

THE OPTICAL EXCITATION OF MAGNETIC RESONANCE IN A ROTATING COORDINATE SYSTEM

L. N. NOVIKOV and V. G. POKAZAN'EV

Ural Polytechnical Institute

Submitted March 11, 1967

Zh. Eksp. Teor. Fiz. 53, 699-704 (August, 1967)

Theoretical and experimental studies are made of the resonances induced by modulated optical excitation in the system of magnetic sublevels of atoms in the ground state, as determined by the effective field in a rotating coordinate system, under the conditions of double radiooptical resonance. The resonances which arise when the modulation frequency coincides with the frequency of the Zeeman splitting in the effective field are detected by means of the absorption of a transverse light beam. The theoretical results are in good agreement with the experimental observations.

INTRODUCTION

WHEN a spin system is acted on simultaneously by a constant magnetic field  $H_0 \parallel Oz$  and a radiofrequency field  $H_1(t)$  lying in the plane perpendicular to  $H_0$  and rotating with the frequency  $\omega$ , in a rotating coordinate system the macroscopic magnetization can be represented as a constant equilibrium magnetization directed along the effective field  $H_e = [(H_0 - \omega/\gamma)^2 + H_1^2]^{1/2}$  and having the Larmor frequency  $\omega_e = \gamma H_e$ . In this coordinate system the averages over the spin ensemble of the values of the components of the magnetization transverse to  $H_e$  are equal to zero; or, using the terminology of quantum theory, we can say that there is no coherence in the system (in the density-matrix formalism this means that the nondiagonal components of the density matrix are zero). Coherence can be introduced in the rotating coordinate system in various ways,<sup>[1,2]</sup> which depend essentially on the use of a second radiofrequency field having the frequency  $\omega_e$ .

A coherent superposition of the states in the system of magnetic sublevels defined by the field  $H_e$  can also be produced by a different method, which is quite independent of the use of a second radiofrequency field. This method consists of the optical excitation of the spin system with resonance radiation whose intensity is modulated at a frequency  $\Omega$  which is equal to  $\omega_e$  or is a multiple of that frequency. The purpose of this paper is a theoretical and experimental investigation of the resonance in the effective field, as induced by modulated light in an optically oriented ensemble of atoms.

THEORY

The occurrence of resonance in the effective field under the action of a modulated optical excitation can be understood from the following qualitative treatment. Suppose the resonance condition  $\omega - \omega_0 = 0$  is satisfied in the laboratory coordinate system; then in the rotating system of coordinates  $H_e = H_1$ . If the intensity  $I_z$  of the light orienting the atoms of the specimen does not depend on the time, it is not hard to see that the average value of the magnetization perpendicular to the field  $H_1$  is equal to zero. If, on the other hand, the intensity  $I_z$

of the light varies with the frequency of the precession of the spins around  $H_1$ , then the magnetization produced by the light at the time  $t_0$  in the direction of the axis  $Oz$  will not be balanced by the magnetization produced at the time  $t_0 + \pi/\omega_e$ , and consequently in the rotating coordinate system there is a nonvanishing macroscopic magnetization precessing around  $H_1$ . From the quantum point of view the modulated optical excitation of the spin system leads to the appearance of a coherent superposition of the atomic states in the effective field.

The quantum theory of the cycle of optical orientation<sup>[3]</sup> leads to the following equations for the density matrix of the atoms in the ground state, interacting with the constant field  $H_0$ , the radiofrequency field  $H_1(t)$ , and the field of the optical resonance radiation:

$$\frac{d}{dt} \sigma_{mm'} = -i[\mathcal{H}, \sigma]_{mm'} - \frac{1}{T_p} (A_{mm} + A_{m'm'}) \sigma_{mm'} + \frac{1}{T_p} \sum_{nn'} B_{nn'}^{mm'} \sigma_{nn'} \quad (1)$$

where

$$\mathcal{H} = \gamma J_z H_0 + \gamma/2 H_1 (J_+ e^{-i\omega t} + J_- e^{i\omega t}) + \mathcal{H}_0 \quad (2)$$

$H_0$  is the Hamiltonian of the unperturbed atom,  $\gamma$  is the gyromagnetic ratio of the atoms in the ground state, and  $J_{\pm} = J_x \pm iJ_y$  are the transverse components of the angular momentum  $J$  of the atom.

