)\lambda\beta\sin(2\psi), \tag{4}$$

where β is the unit pseudoscalar, $\beta=+1$ and after inversion $\beta=-1$.

The resonance interaction of the active atom with the field $\mathbf{E}$ gives rise to redistribution of the populations of the Zeeman sublevels, which in turn gives rise to an uncompensated magnetic moment of the atom. As a result, the atomic gas acquires a magnetic moment $\boldsymbol{\mu}$ per unit volume. This moment is connected with the density matrix ρ in the JM representation by the well-known relation

$$\boldsymbol{\mu}=-\mu_B\int(g_a\rho_{M_aM'_a}\mathbf{J}_{M'_aM_a}+g_b\rho_{M_bM'_b}\mathbf{J}_{M'_bM_b})d\mathbf{v}, \tag{5}$$

where $\mu_B=|e|\hbar/2mc$ is the Bohr magneton, e and m are the electron charge and mass, g_a and g_b are the gyromagnetic factors (g factors), $\mathbf{v}$ is the velocity of the atom, $\rho_{M_aM'_b}(\rho_{M_bM'_b})$ and $\mathbf{J}_{M_aM'_a}(\mathbf{J}_{M_bM'_b})$ are, respectively, the matrix elements of the density matrix ρ and the angular momentum operator $\mathbf{J}$ in the lower (upper) level, and M_a and M_b are the quantum numbers of the projections $\mathbf{l}_z\mathbf{J}_a$ and $\mathbf{l}_z\mathbf{J}_b$ of the angular momenta $\mathbf{J}_a$ and $\mathbf{J}_b$ on the quantization axis, which is oriented along the unit vector $\mathbf{l}_z$ of the Cartesian z axis. Repeated vector indices are summed over.

In order to calculate the photoinduced magnetization (5), taking into account the splitting of the resonance levels into Zeeman sublevels

$$E_{M_a}=E_a+\Omega_aM_a\hbar, \quad E_{M_b}=E_b+\Omega_bM_b\hbar, \tag{6}$$

it is necessary first to solve the quantum-mechanical equations

$$\left[\frac{\partial}{\partial t}+\mathbf{v}\nabla+i(\omega_{ba}+\Omega_bM_b-\Omega_aM_a)+\gamma_{ba}\right]\rho_{M_bM_a}$$

$$=\frac{i}{\hbar}(\mathbf{Ed}_{M_bM'_a}\rho_{M'_aM_a}-\rho_{M_bM'_b}\mathbf{Ed}_{M'_bM_a}), \tag{7}$$

$$\left[\frac{\partial}{\partial t}+\mathbf{v}\nabla+i\Omega_b(M_b-M'_b)+\gamma_b\right]\rho_{M_bM'_b}$$

$$=\frac{\gamma_bN_bf(v)}{2J_b+1}\delta_{M_bM'_b}+\frac{i}{\hbar}(\mathbf{Ed}_{M_bM_a}\rho_{M_aM'_b}-\rho_{M_bM_a}\mathbf{Ed}_{M_aM'_b}), \tag{8}$$

$$\left[\frac{\partial}{\partial t}+\mathbf{v}\nabla+i\Omega_a(M_a-M'_a)+\gamma_a\right]\rho_{M_aM'_a}$$

$$=\frac{(\gamma_aN_a-\gamma N_b)f(v)}{2J_a+1}\delta_{M_aM'_a}+\frac{\gamma(2J_b+1)}{|d_{ba}|^2}\mathbf{d}_{M_aM_b}\rho_{M_bM'_b}$$

$$\times\mathbf{d}_{M'_bM'_a}+\frac{i}{\hbar}(\mathbf{Ed}_{M_aM_b}\rho_{M_bM'_a}-\rho_{M_aM_b}\mathbf{Ed}_{M_bM'_a}), \tag{9}$$

where

$$\Omega_a=g_a\mu_B(\mathbf{l}_z\mathbf{H})/\hbar, \quad \Omega_b=g_b\mu_B(\mathbf{l}_z\mathbf{H})/\hbar,$$

$$\gamma_{ba}=(\gamma_b+\gamma_a)/2, \quad \gamma=4|d_{ba}|^2\omega^3_{ba}/3\hbar c^3(2J_b+1),$$

