MAGNETIC RESONANCE EFFECTS IN AN ALTERNATING EFFECTIVE FIELD

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Submitted May 19, 1967

Zh. Eksp. Teor. Fiz. 53, 1287–1292 (October, 1967)

The conditions for the excitation of parametric and ordinary resonance in an effective field $H_e = [(H_0 - \omega/\gamma)^2 + H_1^2]^{1/2}$ are investigated theoretically and experimentally for the case when the spin system is subjected simultaneously to an amplitude-modulated magnetic field $H_0 + H_2 \cos \Omega t$, parallel to the z axis, and a radio-frequency field $H_1(t)$, rotating about the z axis with a frequency ω. An expression for the M_z component of the macroscopic magnetization of the spin system is derived from the Bloch equations. The existence of parametric and ordinary resonances in an effective field is demonstrated for the case $\Omega = \gamma H_e/k$, where k = 1, 2, 3, ... The experimental results confirm the theoretical predictions.

INTRODUCTION

As demonstrated by Aleksandrov et al.,[1, 2] the modulation of a constant magnetic field H_0 at a frequency Ω, which is equal to or a multiple of the frequency representing transitions between the Zeeman sub-levels of the spin system in this field, $\omega_0 = \gamma H_0$, and a simultaneous coherent optical excitation (with radiation representing a mixture of π and $\sigma^{\pm}$ polarizations) produces a coherent superposition of states in an assembly of atoms. This effect, which can be deduced either from the modulation of fluorescence[1] or from the absorption of resonance radiation,[2] is known as parametric resonance. In contrast to ordinary magnetic resonance, the parametric resonance is characterized by the absence of radio-frequency broadening of the resonance line, because the radio-frequency field, parallel to the field H_0, does not cause real transitions between the magnetic sub-levels.

The conditions for the appearance of parametric and ordinary resonances are considered in the present paper for an effective field $H_e = [(H_0 - \omega/\gamma)^2 + H_1^2]^{1/2}$ when a constant field $\mathbf{H}_0 \parallel z$, rotating about the z axis at a frequency ω, a radio-frequency field $H_1(t)$ and a second radio-frequency field $H_2(t) = H_2 \cos \Omega t$, oscillating along a direction parallel to $\mathbf{H}_0$, are all applied simultaneously to the spin system. A similar situation has been considered earlier by Redfield,[3] who has investigated the saturation and spin temperature in a rotating system of coordinates but who did not complete his analysis.

In a system of coordinates x′y′z′, coupled to the direction of the effective field H_e and rotating about the z axis at a frequency ω, the field $\mathbf{H}_2(t)$ may be expanded into two components, one of which is parallel to $\mathbf{H}_e$. Redfield[3] has ignored the influence of the longitudinal component and has ascribed resonance in the effective field solely to the component of $\mathbf{H}_2$ perpendicular to $\mathbf{H}_e$. Such a treatment is justified only in the case $|\omega - \omega_0| \ll \gamma H_1$, but for arbitrary relationships between these quantities it is necessary to allow also for the influence of the component of $\mathbf{H}_2$ parallel to $\mathbf{H}_e$ since this component induces parametric resonance in the effective field. We shall show below that the contribution of the parametric resonance to the observed signal may, under certain conditions, be dominant.

THEORY

It is convenient to consider these phenomena using Bloch's equations[4] for the components of the macroscopic magnetization of the spin system. It is known[2, 5] that these equations can be used to describe, with satisfactory accuracy, magnetic resonance under optical orientation conditions. As a result of a double transformation of the system of coordinates (transformation to a system rotating about $\mathbf{H}_0 \parallel z$ at a frequency ω and subsequent rotation of the system by an angle θ about the y′ axis so that the new z′ axis coincides with the direction of $\mathbf{H}_e$), Bloch's equations are obtained in the following form:

$$\dot{m}_{\pm} = \pm i[\omega_e + \omega_0 \varepsilon \cos\theta \cos\Omega t]\, m_{\pm} \pm i\omega_0 \varepsilon \sin\theta \cos\Omega t\, m_{z'} - \frac{m_{\pm}}{T} - \frac{M_0 \sin\theta}{T},$$

$$\dot{m}_{z'} = -\frac{\omega_0 \varepsilon \sin\theta \cos\Omega t}{2i}(m_+ - m_-) - \frac{m_{z'}}{T} + \frac{M_0 \cos\theta}{T}, \qquad (1)$$

where M_0 is the equilibrium macroscopic magnetization in the absence of the radio-frequency field $H_1(t)$ and the modulating field $H_2(t)$; $m_{z'}$ and $m_{\pm} = m_{x'} \pm i m_{y'}$ are the components of the macroscopic magnetization in the new system of coordinates

$$\omega_e = \gamma H_e, \quad \varepsilon = \frac{H_2}{H_0}, \quad \omega_1 = \gamma H_1, \quad \tan\theta = \frac{\omega_1}{\Delta\omega}, \quad \Delta\omega = \omega - \omega_0,$$

and the longitudinal and transverse relaxation times are assumed to be equal: $T_1 = T_2 = T$.

