## Mechanisms of precession of a light-induced quadrupole moment of the ground state of atoms with a hyperfine structure in a weak magnetic field and their manifestations in optical characteristics

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An analysis is made of a novel physical effect in the form of free and forced precession of an optically induced quadrupole moment in a weak magnetic field in the absence of a light-induced magnetic moment. The problem is solved for the ground state of an atom allowing for the hyperfine splitting and the Doppler broadening. It is shown that the bounded nature of the light flux gives rise to qualitatively new behavior of the precession relaxation processes. It is suggested that this effect can be observed by utilizing a different novel physical effect associated with the self-rotation of the plane of polarization of light in a weak magnetic field.

1. When atoms are oriented in the ground state by a circularly polarized optical field, a magnetic moment is induced in the system.<sup>1-3</sup> Abrupt application of an external magnetic field H results in free precession of an induced magnetic moment around the direction of H and the frequency of such precession is the Larmor value  $\Omega$  (Refs. 4– 6). Such a simple and clear precession pattern is observed only if the moment of the ground state is J = 1/2. If  $J \ge 1$  or in the case of hyperfine splitting of the ground state  $F \ge 1$  the application of a magnetic field induces not only a magnetic moment, but also multipole moments of higher rank, x > 1, which precess at frequencies  $q\Omega$ , where  $|q| \leq \varkappa$ . For example, a quadrupole moment (x = 2) gives rise to two frequencies in the spectrum:  $\Omega$  and  $2\Omega$ . Hyperfine splitting complicates the spectrum because of the difference between the  $g_n$  factors of the hyperfine components (n is the number of the component). Precession relaxation is usually attributed to depolarizing collisions<sup>3</sup> in the ground state the rates of which are  $\gamma_{\mu}^{0}$  $\sim 10^2 - 10^{-2} \, \mathrm{s}^{-1}$  (Ref. 4).

In the case of orientation by a laser field the rates  $\gamma_{\pi}^{0}$  are low compared with all the characteristic frequencies of the problem. Therefore, relaxation of precession is then governed by other processes, principal among which is a transit effect related to the time of interaction of an atom with a beam of light  $\overline{t} = r_0 / \overline{v}$ , where  $r_0$  is the transverse size of the beam and  $\overline{v}$  is the average velocity. Clearly,  $\gamma_{x}^{0} \overline{t} \ll 1$  and we can then ignore the depolarizing collisions. However, we are not allowed to make the simple substitution  $\gamma_{\kappa}^0 \rightarrow 1/\bar{t}$  and thus correct the problem,<sup>7</sup> because this substitution postulates a priori that the relaxation process is exponential. We shall show below that a finite interaction time results in a slower decay of the precession:  $\propto 1/t$ . In addition to the relaxation processes, we shall draw attention to a novel physical effect of free and forced precession of an optically induced quadrupole moment in a magnetic field when there is no induced magnetic moment. We shall consider the resonant interaction of an atom with a linearly polarized light beam. An external magnetic field is assumed to be so low that we can ignore the Faraday and Hanle effects:  $\Omega \ll \gamma, k\overline{v}$ , where  $2\gamma$  is the rate of relaxation of the upper resonating level and  $k\overline{v}$  is the Doppler width. The third condition, which guarantees the absence of a magnetic moment, is the low intensity of the optical field  $\gamma t G < 1$  when  $\gamma t \gg 1$ ; G is the

saturation parameter described by Eq. (7) below. For example, in the case of alkali metals these conditions can be satisfied if the intensity of a light beam is within the range  $10^{-6}$  mW/mm<sup>2</sup>  $\leq I \leq 10^{-1}$  mW/mm<sup>2</sup>. The intensity of the radiation emitted by cw tunable dye lasers makes it possible to satisfy this condition easily. In this approximation in respect of the field, there is an additional novel physical effect associated with the self-rotation of the plane of polarization,<sup>8</sup> which could be used to study the precession of the quadrupole moment.