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

$d_{M_bM_a}$ is the matrix element of the electric dipole moment operator d of the atom, γ_{ba} is the half-width of the spectral line of the resonance atomic transition, $\hbar\gamma_a$ and $\hbar\gamma_b$ are the homogeneous widths of the lower level E_a and upper level E_b, γ is the probability of spontaneous emission of a quan-

tum $\hbar\omega_{ba}$ by an isolated atom, d_{ba} is the reduced dipole moment,[7] J_a and J_b are the angular momentum quantum numbers in the lower and upper levels, $f(v)$ is Maxwell's distribution, v is the modulus of the velocity $\mathbf{v}$ of the atom, u is the most probable velocity, $\varkappa_B$ is Boltzmann's constant, T is the temperature of the gas, m_{at} is the mass of the active atom, and $N_a(N_b)$ is the stationary density of atoms in the level $E_a(E_b)$ and is determined by he Boltzmann distribution over the energy states in the absence of an external field ($\mathbf{E}=0$). The quantities γ_a and γ_b are determined by radiative decay and inelastic atomic collisions. The terms in Eq. (9) which contain γ describe the arrival of atoms in the lower level E_a due to spontaneous emission on the upper level E_b. Similarly, the terms that contain the factors $\gamma_b N_b f(v)$ and $\gamma_a N_a f(v)$ in Eqs. (8) and (9) describe the flow of atoms into the upper and lower levels, respectively, as a result of the statistical distribution over the energy states and velocities.

In Eqs. (7)–(9) the time origin is

$$t_0'=t_0+\mathbf{k}(\mathbf{r}-\mathbf{r}_0)/\omega,$$

in the initial values of the density matrix components are

$$\rho_{M_bM_a}=0, \quad \rho_{M_bM_b'}=\frac{N_b f(v)}{2J_b+1}\delta_{M_bM_b'},$$

$$\rho_{M_aM_a'}=\frac{N_a f(v)}{2J_a+1}\delta_{M_aM_a'},$$

where

$$\Omega_b|M_b-M_b'|, \quad \Omega_a|M_a-M_a'| \ll \varkappa_B T/2.$$

2. NONLINEAR MAGNETIZATION

The calculation of the photoinduced magnetization (5) for the light wave (1) propagating at an arbitrary angle with respect to $\mathbf{H}$ leads to a very complicated expression. For this reason, we consider the simplest case, when the light wave (1) is collinear with $\mathbf{H}$. Solving the equation for the density matrix by perturbation theory in the region $t_0' \leqslant t$ and using Eq. (5) in the case of the light wave (1) with arbitrary polarization and amplitude $a(t')$, depending arbitrarily on the time $t'=t-t_0'$, we obtain the following expression for the nonstationary photoinduced magnetization:

$$\begin{aligned}\boldsymbol{\mu}(t',\mathbf{H})=&-\mu_B N_{ab}|d_{ba}|^2(2\hbar^2)^{-1}\{\mathbf{l}_z[G(t',1)+G(t',-1)]\\&+(\mathbf{k}/k)\lambda\beta\sin(2\psi)\\&\times[G(t',1)-G(t',-1)]\}+\text{c.c.},\end{aligned} \tag{10}$$

where

$$N_{ab}=N_a/(2J_a+1)-N_b/(2J_b+1),$$

$$\begin{aligned}G(t',q)=&g_a\Phi_a(t',q)+g_b\Phi_b(t',q)\\&+\gamma g_a Q\Phi_{ab}(t',q)/(\gamma_b-\gamma_a),\end{aligned}$$

$$\Phi_a(t',q)=\sum_\mu(q-\mu)\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2 I_a^\mu(t',q), \tag{11}$$

$$\Phi_b(t',q)=\sum_\mu\mu\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2 I_b^\mu(t',q), \tag{12}$$

$$\Phi_{ab}(t',q)=\sum_\mu\mu\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2 [I_a^\mu(t',q)-I_b^\mu(t',q)], \tag{13}$$

$$\begin{aligned}Q=&(-1)^{J_a+J_b}(2J_b+1)\\&\times\left(\frac{J_a(J_a+1)(2J_a+1)}{J_b(J_b+1)(2J_b+1)}\right)^{1/2}\begin{Bmatrix}J_a & 1 & J_a\\ J_b & 1 & J_b\end{Bmatrix},\end{aligned}$$

$$\begin{aligned}I_a^\mu(t',q)=&\exp(-\gamma_a t')\int d\mathbf{v}f(v)\int_0^{t'}d\tau_2 a^*(\tau_2)\\&\times\exp\{[\gamma_a-\gamma_{ba}+i\Delta_\mu(q)]\tau_2\}\int_0^{\tau_2}d\tau_1 a(\tau_1\\&\times\exp\{[\gamma_{ba}-i\Delta_\mu(q)]\tau_1\},\end{aligned} \tag{14}$$

$$\Delta_\mu(q)=\omega-\omega_{ba}-\mathbf{kv}-\mu(\Omega_b-\Omega_a)-q\Omega_a, \quad q=\pm1.$$

The quantity $I_b^\mu(t',q)$ in Eqs. (12) and (13) is obtained from Eq. (14) by making the substitution $\gamma_a\to\gamma_b$. The standard notation is used for the $6j$ symbols.[7]

