COHERENCE TRANSFER IN RADIOOPTICAL RESONANCE AT THE NUTATION FREQUENCY

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We investigate theoretically and experimentally the influence of the transfer of the radio-frequency coherence produced in a system of magnetic sublevels determined by a field H_e in a rotating coordinate system, from the ground state of the atoms to the excited state. It is shown that magnetic resonance can be detected in the field H_e by the fluorescence light.

INTRODUCTION

IT is known that a radio-frequency field $H_1 \cos \omega t$ that varies in a plane perpendicular to a constant magnetic field $\mathbf{H}_0$ leads to the appearance of a component, transverse to this field, of the total magnetic moment of the system whenever the frequency of the Larmor precession $\omega_0 = \gamma H_0$ is of the order of ω. In the language of quantum theory this means that the alternating field introduces a radio-frequency coherence in the system of atomic magnetic sublevels. If the atoms interacting with the radio-frequency field are in the ground state, then optical excitation of the atoms by light of resonance frequency can cause the radio-frequency coherence to be transferred from the ground state to the excited state. This leads to a very complicated motion of the atoms in the excited state: besides the Larmor precession at the frequency of the excited state, the transverse component of the total angular momentum executes also forced motion at the natural frequencies of the ground state[1].

In a coordinate system that rotates with frequency ω around the direction of the constant magnetic field, the magnetic moments of the atoms precess around the direction of the effective field

$$H_e = [(H_0 - \omega/\gamma)^2 + H_1^2]^{1/2}$$

with a nutation frequency $\omega_e = H_e$, where γ is the magnetomechanical ratio of the atoms in the ground state. The mean values of the magnetization components transverse to $\mathbf{H}_e$ vanish in this coordinate system. It can be stated that there is no radio-frequency coherence in this coordinate system. In order to introduce coherence in the rotating coordinate system, it is necessary to apply a second radio-frequency field at the frequency ω_e. This raises the question of the possibility of transferring this coherence by exciting the atoms with light and observing the magnetic resonance in the effective field by means of the fluorescence light[2].

THEORY

To produce coherent precession of the magnetic moments in the effective field $\mathbf{H}_e$, we use a second radio frequency field $\mathbf{H}_2 \cos \omega t$, oscillating with frequency Ω in the direction of the constant magnetic field[3]. In the rotating coordinate system, the field $\mathbf{H}_2$ can be resolved into two components, one of which is parallel to the direction of $\mathbf{H}_e$, and the other is perpendicular to it

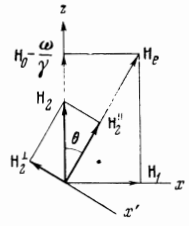

FIG. 1. Geometry of the magnetic fields acting on the magnetic moment of the atom in the rotating coordinate system.

(Fig. 1). The first induces parametric resonance in the system of magnetic sublevels defined by the field $\mathbf{H}_e$, and the other induces ordinary resonance, i.e., a resonance similar to that produced in the laboratory frame by the field H_1. We denote by θ the angle between the vectors $\mathbf{H}_0$ and $\mathbf{H}_e$, defined by the relation $\tan \theta = \gamma H_1/(\omega_0 - \omega)$. It is easy to see that parametric resonance is induced predominantly at small values of θ, and ordinary resonance when $\theta \to \pi/2$. In the case of intermediate values of the angle, the resonance signal is a mixture of both resonances[3].

For optical excitation of the atoms, and also to produce macroscopic magnetization in the ground state, we use circularly-polarized light propagating in the direction of $\mathbf{H}_0$. The influence of the optical excitation and radio-frequency fields $\mathbf{H}_1$ and $\mathbf{H}_2$ on the density matrix $\tilde{\sigma}$ describing the behavior of the atoms in the ground state can be determined from the following equation[4] (the tilde denotes the density matrix in the rotating coordinate frame)

$$\frac{d}{dt}\tilde{\sigma}_{\mu\mu'} = -i[\mathcal{H}, \tilde{\sigma}]_{\mu\mu'} + \Lambda\delta_{\mu\mu'} - \Gamma_0\tilde{\sigma}_{\mu\mu'} + \frac{1}{T_p}\sum_{\mu''\mu'''} B^{\mu\mu'}_{\mu''\mu'''}\tilde{\sigma}_{\mu''\mu'''}\exp\{i\omega(\mu - \mu' - \mu'' + \mu''')t\}, \quad (1)$$

where

$$\mathcal{H} = (\omega_0 - \omega)J_z + \omega_1 J_x + \omega_2 J_z \cos \Omega t;$$
$$\omega_1 = \gamma H_1, \quad \omega_2 = \gamma H_2,$$