2. We shall now formulate the problem.<sup>8</sup> In the quantum part of the problem we shall consider a resonant interaction of a gas of atoms with a given linearly polarized field E. The resonating levels of these atoms, one of which is the ground state, are split by the hyperfine interaction. It is assumed that the optical density of a gas is low:  $\gamma l < 1$ , where  $\gamma$ is the linear absorption coefficient and l is the length of the cell, obeying the condition  $r_0/l \ll 1$ , so that we can ignore all types of collisions and self-consistency with the electrodynamic part of the problem. The gas of atoms can be described by the formalism of the density matrix expanded in terms of irreducible tensor operators ( $\chi$ , q representation<sup>9</sup>). The relationship between the characteristic frequencies in our model corresponds to the usual experimental conditions<sup>5-7</sup>: the Doppler broadening  $k\overline{v}$  and the hyperfine splitting of the ground state  $\omega_{nn_1}$  should be greater than  $\gamma$  and the hyperfine splitting of the upper level should obey  $\omega_{mm_1} \gtrsim \gamma$ . This relationship between the frequencies allows us to consider the upper level as virtual and active in a redistribution of atoms between the magnetic sublevels of the ground state (optical self-pumping) as a result of which an induced quadrupole moment appears for atoms in the ground state. The populations of the upper level can be ignored in view of  $1/\gamma t \ll 1$ .

The electrodynamic part of the problem is used to detect precession of a quadrupole using oscillations of the polarization characteristics of radiation transmitted by a cell containing a gas. Therefore, we shall consider the boundary-value problem relating the initial linearly polarized field at y = 0 to the transmitted field at y = l. The solution of the boundary-value problem can be obtained using perturbation theory for a given polarization (susceptibility) of a medium when the field component E' is orthogonal to E. We shall use a coordinate system (x, y, z) with a suitable basis  $(e_1, e_2, e_3)$ 

where the direction of propagation of a wave is given by the unit vector  $\mathbf{e}_2$ , the initial polarization is given by the unit vector  $\mathbf{e}_3$ , and the direction of the magnetic field **h** is described by the spherical angles  $\theta$  and  $\varphi$ . In this system of coordinates the radiation field transmitted by a medium is elliptically polarized and is characterized by a single complex vector

$$\mu = \frac{(\mathbf{e}_3 \cos \psi + \mathbf{e}_1 \sin \psi) + i \operatorname{tg} \alpha (\mathbf{e}_1 \cos \psi - \mathbf{e}_3 \sin \psi)}{(1 + \operatorname{tg}^2 \alpha)^{\frac{1}{2}}}, \qquad (1)$$

where  $\psi$  is the angle of rotation of the plane of polarization and  $\alpha$  is the ellipticity angle.<sup>10</sup> In the model under discussion, we have  $\alpha$ ,  $\psi \ll 1$  and the component E' is proportional to the quantity  $\psi + i\alpha$ , which in the boundary-value problem is related to the susceptibility component  $\chi_{13}$  by

$$\psi + i\alpha = 2\pi i k l \chi_{13} = -\chi l \xi_{13}.$$

$$(2)$$

**3.** The approximations used in the formulation of the problem allow us to write down the kinetic equation for the polarization moments  $\rho_{xq}^{(n)}$  of the ground-state density matrix:

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right) \rho_{\mathbf{x}}^{(n)} - ig_{n}\Omega\eta(t) \left[\varkappa(\varkappa+1)\right]^{\nu_{2}} \left\{h_{1}\otimes\rho_{\mathbf{x}}^{(n)}\right\}_{\mathbf{x}}$$
$$= \gamma G\left\{\mu_{1}^{*}\otimes\mu_{1}\right\}_{\mathbf{x}} A_{n}^{(\mathbf{x})}f(\mathbf{r})p(t),$$
(3)

where

$$A_{n}^{(x)} = \sum_{n_{i}} \left( A_{x}^{(1)nn_{i}} - A_{x}^{(2)nn_{i}} \right),$$