The expression (10) obtained above is suitable for an amplitude $a(t')$ of arbitrary form. In particular, for the stepped profile

$$a(t')=0 \quad \text{for } t'<0; \quad a(t')=a_0\exp(-i\alpha) \quad \text{for } 0\leqslant t',$$

where a_0 and α are constants, there exists a large time interval

$$1/\gamma_b \ll t' \ll 1/\gamma_a,$$

in which the nonstationary photoinduced magnetization (10) increases linearly with time:

$$\begin{aligned}\mu(t',\mathbf{H})=&-M_0\gamma_{ba}t'\{\mathbf{l}_z[R(1)+R(-1)]\\&+(\mathbf{k}/k)\lambda\beta\sin(2\psi)[R(1)-R(-1)]\},\end{aligned} \tag{15}$$

where

$$M_0=\mu_B N_{ab}|d_{ba}|^2 a_0^2/3\hbar^2\gamma_{ba}^2,$$

$$R(q)=g_a[\Phi_a(q)+\gamma Q\Phi_b(q)/\gamma_b],$$

$$\Phi_a(q)=3\sum_\mu(q-\mu)\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2 I_\mu(q), \tag{16}$$

$$\Phi_b(q)=3\sum_\mu\mu\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2 I_\mu(q), \tag{17}$$

$$I_\mu(q)=\int\frac{\gamma_{ba}^2 f(v)d\mathbf{v}}{[\Delta_\mu(q)]^2+\gamma_{ba}^2}, \quad q=\pm1. \tag{18}$$

In the case of a Maxwellian distribution $f(v)$ the integral (18) assumes the form

$$I_\mu(q)=Y(x_\mu(q),y), \tag{19}$$

where

$$x_\mu(q)=[\omega-\omega_{ba}-\mu(\Omega_b-\Omega_a)-q\Omega_a]/ku,$$

$$y=\gamma_{ba}/ku,$$

and the quantity $Y(x_\mu(q),y)$ can be expressed in terms of the well-known function $Y(x,y)$ with $x=x_\mu(q)$. This function is tabulated in Ref. 8 and has the form

$$Y(x,y)=\frac{y^2}{\pi^{1/2}}\int_{-\infty}^{\infty}\frac{\exp(-\xi^2)d\xi}{(x-\xi)^2+y^2}. \tag{20}$$

For long times

$$1\ll\gamma_a t', \quad 1\ll\gamma_b t',$$

the photoinduced magnetization (10) reaches a stationary state and describes the constant magnetization induced by a traveling monochromatic wave:

$$\mu(\mathbf{H})=-M_0\{l_z[G(1)+G(-1)]+(\mathbf{k}/k)\lambda\beta\sin(2\psi)\times[G(1)-G(-1)]\}, \tag{21}$$

where

$$G(q)=\gamma_{ba}[g_a\Phi_a(q)/\gamma_a+g_b\Phi_b(q)/\gamma_b+\gamma g_a Q\Phi_b(q)/\gamma_a\gamma_b], \quad q=\pm 1.$$

It is evident from Eqs. (16)–(20) that Doppler broadening significantly weakens the photoinduced magnetization (15) and (21), since at exact resonance with the transition frequency between the Zeeman sublevels, $x_\mu(q)=0$, we have

$$Y(0,y)=\pi^{1/2}y \tag{22}$$

for inhomogeneous broadening ($y\ll 1$) and

$$Y(0,y)=1 \tag{23}$$

for homogeneous broadening ($y^2\gg 1$).

Since the most probable velocity u is proportional to the square root of the gas temperature, the photoinduced magnetizations (15) and (21) decrease with increasing temperature as $T^{-1/2}$ if the resonance transition between the Zeeman sublevels is inhomogeneously broadened.

We now assume that an ultrashort pulse with small area (so that perturbation theory is applicable) passes through the atomic gas.[9] The duration τ of the pulse is small compared with the irreversible relaxation time of optical coherence

$$\gamma_{ba}\tau\ll 1, \tag{24}$$

which ensures that the following inequalities are also satisfied:

$$\gamma\tau\ll 1, \quad \gamma_a\tau\ll 1, \quad \gamma_b\tau\ll 1.$$

Thanks to the inequality (24) the nonstationary photoinduced magnetization (10) by the ultrashort pulse, is

$$\mu(t',\mathbf{H})=-\mu_B N_{ab}|d_{ba}|^2(2\hbar^2)^{-1}\{l_z[G_p(t',1)+G_p(t',-1)]+(\mathbf{k}/k)\lambda\beta\sin(2\psi)\times[G_p(t',1)-G_p(t',-1)]\}, \tag{25}$$

where

$$G_p(t',q)=g_a D_a(t',q)\exp(-\gamma_a t')+g_b D_b(t',q)\exp(-\gamma_b t')+[\gamma g_a Q D_b(t',q)/(\gamma_b-\gamma_a)]\times[\exp(-\gamma_a t')-\exp(-\gamma_b t')],$$

$$D_a(t',q)=\sum_\mu(q-\mu)\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2\Theta_\mu(t',q), \tag{26}$$

$$D_b(t',q)=\sum_\mu\mu\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2\Theta_\mu(t',q), \tag{27}$$

$$\Theta_\mu(t',q)=\int f(v)\left|\int_0^{t'}a(\xi)\times\exp[-i\Delta_\mu(q)\xi]d\xi\right|^2 dv, \quad q=\pm 1. \tag{28}$$

