Characteristics of a magnetic resonance in spin systems excited with a wide-band noise rf field

E. I. Dashevskaya and A. N. Kozlov

Institute of Terrestrial Magnetism, Ionosphere, and Radiowave Propagation
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A study was made of a magnetic resonance in a system of optically polarized ^{133}Cs atoms excited by a wide-band noise field of frequencies in the range 30–800 kHz. A study was made of the dependences of the resonance signal amplitude and of the resonance line profile on the noise field intensity. In contrast to monochromatic excitation, there was no distortion of the line profile in strong fields. The experimental results were compared with a theory based on the Bloch equation for a linearly polarized Gaussian random field $\mathbf{H}_1(t)$.

Our aim was to investigate experimentally the saturation of a magnetic resonance in a system of optically oriented ^{133}Cs atoms excited by a random magnetic field.

Nonlinear saturation effects associated with a reduction in the magnetization of a system by an exciting field have been investigated quite thoroughly both experimentally and theoretically in the case of monochromatic excitation. These effects are manifested by a reduction in the signal when the intensity of the exciting field is sufficiently high, by changes in the line profile until a "dip" forms at the center, as well as by line broadening and shift of the resonance frequency. In the case when the exciting field is nonmonochromatic this has, until recently, remained much less clear.

The first report of a study of a resonance in the presence of a nonmonochromatic field was that of Bonch-Bruevich *et al.*[1] who studied the profile of an absorption line of a weak monochromatic signal by a two-level system in the presence of strong "noise" fields of different spectral compositions. Bonch-Bruevich *et al.*[1] also discovered an absorption line shift proportional to the noise intensity, line broadening proportional to the square of the intensity, and reduction in the absorption at the maximum inversely proportional to the square of the intensity. The shift of the line absorption maximum was specifically investigated recently by Novikov,[2] who observed a parametric resonance in the system of ^{133}Cs atoms excited by a weak monochromatic field in the presence of noise. Contrary to the results of Ref. 1, Novikov did not observe a resonance line shift in wide-band noise. The first detailed theoretical investigation of the characteristics of a magnetic resonance in a random field was made by Knight and Kaiser.[3]

In contrast to Refs. 1 and 2, our experimental investigation of resonance properties of a system was not carried out using a monochromatic "probe" signal, but we observed directly the response to a strong wide-band noise signal. We used ^{133}Cs vapor polarized by optical pumping and characterized by a concentration of atoms of the order of 10^{10} cm^{-3}. Measurements were made in a terrestrial magnetic field $\mathbf{H}_0 = 51.6\,\mu T$. It is known that the system of magnetic sublevels of the ^{133}Cs atom represents a set of nine nonequidistant sublevels. Consequently, this spectrum consists of eight magnetic resonance lines satisfying the selection rules $\Delta F = 0$ and $\Delta m_F = \pm 1$. However, since the transition frequencies are close to one another, the shift of the centers of these lines are less than their widths: $\delta\omega < \Gamma$. This is why it is quite difficult to resolve the individual lines under normal experimental conditions and only a certain structured spectrum of a magnetic resonance with a frequency of 179 kHz and a total width of 100 Hz is observed. The transition responsible for this resonance is excited by a wide-band noise field.

Our source of rf noise was a generator of a pseudorandom sequence of pulses[4] with a 33-digit shift register, a modulo 2 logic scheme, and a timing pulse generator operating at 1.88 MHz. The repetition period of the pseudorandom sequence was about 7 min, which ensured the absence of a correlation coupling in the rf noise field during the actual time used to determine the spectrum. The upper frequency limit noise spectrum was 840 kHz at 3 dB. The strays from rf circuits at the input of a photodetector were minimized by setting the lower limit of the noise frequency at about 30 kHz. The maximum spectral density of the rf noise field was 3.1 nT·Hz$^{-1/2}$. Oscillations of a magnetic moment generated as a result of excitation of a resonance were manifested by noise modulation of an optical excitation beam, which was recorded by a photodetector. Such modulation had two components with very different frequencies. The high-frequency component was in the form of a narrow-band signal with frequencies close to the magnetic resonance frequency and a bandwidth of the order of the line width. The low-frequency component had a maximum in the spectral distribution at zero frequency (more exactly at the lowest recorded frequencies) and it decayed strongly at frequencies of the order of the resonance line width. We shall refer to these components as the high- and low-frequency signals, respectively ($I_\perp$ and I_z).