$$A_{x}^{(1)nn_{i}} = 3 \sum_{m} (-1)^{F_{n} - F_{n_{i}} + 1} (2J_{1} + 1) \frac{N^{(n_{i})} C_{mn} C_{mn_{i}}}{(2F_{n_{i}} + 1) (1 + \Delta_{mn_{i}}^{2})}$$

$$\times \left\{ \frac{\varkappa}{1} \frac{F_{n}}{F_{m}} \frac{F_{n}}{F_{m}} \right\} \left\{ \frac{\varkappa}{F_{n_{i}}} \frac{F_{m}}{1} \frac{F_{m}}{1} \right\}.$$
(4)

$$A_{\pi}^{(2)nn_{1}} = 3\delta_{nn_{1}} \sum_{m} (-1)^{F_{n}+F_{m}} \frac{N^{(n_{1})}C_{mn_{1}}}{(2F_{n_{1}}+1)(1+\Delta_{mn_{1}}^{2})} \times \left\{ \frac{\kappa}{F_{m}} \frac{F_{n}}{1} \frac{F_{n}}{1} \right\}.$$
 (5)

The notation for the irreducible tensor products and the 3jn symbols follows Ref. 11. The summation in Eqs. (4) and (5) is over the hyperfine structure of an excited state with an index m. The constants  $C_{mn}$  in Eqs. (4) and (5) and the value of  $g_n$  in Eq. (3) give the dependences of the dipole moment of the d and g factors of the ground state on the total moments F, the nuclear spin I, and the electron moments  $J_0$  and  $J_1$  (0 refers to the lower state and 1 refers to the upper state):

$$C_{mn} = (2F_m + 1) (2F_n + 1) \left\{ \begin{matrix} I & J_0 & F_n \\ 1 & F_m & J_1 \end{matrix} \right\}^2 ,$$
  
$$g_n = (-1)^{F_n + J_0 + I + 1} g_0 (2F_n + 1) \left\{ \begin{matrix} I & J_0 & F_n \\ 1 & F_n & J_0 \end{matrix} \right\} .$$
(6)

The saturation parameter G and the spectral components  $\Delta_{mn}$  obtained using the hyperfine splitting and the Doppler shift are described by the following expressions:

$$G = \left| \frac{Ed}{\hbar \gamma} \right|^2, \quad \Delta_{mn} = (\omega - \omega_{mn} - \mathbf{k}\mathbf{v})/\gamma \tag{7}$$

(frequencies are measured from the field frequency  $\omega$ ). The terms  $A^{(1)}$  and  $A^{(2)}$ , in Eqs. (3)–(5) describe optical pumping of the ground state, i.e., the arrival in the ground state  $A^{(1)}$  as a result of spontaneous emission and departure from the ground state  $A^{(2)}$ , because of an external field.

Equation (3) contains dimensionless functions  $f(\mathbf{r})$ , p(t), and  $\eta(t)$  which describe, respectively, the transverse distribution of the intensity in a light beam  $f(\mathbf{r})$ , the time dependence of the intensity p(t), and the time dependence  $\eta(t)$  of the magnetic field intensity.

We derived the kinetic equation (3) bearing in mind the following points. The right-hand side of Eq. (3) generally depends on all the polarization moments  $\rho_{\chi}^{(n)}$ . Since the solution of Eq. (3) should be obtained in the first order with respect to the saturation parameter G, the polarization moments on the right-hand side of Eq. (3) are assumed to be equal to the initial values<sup>8</sup> (corresponding to adiabatic activation of the interaction):

$$\rho_{x}^{(n)} = \delta_{x,0} \frac{N^{(n)}}{(2F_{n}+1)^{\frac{1}{2}}}, \quad \sum_{n} N^{(n)} = 1,$$
(8)

where  $N^{(n)}$  are the equilibrium population numbers of the hyperfine structure of the ground state. For this reason the unit field vectors  $\mu$  in Eq. (3) are equal to their initial values  $(\mu = e_3)$ .