After expanding the brackets and braces, the photoinduced magnetization (25) separates into three different parts. The first two parts, which are proportional to the exponentials $\exp(-\gamma_a t)$ and $\exp(-\gamma_b t)$ without the factor γ, grow with time from zero to some maximum value at $t'\approx\tau$ (the first maximum of the photoinduced magnetization) and then decrease exponentially as $\exp(-\gamma_a t')$ and $\exp(-\gamma_b t')$, respectively. This decrease is determined by the fact that after the resonance excitation of the atoms the populations of the Zeeman sublevels return to their equilibrium value after the relaxation times $1/\gamma_a$ and $1/\gamma_b$. In contrast to this, after the action of the ultrashort light pulse the third part with the factor γ continues to increase with time because atoms continue to flow into the lower level due to spontaneous emission in the upper level, and after the maximum value is reached (second maximum of photoinduced magnetization) at the time

$$t'_m=(\gamma_b-\gamma_a)^{-1}\ln(\gamma_b/\gamma_a)$$

it is decreased by the relaxational processes described by the difference of two exponential functions. As a result the second maximum of the photoinduced magnetization (25) is delayed by the time t'_m, where $t'_m\gg\tau$ for $\gamma_b\gg\gamma_a$.

The exponential decay law obtained for the photoinduced magnetization (25) makes it possible to determine experimentally the relaxation constants γ_a and γ_b by measuring the nonlinear magnetization as a function of time for any value of $\mathbf{H}$. It should be kept in mind that, in the absence of a magnetic field, the photoinduced magnetizations (25) and (10), (15), and (21) are produced by the circularly or elliptically polarized light wave (1) but are absent in the case of linear polarization, since it is impossible to form from a single real linear-polarization vector an axial vector (4) that determines the direction of the magnetization (5). At the same time, a linearly polarized light wave (1) also induces in a magnetic field a magnetization equal to the term with the factor l_z in the braces (10), (15), (21), and (25), since $\sin(2\psi)=0$ for linear polarization.

3. FUNDAMENTAL PROPERTIES OF PHOTOINDUCED MAGNETIZATION

We can draw from the results (10)–(28) above the basic conclusions that follow from symmetry for gas atoms in external fields.

The substitution $\mathbf{H}\to-\mathbf{H}$ does not change the direction chosen in Eq. (6) for the quantization axis, so that the quantities Ω_a and Ω_b change sign. However, with the substitution $\Omega_a\to-\Omega_a$ and $\Omega_b\to-\Omega_b$ it is also reasonable to change the summation index, $\mu\to-\mu$, in Eqs. (11)–(13), (16), (17), (26), and (27). This leads to the following important transformations:

$$\Phi_a(t',q)\to-\Phi_a(t',-q),\quad \Phi_b(t',q)\to-\Phi_b(t',-q),$$
$$\Phi_{ab}(t',q)\to-\Phi_{ab}(t',-q), \tag{29}$$

$$\Phi_a(q)\to-\Phi_a(-q),\quad \Phi_b(q)\to-\Phi_b(-q),$$
$$G(q)\to-G(-q), \tag{30}$$

$$D_a(t',q)\to-D_a(t',-q),\quad D_b(t',q)\to-D_b(t',-q), \tag{31}$$

$$G(t',q)\to-G(t',-q),\quad G_{\mathbf{p}}(t',q)\to-G_{\mathbf{p}}(t',-q). \tag{32}$$

According to Eqs. (29)–(32), under the substitution $\mathbf{H}\to-\mathbf{H}$, the sum of the terms in the first brackets in Eqs. (10), (15), (21), and (25) changes sign and vanishes when $\mathbf{H}=0$; the expression in the second brackets is preserved and is different from zero when $\mathbf{H}=0$. In other words, the photoinduced magnetizations (10), (15), (21), and (25) separate into two parts, the first an odd and the second an even function of $\mathbf{H}$. For circular and elliptic polarizations both parts are different from zero, while for linear polarization only the odd part exists.

We emphasize that the direction of the odd part of the photoinduced magnetization (10), (15), (21), or (25) is determined by the orientation of the vector $\mathbf{H}$ and does not depend on the orientation of $\mathbf{k}$. This can be easily verified using as the example a stationary state for which the odd part of the photoinduced magnetization is

$$\boldsymbol{\mu}_{\text{odd}}(\mathbf{H})=-\mathbf{l}_z M_0[G(q)+G(-q)]$$
$$=-4\mathbf{H}(\omega-\omega_{ba})\cdot\mu_B M_0\hbar^{-1}\gamma_{ba}^2$$
$$\times\sum_\mu\int\frac{\Phi_{\mu q}[\mu(g_b-g_a)+qg_a]f(v)d\mathbf{v}}{[(\Delta+\Delta_{\mu q})^2+\gamma_{ba}^2][(\Delta-\Delta_{\mu q})^2+\gamma_{ba}^2]},$$