μ is the magnetic quantum number which determines the Zeeman splitting of the atoms in the field H_0; Λ is the rate of regeneration of the atomic sublevels, Γ_0 the damping constant of the ground state, and J the angular momentum of the atom. For convenience, Eq. (1) is written in a coordinate system that rotates with a frequency ω. The last term in (1) depends on the polarization and on the intensity of the optical excitation, the quantity $1/T_p$ being proportional to the light intensity, while $B^{\mu\mu'}_{\mu''\mu'''}$ contains the angular dependence

$$B^{\mu\mu'}_{\mu''\mu'''} = \sum_{\substack{mm' \\ m-m'=\mu-\mu'}} \langle m|\mathbf{e}^0\mathbf{D}|\mu''\rangle \langle \mu'''|\mathbf{e}^0\mathbf{D}|m'\rangle C(J_f m;\ J\mu) C(J_f m';\ J\mu');$$

$C(J_f m;\ J\mu)$ is a Clebsch-Gordan coefficient, J_f and m are the angular momentum and the magnetic quantum number of the excited state, $\mathbf{e}^0$ is the unit vector of the polarization of the exciting light wave, and $\mathbf{D}$ is the dielectric dipole moment of the atom.

We seek the solution of (1) in the form of a series in the optical excitation

$$\tilde{\sigma} = \tilde{\sigma}^{(0)} + \tilde{\sigma}^{(1)} + \tilde{\sigma}^{(2)} + \ldots \tag{2}$$

Confining ourselves to the first order in the expansion, we obtain the following equations for $\tilde{\sigma}^{(0)}$ and $\tilde{\sigma}^{(1)}$:

$$\frac{d}{dt}\tilde{\sigma}^{(0)}_{\mu\mu'} = -i\,[\mathcal{H},\ \tilde{\sigma}^{(0)}]_{\mu\mu'} + \Lambda\delta_{\mu\mu'} - \Gamma_0\tilde{\sigma}^{(0)}_{\mu\mu'}, \tag{3}$$

$$\frac{d}{dt}\tilde{\sigma}^{(1)}_{\mu\mu'} = -i\,[\mathcal{H},\ \tilde{\sigma}^{(1)}]_{\mu\mu'} - \Gamma_0\tilde{\sigma}^{(1)}_{\mu\mu'}$$

$$+\frac{1}{T_p}\sum_{\mu''\mu'''} B^{\mu\mu'}_{\mu''\mu'''}\tilde{\sigma}^{(0)}_{\mu''\mu'''}\exp\{i\omega(\mu-\mu'-\mu''+\mu''')t\}. \tag{4}$$

The solution of (3) has the simple form

$$\tilde{\sigma}^{(0)}_{\mu\mu'} = \Lambda\delta_{\mu\mu'}/\Gamma_0. \tag{5}$$

Substituting (5) in (4) and introducing a new density matrix $\sigma^* = \tilde{\sigma}\exp(\Gamma_0 t)$, we obtain in operator form the following equation for σ^*:

$$d\sigma^{*(1)}/dt = -i\,[\Delta\omega J_z + \omega_1 J_x,\ \sigma^{*(1)}] - i\omega_2\cos\Omega t\,[J_x,\ \sigma^{*(1)}]$$
$$+\frac{\Lambda}{T_p\Gamma_0}\sum_{\mu''}\exp\{i\omega J_z t\}B_{\mu''\mu''}\exp\{-i\omega J_z t\}\exp\{\Gamma_0 t\},$$

$$\Delta\omega = \omega_0 - \omega. \tag{6}$$

We change over to a new coordinate system, the axis z′ of which coincides with the direction of the effective magnetic field. This transition is realized by rotating the coordinate system around the oy′ axis by an angle θ. In the new coordinate system, the density matrix σ^T is expressed in terms of σ^* by the formula