We can see from Eq. (3), as stressed above, that the adopted approximation gives only the polarization moments of the even rank ( $\kappa = 0$  or 2) for  $\rho_{\kappa}^{(n)}$ ; the odd moments can appear only in the next approximation in terms of G.

4. The solution of the kinetic equation (3) can be represented by a sum of the free solution (8) and the forced solution  $\tilde{\rho}_{x}^{(n)}$  describing all the precession effects. We shall now give the expression describing the forced solution:

$$\mathfrak{p}_{\mathbf{x}}^{(n)} = \gamma G \sum_{L} A_{n}^{(\mathbf{x})} \left\{ \left\{ e_{1}^{(3)} \otimes e_{1}^{(3)} \right\}_{\mathbf{x}} \otimes h_{L} \right\}_{\mathbf{x}} t_{\mathbf{x},L}^{(n)} (\mathbf{r}, \mathbf{v}_{\perp}, t),$$

$$t_{\mathbf{x},L}^{(n)} = \sum_{q} (-1)^{q} C_{\mathbf{x}-q,\mathbf{x}q}^{L_{0}} \int_{0}^{\infty} dt' f(\mathbf{r} - \mathbf{v}_{\perp}t') p(t-t')$$

$$(9)$$

$$\operatorname{\mathsf{x}exp}\{-iq\Omega_n(t,t')\}.$$
 (10)

$$\Omega_n(t,t') = \Omega g_n \int_{t-t'}^t dt'' \eta(t'').$$
(11)

Here,  $C_{\kappa-q,\kappa q}^{L0}$  are Clebsch-Gordan coefficients which appear because of double rotation of the coordinate system and  $h_L = h_L(\theta,\varphi)$  are unit spherical functions (Fig. 1).

In a linearly polarized field using the geometry of Fig. 1, we find that

$$\{e_{1}^{*(3)} \otimes e_{1}^{(3)}\}_{*q} = -\delta_{*,0}(1/3)^{1/2} + \delta_{*,2}\delta_{q,0}(2/3)^{1/2}$$

The scalar component of this tensor ( $\varkappa = 0$ ) corresponds to a redistribution of the population numbers  $\tilde{\rho}_{0,0}^{(n)}$  of the ground-state levels by the optical field:

$$\sum_{n} (2F_n+1)^{\frac{1}{2}} \tilde{\rho}_{0,0}^{(n)} = 0.$$





The quantity  $t_{0,0}^{(n)}$  in Eq. (10) is independent of the magnetic field and of the level number *n*, but in the phase space  $(\mathbf{v}_{\perp}, \mathbf{r})$  it governs the interaction time of atoms with the light beam. The time dependence of  $t_{0,0}^{(n)}$  is determined by modulation of the intensity of light. We shall determine the ensemble-average interaction time  $\langle \langle t_{0,0}^{(n)} \rangle_{v_1} = \bar{t}\tau_0$  for a constant intensity:

$$\tau_0 = \frac{1}{2} \pi I_0(\rho^2/2) \exp\{-\rho^2/2\}.$$
(12)

This time depends on two factors: the distribution of the intensity in the light beam and the equilibrium distribution of the velocities of the gas atoms. For example, for a Maxwellian distribution of the atoms and a Gaussian beam,  $f(r) = \exp(-\rho^2)$  and  $\rho = r/r_0$ , we have

$$\tau_{0} = \int_{0}^{\infty} \frac{dt}{\tilde{t}} \langle f(\mathbf{r} - \mathbf{v}_{\perp} t) \rangle_{\mathbf{v}_{\perp}}.$$
(13)

where  $I_0(x)$  is a modified Bessel function; we shall consider this particular case below. The characteristic features associated with atomic beams will be discussed separately. It should be noted that the asymptotic form of Eq. (13) in the range  $\rho \ge 1$  is independent of the beam profile:  $\tau_0 \propto 1/\rho$ .