where

$$\Phi_{\mu q}=3\begin{pmatrix}J_b & J_a & 1\\ \mu & q-\mu & -q\end{pmatrix}^2\gamma_{ba}$$
$$\times\left[\frac{(q-\mu)g_a}{\gamma_a}+\frac{\mu}{\gamma_b}\left(g_b+\gamma\frac{g_a Q}{\gamma_a}\right)\right],$$

$$\Delta_{\mu q}=\mu(\Omega_b-\Omega_a)+q\Omega_a,\quad \Delta=\omega-\omega_{ba}-\mathbf{kv},\quad q=\pm1,$$

and the integral is an even function of $\mathbf{k}$.

The direction of the even part of the photoinduced magnetization (10), (15), (21), or (25) is determined by both the vector $\mathbf{k}$ and the direction of rotation of the electric field $\mathbf{E}$ of the light wave (1), which forms with the even part of the photoinduced magnetization a right-hand screw at $\lambda=-1$ and a left-hand screw at $\lambda=1$, i.e., the even parts of the indicated magnetizations is directed along $\mathbf{k}$ for $\lambda=-1$ and opposite to $\mathbf{k}$ for $\lambda=1$, if the difference in brackets is positive.

In the case when the detuning from resonance changes sign, $\omega-\omega_{ba}\to-(\omega-\omega_{ba})$, the sign of the summation index must be changed, $\mu\to-\mu$, in Eqs. (11)–(13), (16), (17), (26), and (27) and the sign of the integration variable must also be changed, $\mathbf{v}\to-\mathbf{v}$, in Eqs. (14), (18), and (28). Then we arrive once again at the transformations (29)–(32). This means that under the substitution $\omega-\omega_{ba}\to-(\omega-\omega_{ba})$ the photoinduced magnetizations (10), (15), (21), and (25) transform in the same manner as under the substitution $\mathbf{H}\to-\mathbf{H}$, with separation into even and odd parts with respect to $\omega-\omega_{ba}$.

The substitutions $\mathbf{l}_x\to-\mathbf{l}_x$, $\mathbf{l}_y\to-\mathbf{l}_y$ and $\mathbf{l}_z\to-\mathbf{l}_z$ are accompanied by the transformation (29)–(32). Together with this, $\mathbf{k}$ and the unit pseudoscalar β change sign, so that the photoinduced magnetizations (10), (15), (21), and (25) transform as axial vectors.

After the magnetic field $\mathbf{H}$ is switched on, the odd part of the photoinduced magnetizations (10), (15), (21), or (25) in the region

$$|\Delta_{\mu q}|<\max(|\omega-\omega_{ba}|, ku,\gamma_{ba})$$

at first grows in proportion to $\mathbf{H}$. It varies next according to a different law, and as the magnetic field increases further, it approaches zero in the region

$$|\Delta_{\mu q}|\gg\max(|\omega-\omega_{ba}|, ku,\gamma_{ba}),$$

where $q=\pm1$. The magnetization induced by a linearly polarized light wave (1) behaves in the same manner. This magnetization enhances the external field $\mathbf{H}$ for one sign of the detuning from resonance $\omega-\omega_{ba}$ and diminishes the external field for the opposite sign of $\omega-\omega_{ba}$. However, a detailed study of this opposite effect of photoinduced magnetization on the internal magnetic field in a gas requires a separate analysis.

In the case of resonance at the transition frequency $\omega=\omega_{ba}$ the Zeeman sublevels with negative and positive projections of the angular momentum enter symmetrically in all formulas. This has interesting consequences. To see this, we change the sign of the summation index, $\mu\to-\mu$, in Eqs. (11)–(13), (16), (17), (26), and (27) and simul-

taneously the sign of the integration variable $\mathbf{v}\to-\mathbf{v}$ in Eqs. (14), (18), and (28). We obtain, using the fact that $\omega=\omega_{ba}$,

$$G(t',q)=-G(t',-q),\quad R(q)=-R(-q),$$

$$G(q)=-G(-q),\quad G_{\mathrm{p}}(t',q)=-G_{\mathrm{p}}(t',-q). \tag{33}$$

Thus the equivalence of the Zeeman sublevels when $\omega=\omega_{ba}$ implies that the photoinduced magnetizations (10), (15), (21), and (25) are different from zero only for circular and elliptic polarizations, and a linearly polarized light wave in the presence of a magnetic field $\mathbf{H}$ does not induce magnetization, just as in the case $\mathbf{H}=0$.

It also follows from Eq. (33) that replacing in the light wave (1) the right-hand polarization $\lambda=1$ by left-hand polarization $\lambda=-1$ (circular or elliptic) at the transition frequency $\omega=\omega_{ba}$ changes the direction of the photoinduced magnetizations (10), (15), (21), and (25) for any value of $\mathbf{H}$, just as for $\mathbf{H}=0$ and arbitrary detuning from resonance $\omega-\omega_{ba}$.