The system of equations (1) is easily solved if $\varepsilon \omega_0 \Omega^{-1} \sin\theta < 1$. This condition is practically always satisfied in experiments. Then, the component m_+ becomes

$$m_+(t) = -\sum_{k,\, n=-\infty}^{\infty} \frac{J_k(Z) J_{k+n}(Z) e^{in\Omega t}}{T^{-1} - i(k\Omega + \omega_e)} \left(\frac{M_0 \sin\theta}{T} - ik\Omega \tan\theta\, m_{z'} \right), \qquad (2)$$

where $J_n(Z)$ is a Bessel function of the first kind and $Z = \omega_0 \varepsilon \Omega^{-1} \cos\theta$. Substituting Eq. (2) into the equation for $m_{z'}$ in Eq. (1) and averaging the latter with respect to time, we obtain a steady-state solution for $m_{z'}$ which has the following form in the effective field near the k-th resonance

$$m_{z'}^{(k)} = M_0 \cos\theta \frac{T^{-2} + (k\Omega + \omega_e)^2}{T^{-2} + (\Omega k)^2 \tan^2\theta\, J_k^2(Z) + (k\Omega + \omega_e)^2}. \tag{3}$$

Since $m_{x'} = \mathrm{Re}\,\{m_+\}$ and the component of the magnetization M_z in the laboratory system of coordinates is related to $m_{x'}$ and $m_{z'}$ by the expression

$$M_z = -m_{x'} \sin\theta + m_{z'} \cos\theta, \tag{4}$$

the final solution for M_z near the k-th resonance is obtained in the form

$$M_z^{(k)} = M_0 \cos^2\theta \frac{T^{-2} + (k\Omega + \omega_e)^2}{T^{-2} + (\Omega k)^2 \tan^2\theta\, J_k^2(Z) + (k\Omega + \omega_e)^2} + M_0 \sin^2\theta \sum_{n=-\infty}^{\infty} (A_n^{(k)} \cos n\Omega t + B_n^{(k)} \sin n\Omega t), \tag{5}$$

where

$$A_n^{(k)} = \frac{J_k(Z) J_{k+n}(Z)}{T^{-2} + (k\Omega + \omega_e)^2} \left[\frac{1}{T^2} + k\Omega (k\Omega + \omega_e) \frac{T^{-2} + (k\Omega + \omega_e)^2}{T^{-2} + (k\Omega)^2 \tan^2\theta\, J_k^2(Z) + (k\Omega + \omega_e)^2} \right], \tag{6}$$

$$B_n^{(k)} = \frac{J_k(Z) J_{k+n}(Z)}{T^{-2} + (k\Omega + \omega_e)^2} \left[k\Omega + \omega_e - k\Omega \frac{T^{-2} + (k\Omega + \omega_e)^2}{T^{-2} + (k\Omega)^2 \tan^2\theta\, J_k^2(Z) + (k\Omega + \omega_e)^2} \right]. \tag{7}$$

It follows from Eqs. (5)–(7) that the longitudinal component of the total magnetization of the spin system depends resonantly on the frequency Ω of the modulating field $\mathbf{H}_2$ if ω_e = const and $\Omega = \omega_e/|k|$, where $k = -1, -2, \ldots$. It should be mentioned that resonances in the effective field should take place at any value of the angle θ, i.e., at any value of the detuning $\Delta\omega = \omega - \omega_0$ and at any values of the amplitude of the radio-frequency field H_1. The term $(\Omega k)^2 \tan^2\theta\, J_k^2$ in Eq. (7) describes the broadening of the resonance line in the rotating system of coordinates, due to the influence of the component of $\mathbf{H}_2$ perpendicular to $\mathbf{H}_e$, which (as mentioned earlier) induces a magnetic resonance of the ordinary type. This broadening decreases considerably when $\theta \to 0$, i.e., when the dominant effect is the interaction of the spins with the component of $\mathbf{H}_2$ parallel to $\mathbf{H}_e$, which induces a parametric resonance in the effective field. When $\theta \to \pi/2$, only an ordinary resonance is induced in the rotating system of coordinates because the component of $\mathbf{H}_2$ parallel to $\mathbf{H}_e$ tends to zero. In the intermediate case ($0 < \theta < \pi/2$), the resonance signal is a mixture of the parametric and ordinary resonances, observed simultaneously. The relative contributions of the two resonances is determined by the value of the angle θ. Under optical orientation conditions, the intensity of the orienting radiation, parallel to $\mathbf{H}_0$, in an assembly of atoms depends on the state of the longitudinal component of the momentum of the spin system;[6] therefore, resonance in the effective field is observed either as a change in the constant component of the radiation or as a change in the amplitude of the modulation of light at frequencies $n\Omega$ close to $\Omega = \omega_e/|k|$.