The light-induced quadrupole moment proportional to  $\{e_1^{*(3)} \otimes e_1^{(3)}\}_{2,q} = \delta_{q,0} (\frac{2}{3})^{1/2}$  rotates, as is clear from Eq. (9), in a magnetic field, which means that  $\tilde{\rho}_{2q}^{(n)}$  has components with  $q \neq 0$  that depend on the magnetic field in Eq. (10). The interaction times  $t_{2,L}^{(n)}$  of Eq. (10) depend on the hyperfine level number n via the  $g_n$  factors of Eqs. (6) and (11) and also depend in a dual manner on the magnetic field: the index L is governed by the rank of the tensor constructed from a unit vector of the magnetic field and the intensity of the magnetic field gives rise to a strong anisotropy of the "interaction times" depending on q because of the oscillations in Eq. (10). By analogy with Eq. (12), we shall define  $\langle t_{2,L}^{(n)} \rangle_{v_1} = \tilde{t}\tau_{2,L}^{(n)}$ :

$$\tau_{2,L}^{(n)} = \sum_{l} (-1)^{q} C_{2-q,2q}^{L0} \tau_{q}^{(n)}.$$
(14)

The steady-state problem of rotation of the induced quadrupole moment of an atom in a magnetic field and of the polarization of light transmitted by a cell is solved in Ref. 8 for static fields  $\eta = p = 1$ . We shall now consider the processes governing the precession of the quadrupole moment.

We shall assume that the intensity of light remains constant: p = 1. We shall consider transient processes that are induced by the "switching on" of a constant magnetic field. The rate of this switching-on process is high compared with  $1/\bar{t}$ , so that  $\eta(t)$  is represented by the Heaviside step function. The vector components of the interaction times  $\tau_q^{(n)}$ then become

$$\tau_{q}^{(n)}(t) = \int_{0}^{t} \frac{dt'}{\bar{t}} \exp\{-iqg_{n}\Omega t'\} \langle f(\mathbf{r} - \mathbf{v}_{\perp}t') \rangle_{\mathbf{v}_{\perp}} + \exp\{-iqg_{n}\Omega t\} \int_{t}^{\infty} \frac{dt'}{\bar{t}} \langle f(\mathbf{r} - \mathbf{v}_{\perp}t') \rangle_{\mathbf{v}_{\perp}}.$$
(15)

In addition to the monotonically varying first component, which governs the steady-state solution obtained in Ref. 8, Eq. (15) includes also a correction oscillating at frequencies  $qq_n \Omega$  and describing free precession. It is important to note that the harmonics created by oscillations of the quadrupole moment (x = 2) contain  $q = \pm 1$ ,  $\pm 2$  of each of the Larmor frequencies  $\Omega_n = g_n \Omega$ . We shall determine the main precession frequencies by considering the example of the D lines of alkali metals ( $J_0 = 1/2$ ). The g factors are now described by

$$g_{n} = \begin{cases} -2\left(\frac{I(2I-1)}{3(2I+1)}\right)^{1/2}, & F_{n} = I - \frac{1}{2} \\ 2\left(\frac{(I+1)(2I+3)}{3(2I+1)}\right)^{1/2}, & F_{n} = I + \frac{1}{2} \end{cases}$$
(16)

Oscillations of the quadrupole as a result of transmission of linearly polarized light through, for example, sodium vapor (I = 3/2) are observed at four frequencies:

$$|q\Omega_{1}| = 8.78 \cdot 10^{6} H |q| c^{-1}, \quad F_{1} = 1,$$
  
$$|q\Omega_{2}| = 19.63 \cdot 10^{6} H |q| c^{-1}, \quad F_{2} = 2,$$
 (17)

where *H* is expressed in oersteds.

The precession relaxation process is described by the following time dependence of the amplitude:

$$A(t) = \int_{t}^{\infty} \frac{dt'}{\bar{t}} \langle f(r - v_{\perp}t') \rangle_{v_{\perp}}.$$
 (18)

In a typical situation corresponding to Eq. (13) the asymptotic behavior of the oscillation amplitudes at times  $t \ge \overline{t}$  is independent of r and is of universal form

 $A(t) = \bar{t}/t. \tag{19}$ 

In the opposite limiting case of  $t \ll \overline{t}$ , we have  $A(t) = \tau_0$ .