The spectral characteristics of the high-frequency signal were recorded by converting it to the frequency range 1–1.5 kHz and then applying the converted signal to an S4-54 spectrum analyzer with a $F36$ storage device. An optically pumped cesium vapor self-oscillatory magnetic field source[5] was used as a heterodyne for the conversion of the signal frequency. Systems of current-carrying rings were used to shift the oscillation frequency of this source relative to the

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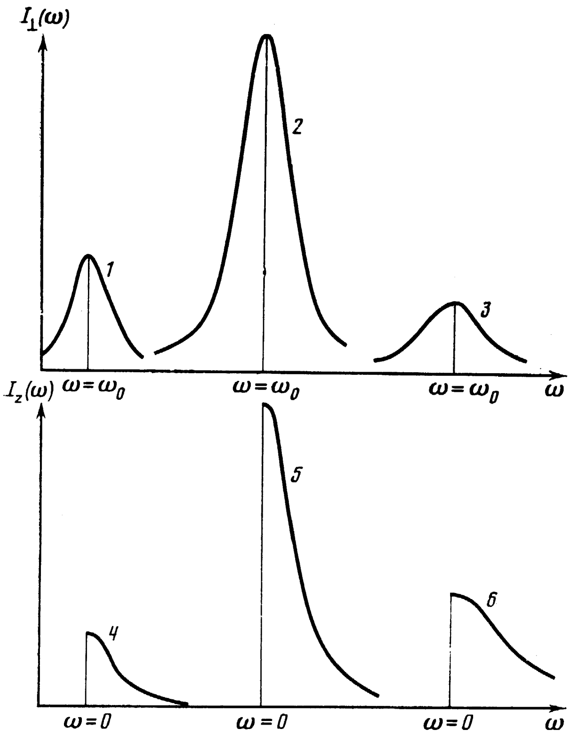

FIG. 1. Experimentally determined spectral distributions of the signal power obtained for different values of the dimensionless intensity of the exciting field. Curves 1–3 are the spectral distributions of the high-frequency signal: 1) $x^2 = 0.015$; 2) 0.10; 3) 0.41. Curves 4–6 gives the spectral distributions of the low-frequency signal: 4) $x^2 = 0.12$; 5) 0.30; 6) 0.065.

average frequency of the signal and thus generate the required frequency difference. This experimental setup made it possible to eliminate the influence of the magnetic field fluctuations. The low-frequency signal I_z was recorded directly by a spectrum analyzer connected to a storage device.

The experimental spectra of the signal power obtained for given optical pump intensity and various intensities of the exciting noise field are plotted in Fig. 1. Curves 1–3 describe the spectral distribution of the power of the high-frequency signal. The frequencies were measured from a resonance frequency $\omega_0 = \gamma \mathbf{H}_0$. The curves demonstrated the main features of the observed effect. The signal intensity first increased on increase in the excitation power and then fell ("saturation"). However, in contrast to the case of monochromatic rf excitation, no dip appeared in the central part of the line. The line intensity decreased without a basic change in its profile: there was only some broadening, but the line remained symmetric. Moreover, there was no shift of the line center, contrary to the shift well established in the case of monochromatic excitation. These differences were due to the fact that basically different physical quantities are recorded in the experiments involving the use of monochromatic and noise fields. In the case of a monochromatic field the curve is obtained by slow variation of the frequency of the exciting signal. At each frequency there is a specific value of the difference between the level populations between which a transition takes place. In the wings of a line these populations are the same as in a weak field, whereas at the center they become equalized in a sufficiently strong field, which gives rise to a dip that distorts the line profile. However, in the case of noise excitation the whole absorption curve is recorded simultaneously and all its parts correspond to the same population difference, which decreases as a result of saturation causing a uniform reduction in the intensity of the whole line.

Moreover, the line broadening is physically different in these two cases. In a monochromatic field it is manifested by a distortion of the line profile: the absorption at the center decreases and the wings become stronger. In the case of noise excitation the observed broadening is due to acceleration of the relaxation of the magnetic moment under the influence of the random field.

Curves 4–6 in Fig. 1 show the spectral distribution of the low-frequency signal. The width of the curve increases on increase in the intensity of the exciting noise field.

We shall now compare our experimental results with the theory of Ref. 3, where an analysis is made of a system of magnetic moments described by the Bloch equations with longitudinal and transverse relaxation times T_1 and T_2, subjected to a linearly polarized Gaussian random field $H_1(t)$ with a δ-like correlation function:

$$\gamma^2\langle H_1(t) H_1(t')\rangle = \sigma^2 \delta(t-t'). \tag{1}$$

Here, γ is the gyromagnetic ratio. For the Cs atom, we have $\gamma = 3.5$ Hz/nT. Knight and Kaiser were able to carry out rigorous averaging of the equations over the field fluctuations and, in particular, they were able to calculate the correlation functions of the magnetic field components.

As is known, the Bloch equations are sufficiently rigorous for the description of a magnetic resonance in a two-level quantum-mechanical system.[1)] However, it is reasonable to apply the theory developed in Ref. 3 to describe the experimental results obtained for ^{133}Cs atoms. An additional argument for the use of the noise variant of the Bloch equations in the description of the system of levels of the ^{133}Cs atom is the fact that the classical variant of the Bloch equations describes the system quite satisfactorily in the case of excitation by a monochromatic field $H_1(t)$.