This theoretical discussion is based on Bloch's equations in order to stress that the phenomena described here do not represent some special case which is typical only of optically oriented spin systems but that these effects represent general properties of magnetic resonance in an effective field and that they should occur at least in those systems which are described satisfactorily by Bloch's equations.

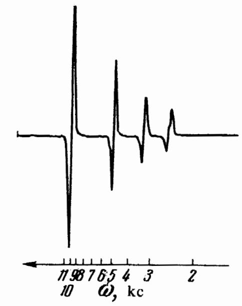

FIG. 1. Magnetic resonance of Cs^{133} atoms in an effective field at $\omega_e/2$ = 10 kc.

EXPERIMENTAL

In order to check the main conclusions of the theory, we carried out an experiment using a system of optically oriented Cs^{133} atoms. The apparatus used was in its main respects similar to that described earlier.[7] The amplitude of the radio-frequency field $H_1(t)$ and the detuning $\omega - \omega_0$ were selected so that $\omega_e/2\pi$ = 10 kc. The magnetic field $\mathbf{H}_2(t)$ was directed parallel to the field $\mathbf{H}_0$ and its frequency $\Omega/2\pi$ could be varied smoothly within the limits 2–15 kc.

We recorded the amplitude of the intensity of a longitudinal beam of the circularly polarized D_1 component of the resonance radiation modulated at a frequency Ω [this amplitude was proportional to the first harmonics of $M_z^{(k)}$ in Eq. (5)], and we also measured the signal proportional to the zeroth harmonic of $M_z^{(k)}$. In agreement with the theory, when ω_e = const, we observed a series of resonances in the effective field at the following frequencies: $\Omega = \omega_e = 2\pi \times 10$ kc, $\Omega = \omega_e/2 = 2\pi \times 5$ kc, $\Omega = \omega_e/3 = 2\pi \times 3.33$ kc, and $\Omega = \omega_e/4 = 2\pi \times 2.5$ kc (Fig. 1).

According to Eqs. (5) and (7), the dependence of the amplitude of the signal, proportional to $\sin \Omega t$, on the angle θ for the resonance $k = -1$ has the form

$$B_1^{(-1)} = M_0 \sin^2\theta \frac{\Omega T J_0(Z) J_1(Z)}{1 + (\Omega T)^2 \tan^2\theta\, J_1^2(Z)}. \tag{8}$$

When $\omega_2 \ll \Omega$, we can use the approximate representation of the Bessel functions $J_0 \approx 1 - Z^2/4$ and $J_1 \approx Z/2$, where $Z = \omega_2 \Omega^{-1} \cos\theta$. Substituting into Eq. (8) the values $\sin\theta = \omega_1/\omega_e$ and $\cos\theta = \Delta\omega/\omega_e$, we obtain

$$B_1^{(-1)} \propto \frac{\omega_2 T}{2} \left(\frac{\omega_1}{\omega_e}\right)^2 \sqrt{1 - \left(\frac{\omega_1}{\omega_e}\right)^2} \Big/ \left[1 + \left(\frac{\omega_2 T}{2}\right)^2 \left(\frac{\omega_1}{\omega_e}\right)^2\right]. \tag{9}$$

The dependence (9) for ω_e = const is represented in Fig. 2 by continuous curves and the experimental values

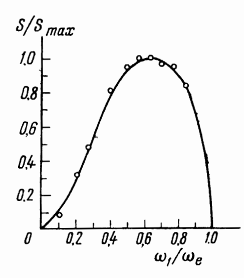

FIG. 2. Dependence of the resonance signal on the angle θ for ω_e = const. The abscissa gives the value of $\sin\theta = \omega_1/\omega_e$, while the ordinate gives the relative amplitude of the resonance signal

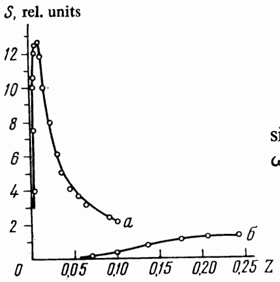

FIG. 3. Dependence of the resonance signal on $Z = \omega_2/\Omega\sqrt{2}$ for $\theta = 45°$; a) $\Omega = \omega_e$ resonance; b) $\Omega = \omega_e/2$ resonance.

are denoted by points. The agreement between the theoretical curve and the experimental data is satisfactory.