In the first part of this paper we have mentioned that it is not possible to model the finite size of a beam by the simple substitution  $\gamma_{x}^{0} \rightarrow 1/\bar{t}$ . In Eq. (18) such a substitution is equivalent to  $\langle f \rangle_{v_{1}} \rightarrow \exp\{-t/\bar{t}\}$ , and it is easy to show that in this case the behavior of  $A(t) = \exp\{-t/\bar{t}\}$  differs qualitatively from that described by Eq. (19). This qualitative difference is manifested largely in the form of forced precession resonances and examples of such resonances will be considered later.

## b) Modulation of the intensity of pump light

We shall now assume that the intensity of light varies with time in accordance with  $p = 1 + \alpha \cos \nu t$  ( $\nu \ll \gamma$ ) and the magnetic field is constant:  $\eta = 1$ . This corresponds to the excitation of forced precession of the quadrupole moment by modulation of the intensity of light. We shall write down the expression for the time dependences of the vector interaction times  $\tilde{\tau}_q^{(n)}(t)$  in the resonance case when  $|\Delta| \ll 1$ , where  $\Delta = (\nu - |q\Omega_n|)\overline{t}$ :

$$\tilde{\tau}_{q}^{(n)} = \frac{i}{2\alpha} \exp\{-i\beta vt\} \left(\tau_{cq}^{(n)} + i\beta \tau_{sq}^{(n)}\right), \qquad (20)$$

$$\tau_{cq}^{(n)} = \tau_0 - \frac{1}{2}\pi |\Delta|, \qquad (21)$$

$$\tau_{sq}^{(n)} = \Delta \ln |\Delta|, \qquad (22)$$

and  $\beta = \operatorname{sign}(q\Omega_n)$ .

The frequency dependences  $\tau_q^{(n)}(\Delta)$  of the precession amplitude of Eqs. (21) and (22) are independent of the beam profile and differ qualitatively from the Lorentzian and the dispersion profiles obtained using a model of exponential decay of the optical orientation. Figure 2 shows, for the sake of comparison, the dependences  $\tau_{cq}^{(n)}(\Delta)$  and Re  $L(\Delta)$ , and  $\tau_{sq}^{(n)}(\Delta)$  and Im  $L(\Delta)$ , where  $L(\Delta)$  (dashed curve) has the Lorentzian profile characterized by  $\gamma = 1/\overline{t}$ . The functions  $\tau_{cq}^{(n)}$  and  $\tau_{sq}^{(n)}$  have important singularities at zero: the derivative  $d\tau_{cq}^{(n)}/d\Delta$  has a discontinuity and  $d\tau_{sq}^{(n)}/d\Delta$  is infinite. Asymptotes of the functions  $\tau_{cq}^{(n)}(\Delta)$ and  $\tau_s^{(n)}(\Delta)$  depend on  $\rho$  if  $\Delta \ge 1$ , as deduced from Eqs. (10) and (14). If  $\rho = 0$ , they are described by

$$\tau_{cq}^{(n)} \rightarrow e^{-|\Delta|}, \quad \tau_{sq}^{(n)} \rightarrow 1/\Delta.$$
 (23)