To investigate the photoinduced magnetization near Zeeman resonances we set $\mathbf{k}/k=\mathbf{l}_z$ and $\mathbf{l}_z\|\mathbf{H}$ and write the expression in the braces in Eq. (21) in a different form:

$$\mathbf{l}_z[G(1)+G(-1)]+(\mathbf{k}/k)\lambda\beta\sin(2\psi)[G(1)-G(-1)]$$

$$=\mathbf{l}_z[(\sin\psi+\lambda\beta\cos\psi)^2G(1)$$

$$+(\sin\psi-\lambda\beta\cos\psi)^2G(-1)] \tag{34}$$

and make the same transformation in Eq. (15), which leads to the expression (34) with the substitution $G(q)\to R(q)$, where $q=\pm1$.

For the light wave (1) with right-hand circular polarization $\lambda=1$ and $\psi=\pi/4$ the expression (34) is $2\mathbf{l}_zG(1)$, where in the absence of inversion we set $\beta=1$. This means that for the Zeeman sublevels (6) the selection rule $M_b-M_a=1$ is satisfied, and for right-hand circular polarization a resonance is realized at the frequency

$$\omega=\omega_{ba}+M_b(\Omega_b-\Omega_a)+\Omega_a. \tag{35}$$

In a sufficiently strong magnetic field

$$\Omega_a^2,\Omega_b^2\gg\max[\gamma_{ba}^2,(ku)^2]$$

near resonance (35) all quantities in Eqs. (10)–(28) with arbitrary ψ and $q=-1$ are less than the corresponding quantities with $q=1$ by a factor of γ_{ba}^2/Ω_a^2 or γ_{ba}^2/Ω_b^2 for homogeneous broadening ($\gamma_{ba}^2\gg(ku)^2$), and by a factor of $\gamma_{ba}ku/\Omega_a^2$ or $\gamma_{ba}ku/\Omega_b^2$ for inhomogeneous broadening ($\gamma_{ba}\ll ku$). This makes it possible to drop all small terms with $q=-1$ and retain in the quantities with $q=1$ only resonance terms with $\mu=M_b$.

Similarly, for the light wave (1) with left-hand circular polarization $\lambda=-1$ and $\psi=\pi/4$ the value of (34) is $2\mathbf{l}_zG(-1)$. For this reason the selection rule $M_b-M_a=-1$ is satisfied in Eq. (6), and resonance is realized for left-hand polarization at the frequency

$$\omega=\omega_{ba}+M_b(\Omega_b-\Omega_a)-\Omega_a. \tag{36}$$

In the indicated strong magnetic field, the small quantities with $q=1$ can be dropped in Eqs. (10)–(28) with arbitrary ψ near resonance (36), and only resonance terms with $\mu=M_b$ need to be retained in terms with $q=-1$.

Thus near the resonances (35) and (36) in the frequency interval

$$[\omega-\omega_{ba}-M_b(\Omega_b-\Omega_a)\mp\Omega_a]^2\lesssim\max[\gamma_{ba}^2,(ku)^2]$$

the quantities $G(1)$ and $G(-1)$ have in a sufficiently strong magnetic field the same sign for resonance terms with $\mu=M_b$, so that, in contradistinction to the case of resonance at the transition frequency $\omega=\omega_{ba}$, replacement of right-hand circular polarization $\lambda=1$ and $\psi=\pi/4$ in the light wave (1) by left-hand circular polarization $\lambda=-1$ and $\psi=\pi/4$ does not, according to Eq. (34), change the direction of the photoinduced magnetizations (15) and (21).

For the resonances (35) and (36) in the cases Eqs. (15) and (21), the effect of Doppler broadening is described by the integral (18), which satisfies Eqs. (22) and (23). For this reason, the photoinduced magnetizations (15) and (21) decrease with increasing gas temperature for an inhomogeneously broadened line.

It is known that right- and left-hand circularly polarized waves are natural modes in the case of propagation parallel to $\mathbf{H}$. In the case of the light wave (1) with arbitrary polarization the transformation (34) means that the photoinduced magnetizations (15) and (21) separate into two parts, referring to the indicated natural modes.

With the help of a transformation similar to (34) it is also possible to resolve the photoinduced magnetizations (10) and (25) into natural modes in order to study their behavior near the resonances (35) and (36). As a result we find that in a sufficiently strong magnetic field near resonance (35) only resonance terms with $q=1$, $\mu=M_b$, and arbitrary ψ remain in Eqs. (10)–(14) and (25)–(28), and terms with $q=-1$ must be dropped. Similarly, near resonance (36) only terms with $q=-1$, $\mu=M_b$, and arbitrary ψ must be retained, while terms with $q=1$ can be dropped. For Eqs. (10) and (25) the law of conservation of the direction of the photoinduced magnetization remains in force on going from right- to left-hand circular polarization, and the photoinduced magnetization likewise decreases with increasing gas temperature in the case of inhomogeneous broadening.