$$\sigma^T = \exp\{iJ_{y'}\theta\}\sigma^*\exp\{-iJ_{y'}\theta\}. \tag{7}$$

Substituting (7) in (6) and recognizing that $\omega_e = [\Delta\omega^2 + \omega_1^2]^{1/2}$, we obtain

$$d\sigma^{T_{(1)}}/dt = -i\omega_e[J_{z'},\ \sigma^{T_{(1)}}] - i\omega_2\cos\Omega t\,[J_{z'}\cos\theta - J_{x'}\sin\theta,\ \sigma^{T_{(1)}}]$$
$$+\frac{\Lambda}{T_p\Gamma_0}\sum_{\mu''}\exp\{iJ_{y'}\theta\}\exp\{i\omega J_z t\}B_{\mu''\mu''}\exp\{-i\omega J_z t\}\exp\{-iJ_{y'}\theta\}. \tag{8}$$

The first term of (8) describes the Larmor precession of the moments with frequency ω_e around the direction of the effective field. The second term represents the action of the radio-frequency field $H_2\cos\omega t$, and breaks up in turn into two components. The term $\omega_2\sin\theta\cos\Omega t[J_{x'}, \sigma^T_{(1)}]$ describes the interaction of the system with the H_2-component that oscillates in a plane perpendicular to the direction of the effective field and induces ordinary resonance. The second term describes the modulation of the effective field. As shown in [3], the H_2-component that oscillates in the direction of H_e induces parametric resonance at frequencies $\Omega = \omega_e/n$, where n = 1, 2, ... At small angles θ, as noted above, when the detuning $\Delta\omega$ is much larger than ω_1, practically only the parametric resonance is induced, and the influence of the field $H_2\sin\theta$ on the production of radio-frequency coherence in the system of the sublevels determined by the field H_e can be disregarded. Omitting the corresponding term from (8) and solving the remaining equation, we obtain

$$\sigma^{T_{(1)}} = \frac{\Lambda}{T_p\Gamma_0}\sum_{\mu''}\int_0^t T^*(t)U(t')B_{\mu''\mu''}U^*(t')T(t)\exp\{\Gamma_0 t'\}\,dt', \tag{9}$$

where

$$T(t) = \exp[iJ_{z'}(\omega_e t + (\omega_2/\Omega)\cos\theta\sin\Omega t)],$$

$$U(t) = T(t)\exp(iJ_{y'}\theta)\exp(i\omega J_z t),$$

T^* is the complex conjugate of T.

Applying to (9) the inverse transformation (7), and then going over from σ^* to $\tilde{\sigma}$, and also recognizing that the density matrix σ in the laboratory system is connected with $\tilde{\sigma}$ in the rotating system by the relation $\sigma = \exp(-i\omega J_z t)\tilde{\sigma}\cdot\exp(i\omega J_z t)$, we obtain for $\sigma^{(1)}_{\mu\mu'}$ the following expression:

$$\sigma^{(1)}_{\mu\mu'} = \frac{\Lambda}{T_p\Gamma_0}\sum_{\mu\mu'\mu''\eta\eta'}\sum_{MM'}\sum_{n,k=-\infty}^{\infty}(-1)^n\frac{J_n(z-z')J_{k-n}(z-z')\exp\{ik\Omega t\}}{\Gamma_0 + i[\omega_e(M-M') + \omega(\eta-\eta') + n\Omega]}$$
$$\times\langle\mu|M\rangle\langle M|\eta\rangle B^{\eta\eta'}_{\mu''\mu''}\langle\eta'|M'\rangle\langle M'|\mu'\rangle\exp[-i\omega(\mu-\mu'+\eta'-\eta)t], \tag{10}$$

where

$$z = \frac{\omega_2}{\Omega}M\cos\theta; \qquad z' = \frac{\omega_2}{\Omega}M'\cos\theta;$$

$\langle\mu|M\rangle$ is the matrix of rotation of the coordinate frame through an angle θ around the oy′ axis; M is the magnetic quantum number, which determines the Zeeman splitting of the atoms in the effective field; $J_n(z)$ is a Bessel function.

Expression (10) for the density matrix has a resonant dependence on the frequency Ω and assumes a maximum value when the condition $(M - M')\omega_e + (\eta - \eta')\omega + n\Omega = 0$ is satisfied. This resonance can be revealed in two ways: by absorption of the pump light, as already reported in [3], and by the fluorescence emitted by the atoms on going from the excited state to the ground state. The second method is possible if the coherence is transferred from the ground state to the excited state upon absorption of the light. Let us proceed to derive an expression for the parametric-resonance signal contained in the fluorescence light.