## c) Modulation of the magnetic field amplitude (parametric resonance)

We shall carry out a similar analysis for the case of constant intensity of light (p = 1) and an alternating magnetic field  $\eta = 1 + \alpha \cos \nu t$ . In contrast to forced precession, which is due to modulation of the light intensity, when a resonance may be observed at one frequency  $\nu_1 = |q\Omega_n|$ , in the present case we find from an analysis of Eqs. (10), (11),





and (14) that the resonance oscillations of the quadrupole appear if we select the rf magnetic field frequency to be  $v_k = -q\Omega_n/k$  (k is an integer) (if  $q\Omega_n < 0$ , we have k > 0, whereas for  $q\Omega_n > 0$ , we have k < 0). Each frequency  $v_k$  of the magnetic field oscillations corresponds to a discrete series of frequencies of the oscillations of the quadrupole  $mv_k$ . In the case when  $|\alpha| \ll 1$ , which is of practical interest (when the amplitude of the rf field is small compared with the static field H), the term with m = 1 (first harmonic) in this series corresponds to the highest amplitude. We shall now give the expression for the component  $\tilde{\tau}_q^{(n)}(t)$  in this limiting case when  $|\Delta_h| \ll 1$  and  $\Delta_k = (v - v_k)\overline{t}$ :

$$\tilde{\tau}_{q}^{(n)}(t) = (-\alpha k)^{|\mathbf{k}|} \exp(ikvt) \left(\tau_{cqk}^{(n)} + i\beta\tau_{sqk}^{(n)}\right).$$
(24)

The spectral dependences  $\tau_{cqk}^{(n)}$  and  $\tau_{sqk}^{(n)}$  are then identical with Eqs. (21)–(23) if we make the substitution  $\Delta \rightarrow \Delta_k$ .

5. This precession of the quadrupole moment leads to modulation of the intensity, angle of rotation, and degree of ellipticity of a light beam transmitted by a cell. In particular, the susceptibility component  $\chi_{13}$  governing the rotation angle  $\psi$  and the ellipticity  $\alpha$  [Eq. (2)] can be expressed in terms of solutions of the kinetic equation for  $\tilde{\rho}_{2q}^{(n)}$ . The time dependence  $\tilde{\rho}_{2q}^{(n)}(t)$  for each type of precession is described by the behavior of the components of interaction times  $\tau_q^{(n)}(t)$  considered above. We shall write down the expression for  $\chi_{13}$  for an arbitrary relationship between the hyperfine splitting and the Doppler width for the atoms:

$$\chi_{13} = i \frac{|d|^2 N}{\hbar \gamma} \sum_{m,n,\varkappa} (-1)^{F_m + F_n} C_{mn} (1 + i\Delta_{mn})^{-1} \begin{cases} F_m & 1 & F_n \\ 2 & F_n & 1 \end{cases}$$
$$\times (\{e_1^{(1)} \otimes e_1^{(3)}\}_{\varkappa} \rho_{\varkappa}^{(n)}). \tag{25}$$

Here, N is the density of the atoms.

Before discussing the results of averaging over the velocities in Eq. (25), we should note that if we ignore the hyperfine splitting of an excited state (when the value of  $\Delta_{mn}$ is the same for all m), we can sum in Eq. (25) over m and the answer, irrespective of the value of  $\rho_2^{(n)}$  is proportional to a 6jsymbol:

$$\left\{\begin{array}{ccc}J_1 & J_0 & 1\\2 & 1 & J_0\end{array}\right\},\$$

which according to the selection rules does not vanish only if  $J_0 \ge 1$ . Hence, we can draw the important conclusion that rotation of the plane of polarization of light and, consequently, the ability to record in this way the precession of the quadrupole of the atoms with  $J_0 < 1$  and possible only if we allow for the hyperfine splitting (for example, in the case of alkali metals with  $J_0 = 1/2$ ).

We shall average over the velocities in Eq. (25) in the special case when the Doppler width is much greater than the characteristic magnitudes of the hyperfine splitting  $\Delta \omega_{+} = \max\{\omega_{nm_1}\}$  and  $\Delta \omega_{-} = \max\{\omega_{nn_1}\}$ . It is convenient to write down the result in terms of the orthonormalized quantity  $\xi_{13}$  [see Eq. (2)] which is independent of the gas density and the cell length:

$$\xi_{1s}(t) = (2J_0 + 1) \sum_{L,n} \frac{2\sqrt{6}}{(L(L+1))^{\frac{1}{2}}} C_{2120}^{L1} P_L^{(1)}(\theta)$$