The frequency dependences of the photoinduced magnetization in the presence of a magnetic field $\mathbf{H}$ is significantly different from the case when there is no field. In particular, in a strong magnetic field the magnetization decreases when the frequency ω deviates from the resonances (35) and (36), so that as the frequency ω is scanned, separate peaks of the photoinduced magnetization, corresponding to the resonances (35) and (36) with different values of M_b, appear.

Far from all resonances

$$(\omega-\omega_{ba})^2\gg[M_b(\Omega_b-\Omega_a)\pm\Omega_a]^2$$

the quantity $\mu(\Omega_b-\Omega_a)+q\Omega_a$ can be neglected and then, after summing over the index μ, the expressions for the magnetizations (10), (15), (21), and (25) describe nonlinear magnetization in the absence of a magnetic field.

The fundamental properties found above for photoinduced magnetization in an atomic gas are different from the properties of photoinduced magnetization in solids, studied in Refs. 10–14, because of both the symmetry and the active centers (atoms) are different.

4. PRECESSION OF PHOTOINDUCED MAGNETIZATION IN A MAGNETIC FIELD

For an arbitrary angle θ between the vectors $\mathbf{k}$ and $\mathbf{H}$ the photoinduced magnetization has in the case of an ultrashort light pulse (1) of duration τ, a simple form which satisfies together with the inequality (24) the requirements

$$J_a\Omega_a\tau \ll 1, \quad J_b\Omega_b\tau \ll 1. \tag{37}$$

Thanks to the inequalities (37) the effect of $\mathbf{H}$ on the density matrix $\rho(t')$ in Eqs. (7)–(9) in the time interval $0 \leqslant t' \leqslant \tau$ can be neglected, while in the region $\tau < t'$ Eqs. (7)–(9) describe the behavior of atoms in the magnetic field $\mathbf{H}$ after the passage of an ultrashort pulse. In the region $\tau \leqslant t'$ the initial condition is the density matrix $\rho(\tau)$ describing active atoms at the time $t'=\tau$ when the action of the short pulse ceases.

As a result, the photoinduced magnetization (5) in the magnetic field $\mathbf{H}$ for $0 \leqslant t'$ assumes the form

$$\boldsymbol{\mu}(t',\mathbf{H}) = \mathbf{l}_x\mu_x(t',\mathbf{H}) + \mathbf{l}_y\mu_y(t',\mathbf{H}) + \mathbf{l}_z\mu_z(t',\mathbf{H}), \tag{38}$$

where

$$\begin{aligned}
\mu_x(t',\mathbf{H}) &= \sin\theta[M_a(t')\cos(\Omega_a t'+\varphi) + M_b(t') \\
&\times\cos(\Omega_b t'+\varphi) + M_{a\gamma}(t',\mathbf{H})\cos(\Omega_a t'+\Phi \\
&+\varphi) + M_{b\gamma}(t',\mathbf{H})\cos(\Omega_b t'+\Phi+\varphi)], \\
\mu_y(t',\mathbf{H}) &= \sin\theta[M_a(t')\sin(\Omega_a t'+\varphi) + M_b(t') \\
&\times\sin(\Omega_b t'+\varphi) + M_{a\gamma}(t',\mathbf{H})\sin(\Omega_a t'+\Phi \\
&+\varphi) + M_{b\gamma}(t',\mathbf{H})\sin(\Omega_b t'+\Phi+\varphi)], \\
\mu_z(t',\mathbf{H}) &= \cos\theta[M_a(t') + M_b(t') + M_{a\gamma}(t',0) \\
&+ M_{b\gamma}(t',0)],
\end{aligned}$$

$$M_a(t') = (\mathbf{k}/k)M_a(t'), \quad M_{a\gamma}(t',\mathbf{H}) = (\mathbf{k}/k)M_{a\gamma}(t',\mathbf{H}),$$

$$M_a(t') = -\lambda\beta\sin(2\psi)M(t')g_aQ_a\exp(-\gamma_a t'),$$

$$\begin{aligned}
M_{a\gamma}(t',\mathbf{H}) &= -\lambda\beta\sin(2\psi)M(t')g_aQ_{ab}\gamma(\gamma_b-\gamma_a)^{-1} \\
&\times\cos\Phi\exp(-\gamma_a t'),
\end{aligned}$$

$$M(t') = \mu_B N_{ab}|d_{ba}|^2(6\hbar^2)^{-1}\int f(v)\left|\int_0^{t'} a(\xi) \times\exp(-i\Delta\xi)d\xi\right|^2 dv,$$

$$\sin\Phi = (\Omega_b-\Omega_a)[(\gamma_b-\gamma_a)^2+(\Omega_b-\Omega_a)^2]^{-1/2},$$

$$\cos\Phi = (\gamma_b-\gamma_a)[(\gamma_b-\gamma_a)^2+(\Omega_b-\Omega_a)^2]^{-1/2}.$$