The greatest interest lies in the experimental proof of the existence of parametric resonance and of ordinary resonance. To provide such a proof, we measured the dependence of the resonance signals in an effective field on the amplitude of the field $H_2(t)$ for various values of the angle θ. Figure 3 shows the theoretical dependences, obtained from Eq. (7),

$$B_1^{(-1)} \propto \frac{\Omega T J_0(Z) J_1(Z)}{1+(\Omega T)^2 J_1^2(Z)}, \quad B_1^{(-2)} \propto \frac{2\Omega T J_1(Z) J_2(Z)}{1+(2\Omega T)^2 J_2^2(Z)} \qquad \text{b}$$

for $\theta = 45°$ and resonances $k = -1$ and $k = -2$, together with the experimental points. Figure 4 includes, in addition to the experimental results, the theoretical dependences of the resonance signal

$$S^{(k)} \propto \frac{(k\Omega T \operatorname{tg}\theta)^2 J_k^2(Z)}{[1+(k\Omega T \operatorname{tg}\theta)^2 J_k^2(Z)]^{3/2}} \qquad \text{c}$$

for $\theta = 0.5°$ and resonances $k = -1, -2, -3$, obtained from Eq. (5) making allowance for the low-frequency modulation and for the slow passage through the resonance line used in our experiments. In spite of the good agreement between the theoretical curves and the experimental results, one should still note the basic difference between these dependences.

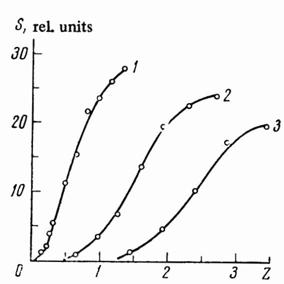

FIG. 4. Dependence of the resonance signal on $Z = \omega_2 \Omega^{-1} \cos\theta$ for $\theta \approx 0.5°$. 1) $\Omega = \omega_e$ resonance; 2) $\Omega = \omega_e/2$ resonance; 3) $\Omega = \omega_e/3$ resonance.

In the former case (Fig. 3), saturation of the first resonance takes place already at values of $Z < 0.01$, whereas in the latter case (Fig. 4), saturation is not reached in any of the investigated resonances even at the maximum available amplitudes of the field $H_2(t)$, corresponding to $Z > 1$. This difference indicates that, in the latter case, we are dealing mainly with parametric resonance in an effective field, i.e., with a resonance due to the component of $\mathbf{H}_2(t)$, parallel to the field $\mathbf{H}_e$, whereas in the former case, we have basically the conditions for an ordinary resonance and the component of $\mathbf{H}_2(t)$ perpendicular to the field $\mathbf{H}_e$ induces real transitions in a system of magnetic sub-levels governed by the effective field. Such transitions broaden the resonance lines considerably and saturate them rapidly.

These results confirm that, as mentioned in the introduction, it is necessary to allow for the influence of the longitudinal component of $\mathbf{H}_2$ in experiments of this type; the results allow us also to attribute side resonance lines[8] at $\Delta\omega \gg \gamma H_1$ to a parametric resonance in an effective field.

[1] E. B. Aleksandrov, O. V. Konstantinov, V. I. Perel', and V. A. Khodovoĭ, Zh. Eksp. Teor. Fiz. **45**, 503 (1963) [Sov. Phys.-JETP **18**, 346 (1964)].

[2] E. B. Aleksandrov, O. V. Konstantinov, V. I. Perel', Zh. Eksp. Teor. Fiz. **49**, 97 (1965) [Sov. Phys.-JETP **22**, 70 (1966)].

[3] A. G. Redfield, Phys. Rev., **98**, 1787 (1955).

[4] W. E. Bell and A. L. Bloom, Phys. Rev. **107**, 1559 (1957).

[5] C. Cohen-Tannoudji, Theses, l'Université de Paris, 1962.

[6] M. A. Bouchiat, J. Phys. **26**, 415 (1965).

[7] L. N. Novikov and V. G. Pokazan'ev, ZhETF Pis. Red. **4**, 393 (1966) [JETP Lett. **4**, 266 (1966)].

[8] K. Halbach, Phys. Rev. **119**, 1230 (1960).

Translated by A. Tybulewicz
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