$$\times \left\{ \frac{1 + (-1)^L}{2} \operatorname{Re} \langle \tau_{2L}^{(n)}(t) \rangle_v \cos \varphi + \frac{1 - (-1)^L}{2} \operatorname{Im} \langle \tau_{2L}^{(n)}(t) \rangle_v \sin \varphi \right\} B_n, \qquad (26)$$

where

$$B_{n} = \sum_{m_{1}n_{1}} (-1)^{F_{n}+F_{m}} C_{mn} \left\{ 1 - i \frac{\omega_{nn} - \omega_{mm_{1}}}{2\gamma} \right\}^{-1} \\ \times \left\{ \frac{F_{m}}{2} \frac{1}{F_{n}} \frac{F_{n}}{1} \right\} (\tilde{A}_{2}^{(1)\vec{n}n_{1}} - \tilde{A}_{2}^{(2)nn_{1}}).$$
(27)

Here  $\widetilde{A}_{2}^{(1,2)nn_{1}}$  is obtained from Eqs. (4) and (5) if  $\Delta_{mn_{1}} = 0$  in  $A_{2}^{(1,2)nn_{1}}$ .

We must stress once again that the polarization characteristics in Eq. (26) have a time dependence due to the precession of the quadrupole as defined in terms of  $\langle \tau_{2L}^{(n)}(t) \rangle_{v}$ .

We shall analyze the relationship between the angle of rotation  $\psi = -\chi l \operatorname{Re} \xi_{13}$  and the ellipticity angle  $\alpha = -\chi l \operatorname{Im} \xi_{13}$  for possible special cases. As a rule, the characteristic splitting of an upper state  $\Delta \omega_+$  is less than the characteristic splitting of a lower state  $\Delta \omega_-$ , so that we shall assume that  $\Delta \omega_+ < \Delta \omega_-$ . For atoms with the moment of the electron shell of the ground state  $J_0 \ge 1$  in Eq. (26) we find that for both  $\Delta \omega_- \ll \gamma$  and  $\Delta \omega_+ \gg \gamma$ , the angle of rotation  $\psi$  exceeds greatly the ellipticity angle  $\alpha$ . In the intermediate case when  $\Delta \omega_+ \propto \gamma$  the angles  $\psi$  and  $\alpha$  are of the same order of magnitude.

For atoms with  $J_0 < 1$  we have to allow for the hyperfine splitting, as pointed out above. If  $\Delta \omega_-$ ,  $\Delta \omega_+ \ll \gamma$ , the summing over *m* in the zeroth order with respect to  $\Delta \omega_-/\gamma$  gives  $\xi_{13} = 0$  in Eq. (27) and the first nonzero correction in respect of the parameter  $\Delta \omega_-/\gamma$  is purely imaginary. This means that in this case an elliptically polarized wave without rotation of the plane of polarization emerges from the cell. If  $\Delta \omega_+ \gg \gamma$  (when we know that  $\Delta \omega_- \gg \gamma$ ), the sums in Eq. (26) are dominated by the terms with  $\omega_{nn_1} = \omega_{mm_1} = 0$  and these determine the angle of rotation; we therefore have  $\psi \gg \alpha$ .

If the hyperfine splitting in the ground state exceeds the Doppler broadening, as is true for example of alkali metals, the resonant interaction of light occurs only with just one ground-state level with a fixed number  $n^*$ . We then have to substitute in Eqs. (26) and (27) the value  $n = n_1 = n^*$  ( $\omega_{nn_1} = 0$ ). A similar situation occurs if  $J_0 = 0$  ( $F_0 \ge 1$ ), i.e., when the ground state is not split and only the nuclear moment becomes oriented.

We shall conclude by noting that we have ignored a typical experimental setup<sup>4-7</sup> for the orientation by a circularly polarized field in order to draw attention to the novel physical effect of precession of the quadrupole moment. The problem of precession of multipole moments, oriented by an elliptically polarized field, will be considered in the future.

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