Here $M_b(t')$ differs from $M_a(t')$ in that b is replaced by a, while $M_{b\gamma}(t',\mathbf{H})$ is obtained from $M_{a\gamma}(t',\mathbf{H})$ by replacing $\exp(-\gamma_a t')$ by $-\exp(-\gamma_b t')$. The polar angle θ and the azimuthal angle φ determine the direction of k in a spherical coordinate system whose polar axis z is parallel to $\mathbf{H}$. The quantities Q_a, Q_b, and Q_{ab} assume the following values, depending on the type of atomic transition $J_a \to J_b$:

1) for $J_a = J \to J_b = J$ $(J>0)$

$Q_a = Q_b = 1$, $Q_{ab} = (J^2+J-1)/J(J+1)$;

2) for $J_a = J \to J_b = J+1$

$Q_a = -J$, $Q_b = J+2$, $Q_{ab} = J(J+2)/(J+1)$;

3) for $J_a = J+1 \to J_b = J$

$Q_a = J+2$, $Q_b = -J$, $Q_{ab} = -J(J+2)/(J+1)$.

After the passage of the ultrashort pulse in the region $\tau \leqslant t'$ Eq. (38) describes the complicated precession of the photoinduced magnetization in the magnetic field $\mathbf{H}$. This precession is the sum of the precessions of separate independent vectors $\mathbf{M}_a(t')$, $\mathbf{M}_b(t')$, $\mathbf{M}_{a\gamma}(t',\mathbf{H})$ and $\mathbf{M}_{b\gamma}(t',\mathbf{H})$. In addition, the magnetizations $\mathbf{M}_{a\gamma}(t',\mathbf{H})$ and $\mathbf{M}_{b\gamma}(t',\mathbf{H})$ are determined by the flow of atoms into the lower level due to spontaneous emission in the upper level after passage of the exciting pulse. The quantities $M_a(t')$ and $M_b(t')$ reach their maximum values at the time $t' \approx \tau$, while the maximum of the sum $M_{a\gamma}(t',0)+M_{b\gamma}(t',0)$ is delayed in time by an amount t'_m. The obtained precession of photoinduced magnetization is significantly different from the model variant proposed in Ref. 4.

With the substitution $\mathbf{H} \to -\mathbf{H}$ the quantities Ω_a, Ω_b, and Φ in Eq. (38) change sign, so that right-hand rotation of the magnetization vectors changes to left-hand rotation. If we set $\mathbf{H}=0$, then the photoinduced magnetization (38) is

$$\boldsymbol{\mu}(t',0) = \mathbf{M}_a(t') + \mathbf{M}_b(t') + \mathbf{M}_{a\gamma}(t',0) + \mathbf{M}_{b\gamma}(t',0),$$

which is identical to Eq. (25) in the absence of a magnetic field.

5. DISCUSSION

The series of experiments performed in Ref. 1 with potassium and rubidium vapors with resonance excitation of atoms by an ultrashort pulse established that in the case $\omega \neq \omega_{ba}$ and a linearly polarized light wave the photoinduced magnetization increases as $\mathbf{H}$ and it also changes sign under the substitution $\mathbf{H} \to -\mathbf{H}$ in a wide range of values of $\mathbf{H}$. In addition, it was found that the direction of magnetization does not change when the polarization of the ultrashort pulse is changed from right- to left-hand polarization in a strong magnetic field near Zeeman resonances. These experimental results are in complete agreement with the theoretical results and are solely symmetry determined.

The experiments of Refs. 1–5 were performed on atoms with hyperfine structure. This greatly complicates the formulas for the photoinduced magnetization. In particular, new terms, which correspond to resonance at zero frequency and which are absent for atoms with spinless nuclei, appear in the magnetization terms determined by the

spontaneous emission of atoms in the upper level. Similar resonance terms make an appreciable contribution to self-diffraction and phase conjugation.[15] However, in spite of the fact that the formulas for the photoinduced magnetization become much more complicated, the fundamental properties found to be associated with symmetry remain for atoms with hyperfine structure. The law of decay of photoinduced magnetization after the passage of an ultrashort light pulse also remains in force. This provided a basis for comparing the theoretical results with the experimental results.[1-5]

It should be kept in mind, however, that for nuclei with nonzero spin the frequency dependence of the photoinduced magnetization is much more complicated, both in the case $\mathbf{H}=0$ and for $\mathbf{H}\neq 0$, because of resonances with the hyperfine components. These resonances also influence the temperature dependence of the magnetization, especially in cases when the splitting of the hyperfine components is of the order of the Doppler width multiplied by $\hbar$. In addition, owing to the smallness of the hyperfine splitting, the Paschen–Back effect appears in comparatively weak fields $\mathbf{H}$. For this reason, separate calculations, taking into account the hyperfine structure, must be performed in order to analyze the frequency and temperature dependences of the photoinduced magnetization in the experiment of Ref. 1.

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