By definition [1], the intensity S_F of the spontaneous emission is

$$S_F = \frac{3\Gamma}{8\pi}\sum_{mm'}G_{mm'}\sigma_{m'm}, \tag{11}$$

where Γ is the damping constant of the excited state, and $G_{mm'}$ is the matrix for the emission of light with polarization characterized by a unit vector $\mathbf{e}$:

$$G_{mm'} = \sum_{\mu}\langle m|\mathbf{e}\mathbf{D}|\mu\rangle\langle\mu|\mathbf{e}\mathbf{D}|m'\rangle. \tag{12}$$

The density matrix $\sigma_{mm'}$ of the excited state is connected by a simple relation with the density matrix $\sigma_{\mu\mu'}$ of the ground state:

$$\sigma_{mm'} = \frac{1}{T_p\Gamma}\sum_{\mu\mu'}\langle m|\mathbf{e}^0\mathbf{D}|\mu\rangle\,\sigma_{\mu\mu'}\langle\mu'|\mathbf{e}^0\mathbf{D}|m'\rangle. \tag{13}$$

Substituting (13) in (11) with allowance for expression (10) for $\sigma_{\mu\mu'}$ (the zeroth approximation of $\sigma_{\mu\mu'}$ is of no interest), we obtain the following expression for the fluorescence-light intensity:

$$S_F = \frac{3\Lambda}{8\pi T_p^2\Gamma_0}\sum_{mm'MM'}\sum_{\mu\mu'\mu''\eta\eta'}\sum_{n,k=-\infty}^{\infty}\frac{(-1)^n J_n(z-z')J_{k-n}(z-z')\exp\{ik\Omega t\}}{\Gamma_0 + i[\omega_e(M-M') + \omega(\eta-\eta') + n\Omega]}$$
$$\times G_{m'm}\langle m|\mathbf{e}^0\mathbf{D}|\mu\rangle\langle\mu|M\rangle\langle M|\eta\rangle B^{\eta\eta'}_{\mu''\mu''}\langle\eta'|M'\rangle\langle M'|\mu'\rangle$$
$$\times\langle\mu'|\mathbf{e}^0\mathbf{D}|m'\rangle\exp\{-i\omega(\mu-\mu'-\eta+\eta')t\}. \tag{14}$$

Modulation at a frequency ω and at its multipoles is the consequence of the creation of radio-frequency co-

herence by the field H_1 in the system of sublevels μ of the ground state, and of its transfer to the excited state upon absorption of the pump light by the atoms. The transfer of this coherence was recently investigated experimentally in[5]. The appearance of modulation of the light re-radiated at the frequencies $k\Omega$ is the result of interference between the magnetic sublevels M of the ground state, but already determined by the field H_e. The amplitudes of the individual harmonics depend on the Bessel functions and have a resonant character. As follows from (14), in the case of a suitable choice of the frequency Ω and of the characteristics of the optical excitation, it is possible to detect quite independently, by means of the re-radiated light, the parametric resonance induced in the system of magnetic sublevels μ, representing the Zeeman splitting in the field H_0, and in the system of sublevels M, determined by the field H_e. Thus, in the case of incoherent optical excitation (the radiation is represented by either σ_+ or σ_- polarization) we have $\eta = \eta'$ and the resonance occurs if the following condition is satisfied

$$\omega_e(M - M') + n\Omega = 0, \tag{15}$$

i.e., the condition for parametric resonance of the atoms of the ground state in the rotating system of coordinates. The width of the resonance line is determined only by the times of the transverse thermal and optical relaxations of the atoms in the ground state. The radio-frequency broadening accompanying the usual resonance is missing.