The quantities  $A_{mm}$  and  $B_{nn'}^{mm'}$  can be expressed in terms of the matrix elements of the electric dipole transition:

$$A_{mm} = \sum_{\mu} \langle \mu | e^0 D | m \rangle \langle m | e^0 D | \mu \rangle,$$

$$B_{nn'}^{mm'} = \sum_{\mu\mu'} \langle \mu | e^0 D | n \rangle \langle n' | e^0 D | \mu' \rangle C(J_e, \mu; J, m) C(J_e, \mu'; J, m'),$$

where  $D$  is the operator for the electric dipole moment,  $e^0$  is the polarization vector of the incident light wave,  $J_e$  and  $\mu$  are the angular momentum and magnetic quantum number of the excited state, and  $C(J_e, \mu; J, m)$  are Clebsch-Gordan coefficients.

Equation (1) is written on the assumption that the Zeeman splitting of the ground and excited states is much smaller than the decay constant of the excited state. The expression (1) can be extended to the case of a modulated optical excitation. Let the intensity of the light which produces the pumping (propagated along the axis  $Oz$ ) have the following time dependence:

$$I = I_0(1 + \epsilon \cos \Omega t), \quad (3)$$

where  $\epsilon$  and  $\Omega$  are the index and frequency of the modulation. Since the intensity of the light occurs in (1) through the quantity  $1/T_r$ , which is given by

$$\frac{1}{T_r} = K = \int_0^\infty U(k) |A_k|^2 \frac{\Gamma/2}{(k - k_0)^2 + \Gamma^2/4} dk,$$

where  $U(k)$  is the spectral density of the incident light and  $\Gamma$  is the natural width of the excited state, in this case  $K$  must be a function of the time analogous to (3).

Equation (2) has been written without taking account of the effect of the various relaxation mechanisms on the character of the variation of the density matrix. The effects of thermal reaction can be included in the treatment by a generally accepted method, which is to add to the right member of (1) appropriate terms determined in a purely phenomenological way. Let  $T_1$  and  $T_2$  be certain generalized characteristic times for longitudinal and transverse relaxation. Assuming that  $T_1$  and  $T_2$  are much smaller than the time for optical relaxation, given by  $1/T_{mm'} = (A_{mm} + A_{m'm})/T_r$ , and that in the state of statistical equilibrium the populations of the sublevels are equal, we can rewrite Eq. (1) in the following form:

$$\frac{d}{dt} \sigma_{nn'} = -i[\mathcal{H}, \sigma]_{nn'} + \Lambda \delta_{nn'} - \Gamma_0 \sigma_{nn'} + K(t) \sum_{kk'} B_{kk'}^{nn'} \sigma_{kk'}, \quad (4)$$

where for simplicity we have assumed  $1/T_1 = 1/T_2 = \Gamma_0$ , and  $\Lambda$  is the rate of regeneration of the atomic states.

To solve Eq. (4) it is convenient to go over to a system of coordinates rotating with the frequency  $\omega$ . Then we have for the density matrix

$$\tilde{\sigma} = e^{i\omega J_z} \sigma e^{-i\omega J_z}, \quad (5)$$

and (4) can be written in the form

$$\frac{d}{dt} \tilde{\sigma}_{nn'} = -i[\mathcal{H}^T - \omega J_z, \tilde{\sigma}]_{nn'} + \Lambda \delta_{nn'} + \Gamma_0 \tilde{\sigma}_{nn'} + K(t) \sum_{kk'} B_{kk'}^{nn'} \tilde{\sigma}_{kk'}, \quad (6)$$

where

$$\mathcal{H}^T = \mathcal{H}_0 + \omega_0 J_z + \omega_l J_x, \quad \omega_0 = \gamma H_0, \quad \omega_l = \gamma H_l.$$

We shall look for the solution of (6) in the form of an expansion in terms of the optical perturbation:

$$\tilde{\sigma} = \tilde{\sigma}^{(0)} + \tilde{\sigma}^{(1)} + \dots \quad (7)$$

Confining ourselves to the first-order approximation, we get two equations:

$$\frac{d}{dt} \tilde{\sigma}_{nn'}^{(0)} = -i[\mathcal{H}^T - \omega J_z, \tilde{\sigma}^{(0)}]_{nn'} + \Lambda \delta_{nn'} - \Gamma_0 \tilde{\sigma}_{nn'}^{(0)}, \quad (8)$$

$$\frac{d}{dt} \tilde{\sigma}_{nn'}^{(1)} = -i[\mathcal{H}^T - \omega J_z, \tilde{\sigma}^{(1)}]_{nn'} - \Gamma_0 \tilde{\sigma}_{nn'}^{(1)} + K(t) \sum_{kk'} B_{kk'}^{nn'} \tilde{\sigma}_{kk'}^{(0)}. \quad (9)$$

The solution of (8) is

$$\tilde{\sigma}_{nn'}^{(0)} = \frac{\Lambda}{\Gamma_0} \delta_{nn'}. \quad (10)$$

Integrating (9) with the use of (10) and returning to the laboratory coordinate system, we get

$$\begin{aligned} \sigma_{nn'}^{(1)} = & K_0 \sum_{MM'} \sum_{mm'} \exp \{i\omega(n' - m' + m - n)t\} \langle n|M \rangle \langle M|m \rangle \langle m'|M' \rangle \\ & \times \langle M'|n' \rangle \frac{\Lambda}{\Gamma_0} B_{kk'}^{mm'} \delta_{kk'} \left\{ \frac{1}{\Gamma_0 - iX} + \frac{\epsilon}{2} \left( \frac{e^{i\Omega t}}{\Gamma_0 - i(X - \Omega)} \right. \right. \\ & \left. \left. + \frac{e^{-i\Omega t}}{\Gamma_0 - i(X + \Omega)} \right) \right\}, \quad (11) \end{aligned}$$

where

$$X = (M' - M)\omega_e + (m' - m)\omega, \quad \omega_c = \gamma H_e,$$

$M$  is the magnetic quantum number that characterizes the Zeeman splitting in the effective field  $H_e$ , and  $\langle n|M \rangle$  is the matrix for a rotation through the angle  $\vartheta$  around the axis  $Oy'$  in the rotating coordinate system, where  $\vartheta$  is defined by the expression  $\vartheta = \arcsin(\omega_l/\omega_e)$ , and is the angle between the directions of the constant field  $H_0$  and the effective field  $H_e$ .

It follows from (11) that the coherence  $\sigma_{nn'}^{(1)}$ , oscillates with frequencies which are multiples of  $\omega$ , and undergoes beats with the frequency  $\Omega$ , the amplitudes of the various harmonics having a resonance character (a resonance type of frequency dependence). When the condition

$$(M' - M)\omega_e + (m' - m)\omega \pm \Omega = 0 \quad (12)$$

is satisfied the modulated optical excitation leads to the appearance of a coherent superposition of the atomic states in the most general case. If  $m = m'$ , the relation (12) goes over into  $(M' - M)\omega_e \pm \Omega = 0$ , which is the condition for resonance in the system of magnetic sublevels in the effective magnetic field  $H_e$ .

If the resonance signal is registered by the change of the absorption of the resonance radiation by the atoms, the amount of light absorbed in unit time is described by the well known expression<sup>[3]</sup>

$$S = \frac{1}{T_r} \sum_{n'n} A_{nn'} \sigma_{n'n}, \quad (13)$$

where

$$A_{nn'} = \sum_{\mu} \langle n|e_{\alpha} D|\mu\rangle \langle \mu|e_{\alpha} D|n'\rangle.$$

The index  $\alpha$  on the polarization vector indicates that for the registration of the magnetic resonance one can use either the main (modulated) light beam (in this case  $e_{\alpha}^0 = e^0$ ) or an auxiliary beam making any angle with the field  $H_0$  and in general having a different polarization  $e_{\alpha}^0 \neq e^0$ . If the resonance is observed by means of the absorption of the main light beam, then substituting (11) in (13) we get

$$\begin{aligned} S = & \frac{\epsilon K_0}{2T_r} \sum_{MM'} \sum_{mm'} \sum_{nn'} \exp \{i\omega(n' - m' + m - n)t\} \langle n|M \rangle \langle M|m \rangle B_{kk'}^{mm'} \\ & \times \langle M'|M' \rangle \langle M'|n' \rangle A_{n'n} \left[ \frac{2}{\epsilon} \frac{1}{\Gamma_0 - iX} + \frac{e^{i\Omega t}}{\Gamma_0 - i(X - \Omega)} \right. \\ & \left. + \frac{e^{-i\Omega t}}{\Gamma_0 - i(X + \Omega)} \right]. \quad (14) \end{aligned}$$