Expression (14) is general. In the particular case of an atomic system having a spin J = 1/2, it simplifies greatly and assumes a form convenient for experimental verification[6]. Assuming k = n = 1 and $|\mu - \mu'| = |M - M'| = 1$, we have

$$S_F = \frac{\text{const}\cdot J_0(z)J_1(z)}{\Gamma_0^2 + (\omega_c - \Omega)^2}[\sin\theta(\Gamma_0 \sin\omega t - \Delta\omega_c \cos\omega t)\sin\Omega t - {}^1/_2 \sin 2\theta(\Gamma_0 \cos\omega t + \Delta\omega_c \sin\omega t)\cos\Omega t], \tag{16}$$

$$\Delta\omega_c = \omega_e - \Omega, \quad z = \omega_2 \cos\theta / \Omega.$$

On the basis of (14) and (16) we thus arrive at the following conclusions: a) the radio-frequency coherence produced in the ground state in the system of magnetic sublevels determined by the effective magnetic field H_e can be transferred during the process of optical pumping to the excited state, as a result of which the fluorescence light becomes modulated at the frequency $k\Omega$; b) owing to the coherence transfer, the resonance in the system of "effective" sublevels of the ground state can be detected by means of the fluorescence light.

EXPERIMENT

The experiment was performed with optically oriented Cs^{133} atoms. A cell of cylindrical form with walls coated with paraffin, containing saturated cesium vapor at a temperature 25° C, was located in the center of Helmholtz coils producing a constant field $H_0 \sim 0.28$ Oe. A radio-frequency field H_1 with frequency $\omega = 100$ kHz was applied perpendicular to the field H_0. The optical orientation of the atoms was by means of circularly polarized light from an electrodeless cesium lamp, propagating along H_0. A radio-frequency field H_2, oscillating in the direction of the constant magnetic field, was produced by a low-frequency generator. The fluorescence light was registered at right angle to the constant magnetic field. A narrow-band amplifier tuned to 100 kHz was used to register the harmonic of the light at the frequency ω. Detection separated the signal envelope of frequency Ω. The amplitude of the envelope was recorded, after synchronous detection, by an automatic potentiometer as the detuning $\Delta\omega$ was continuously varied. The detuning $\Delta\omega$ and the amplitude of the radio-frequency field H_2 were chosen in such a way that the frequency of the Zeeman splitting in the effective field was $\omega_e/2\pi = 2$ kHz, and the ratio of ω to ω_e was chosen such as to make $\sin\theta \sim 0.05$, in which case the parametric-resonance condition was satisfied.

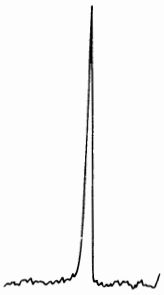

FIG. 2. Plot of parametric-resonance signal obtained from the fluorescence light under slow variation of $\Delta\omega$.

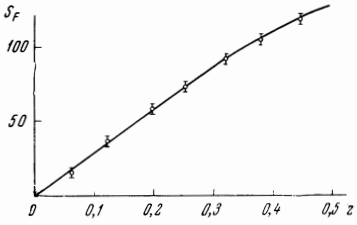

FIG. 3. Dependence of the resonance signal on the field H_2. The points denote the experimental results and the continuous curve the theoretical plot, in accordance with (16), proportional to $J_0(z)J_1(z)$.

Resonant intensification of the signal at the frequency $\Omega/2\pi = 2$ kHz was observed in the fluorescence light, in accord with the prediction of the theory. The recorded signal is shown in Fig. 2. The magnetic-resonance line width was of the order of 70 Hz, in satisfactory agreement with the width of the same resonance observed by means of absorption of the pump light[3].

We also investigated the dependence of the resonance signal on the field H_2. In accordance with (16), it is proportional to $J_0(z)J_1(z)$ at exact resonance. The curve of Fig. 3 shows the theoretical dependence of the signal on z, and the points show the experimental results. The agreement between the experimental and predicted results is good. No broadening of the resonance line was observed with increasing field H_2, a feature characteristic of the type of resonance under consideration.

[1] J. Barrat and C. Cohen-Tannoudji, J. phys. radium **22**, 329, 443 (1961).
[2] V. G. Pokazan'ev, Opt. Spektrosk. **24**, 348 (1968).
[3] L. N. Novikov, V. G. Pokazan'ev, and L. I. Yakub, Zh. Eksp. Teor. Fiz. **53**, 1287 (1967) [Sov. Phys.-JETP **26**, 752 (1968)].
[4] L. N. Novikov and V. G. Pokazan'ev, ibid. **53**, 699 (1967) [**26**, 438 (1968)].
[5] K. Rosinski, Acta Phys. Polonica **31**, 173 (1967).
[6] M. Bouchiat, J. Phys. **26**, 415 (1965).

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