This is a rather general expression, and in the special case in which there is no radiofrequency field it leads directly to the results obtained by Bell and Bloom<sup>[4]</sup> for the optical excitation of magnetic resonance in the laboratory coordinate system:

$$\begin{aligned} S = & \frac{\epsilon K_0}{2T_r} \sum_{mm'} B_{kk'}^{mm'} A_{m'm} \left[ \frac{e^{i\Omega t}}{\Gamma_0 + i[(m - m')\omega_0 + \Omega]} \right. \\ & \left. + \frac{e^{-i\Omega t}}{\Gamma_0 + i[(m - m')\omega_0 - \Omega]} + \frac{2}{\epsilon} \frac{1}{\Gamma_0 + i\omega_0(m - m')} \right]. \quad (15) \end{aligned}$$

Since in the case considered the quantity  $1/T_r$  appearing in (14) and (15) is a function of the time, of the form

of Eq. (3), in the absorbed light the harmonic of the signal at the frequency  $\Omega$  is proportional to  $\epsilon$ , the harmonic at frequency  $2\Omega$  is proportional to  $\epsilon^2$ , and the zero-frequency resonance term is proportional to  $\epsilon^2$ . It must be pointed out that resonance occurs only with coherent excitation, with  $m \neq m'$ . These are the results that were obtained in [4] on the basis of a phenomenological theory of optical orientation.

Let us now consider noncoherent excitation ( $m = m'$ ) by modulated light propagated along  $H_0$  and with circular polarization. The expression (14) is now rewritten in the form

$$S = -\frac{\epsilon K_0}{2T} \sum_{MM'n'n'} \sum_{mh} \exp\{i\omega(n' - n)t\} \langle n|M \rangle \langle M|m \rangle B_{hk}^{mm'} \langle m|M' \rangle \times \langle M'|n' \rangle A_{n'n} \left[ \frac{2}{\epsilon} \frac{1}{\Gamma_0 + i(M - M')\omega_e} + \frac{e^{i\Omega t}}{\Gamma_0 + i[(M - M')\omega_e + \Omega]} + \frac{e^{-i\Omega t}}{\Gamma_0 + i[(M - M')\omega_e - \Omega]} \right]. \quad (16)$$

In the special case of an atomic system with spin  $1/2$  and of registration of the signal by means of the absorption of a linearly polarized light beam propagated perpendicular to  $H_0$ , the general expression (16) is decidedly simplified and takes a form convenient for experimental testing:

$$S = c\epsilon \frac{\sin 2\alpha}{\Gamma_0^2 + (\omega_e - \Omega)^2} [\sin \varphi (\Gamma_0 \sin \omega t - \Delta\omega_e \cos \omega t) \sin \Omega t - 1/2 \sin 2\varphi \times (\Gamma_0 \cos \omega t + \Delta\omega_e \sin \omega t) \cos \Omega t], \quad (17)$$

where  $\alpha$  is the angle between the polarization vector of the light and the field  $H_0$ , and  $\Delta\omega_e = \omega_e - \Omega$ .

## EXPERIMENT

For the purpose of confirming the conclusions of the theory an experiment was made with optically oriented atoms of  $Cs^{133}$ . A spherical cell with walls coated with paraffin, containing saturated vapor of  $Cs^{133}$  at temperature  $25^\circ C$ , was at the center of a system of Helmholtz coils which produced a constant field  $H_0 \sim 0.28$  Oe and a radiofrequency field perpendicular to it with frequency  $\omega = 100$  kHz. The intensity of the

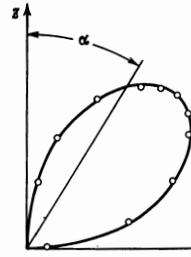


FIG. 2. Dependence of the size of the magnetic-resonance signal in the effective field on the angle  $\alpha$  between the polarization vector of the light and the field  $H_0$ . The value of the signal in relative units is plotted as radius vector.

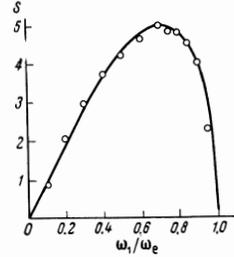


FIG. 3. Dependence of the size of the magnetic-resonance signal on the amplitude of the radiofrequency field  $H_1$ , for  $\omega_e = \text{const}$ . Values of the ratio  $\omega_1/\omega_e$  are plotted as abscissas, and values of the resonance signal in relative units or ordinates.

beam of circularly polarized light coming from an electrodeless cesium lamp and travelling along the field  $H_0$  was modulated at frequency  $\Omega$  by means of a low frequency generator. The frequency of the generator could be varied smoothly over a wide range. The registration of the magnetic-resonance signal was secured from the change of the modulation of the absorption of the auxiliary light beam, linearly polarized and directed perpendicular to  $H_0$ . The signal harmonic with frequency 100 kHz was amplified with a narrow-band amplifier, after which detection gave its envelope, with frequency  $\Omega$ . After the synchronous detection the amplitude of the envelope was recorded with a recording potentiometer while  $\Omega$  was varied in a smooth manner. The amplitude of the radiofrequency field  $H_1$  and the tuning difference  $\omega - \omega_0$  were chosen so that the frequency of the resonance in the effective field  $H_e$  was  $\omega_e/2\pi = 2.5$  kHz.

As shown in Fig. 1, a resonance was observed in the rotating coordinate system at a frequency of modulation of the light  $\Omega/2\pi = \omega_e/2\pi = 2.5$  kHz, in exact agreement with the prediction of the theory. According to (17) the magnitude of the resonance signal depends on the angle  $\alpha$  as  $\sin 2\alpha$ . In Fig. 2 this dependence is plotted in polar coordinates as the solid curve, and the values obtained experimentally are plotted as points. The dependence on the angle  $\varphi$  between the effective field  $H_e$  and  $H_0$  is given by the factor  $\sin 2\varphi$  for the term in (17) containing  $\cos \Omega t$ . Since  $\sin \varphi = \omega_1/\omega_e$  and  $\cos \varphi = \Delta\omega/\omega_e$ , for  $\omega_e = \text{const}$  the resonance signal  $S$  is proportional to  $(\omega_1/\omega_e)[1 - (\omega_1/\omega_e)^2]^{1/2}$ . This theoretical dependence is represented by the solid curve in Fig. 3, and the experimental values are plotted as points. It can be seen from Figs. 2 and 3 that the experiment fully confirms the conclusions of the theory we have given.

The resonance signal at the doubled frequency  $\omega_e$  was also observed. This directly confirms the resonance condition  $(M - M')\omega_e \pm \Omega = 0$ . In conclusion it must be noted that the width of the observed resonances was  $\sim 60$  Hz, which is due to absence of the radiofrequency broadening which usually accompanies radiofrequency transitions.

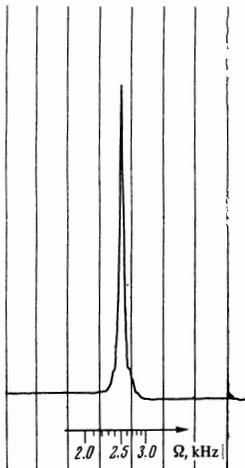


FIG. 1. Optically induced resonance of  $Cs^{133}$  in effective field at frequency  $\omega_e/2\pi = \Omega/2\pi = 2.5$  kHz.

- <sup>1</sup>A. Redfield, Phys. Rev. 98, 1787 (1955).  
<sup>2</sup>L. N. Novikov and V. G. Pokazan'ev, ZhETF Pis. Red. 4, 393 (1966) [JETP Lett. 4, 266 (1966)].  
<sup>3</sup>J. P. Barrat and C. Cohen-Tannoudji, J. phys. radium 22, 329, 443 (1961).

<sup>4</sup>W. E. Bell and A. L. Bloom, Phys. Rev. Letters 6, 280 (1961).

Translated by W. H. Furry  
78