It may also be assumed that the hypothesis of the δ-like correlation of the noise applies sufficiently well to our wideband rf field $H_1(t)$. The mean-square value of the field H_1^2 is linked to a parameter σ^2, described by Eq. (1), by the relationship $\gamma^2 H_1^2/\Delta f_n = \sigma^2$ (Δf_n is the noise field band, which in our case amounts to $\Delta f_n = 800$ kHz).

The investigated signals are created by oscillations of the magnetic moment excited by the applied *rf* noise field. The oscillations of the transverse components of the magnetic moments M_x and M_y are of frequency close to $\omega_0 = \gamma H_0$, and they create the high-frequency signal. The low-frequency signal can naturally be attributed to oscillations of the longitudinal component of the magnetic moment M_z. If these assumptions are correct, then the spectral distribution of the power of the high-frequency signal should be governed by the spectral density of oscillations of the transverse component of the magnetic moment, i.e., it should be governed by the Fourier transform of the time correlation function

$$I_\perp(t) = \langle M_x(t) M_x(0) + M_y(t) M_y(0)\rangle,$$

whereas the spectral distribution of the low-frequency signal should be governed by the Fourier transform of the correlation function $I_z = \langle M_z(t) M_z(0) \rangle$ The required correlation functions are calculated in Ref. 3 and are given by Eqs. (50) and (61) in that paper. Fourier transformation yields the following expressions for the spectral densities:

$$I_\perp(\omega) = 2M_0T_2x^2(1+x^2) \times \{(1+2lx^2)[1+(2l+1)x^2][(1+x^2)^2+\varepsilon^2]\}^{-1} \quad (2)$$

and

$$I_z(\omega) = 4M_0T_1lx^4 \times \{(1+2lx^2)[1+(2l+1)x^2][(1+2lx^2)^2+\rho^2]\}^{-1}. \quad (3)$$

The notation used in these expressions is as follows:

$$x^2=\sigma^2T_2/4, \quad l=T_1/T_2, \quad \varepsilon=(\omega-\omega_0)T_2, \quad \rho=\omega T_1,$$

and M_0 is the static magnetic moment. Equation (2) is simplified by omitting the resonance frequency shift induced by the noise field:

$$\delta\omega = -T_2^{-1}(x^4/2\gamma H_0T_2).$$

This shift is proportional to the fourth power of the noise amplitude and under our conditions it is negligible (much less than the line width), which accounts for the experimentally observed absence of the line center shift. We recall that when a resonance is excited by a linearly polarized monochromatic field there is a considerable shift of the resonance frequency proportional to the square of the field amplitude (Bloch-Siegert shift).

Equation (2) accounts for another qualitative feature of our experimental results, which is the absence of distortion of the line profile at saturation. According to Eq. (2) the line remains Lorentzian for any intensity of the noise field. The line width of the high-frequency signal at midintensity is then $\Gamma = (1/\pi T_2)(1+x^2)$. It is clear from Eq. (2) that deviation from the linear regime is governed by the parameter x. The saturation effects appear for $x^2 = \gamma^2 H_1^2 T/\Delta f_n \sim 1$. (In these estimates we make no distinction between the longitudinal and transverse relaxation times: $T_1 = T_2 = T$.) It is known that in the case of a monochromatic field H_1 the saturation appears for $\gamma^2 H_1^2 T^2 \sim 1$. This means that the saturation effects are observed in the case of noise excitation when the noise intensity in the absorption band of the spin system $H_1^2/\Delta f_n T$ is of the order of the intensity of the field which causes saturation in the monochromatic case.

In a quantitative comparison of the experiment with theory it is necessary to know the values of the relaxation times T_1 and T_2. These relaxation times are not easy to find because under realistic experimental conditions the process of relaxation is due to collisions of atoms with holes and the effective relaxation times depend on the rate of optical pumping, which is naturally not described by the Bloch equations. We shall use the following relaxation times which are typical cesium absorption cells coated with paraffin:

$$T_1=25\cdot 10^{-3}\text{ sec}, \quad T_2=7\cdot 10^{-3}\text{ sec}, \quad l=3.57, \quad (4)$$

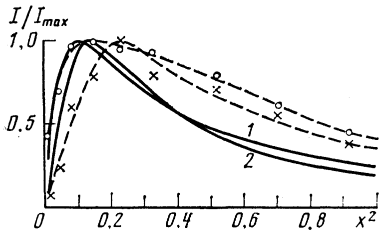

FIG. 2. Dependence of the spectral density at the line center for $l = 3.57$ on the dimensionless intensity of the exciting field x^2. Curve 1 is the theoretical dependence for the high-frequency signal and the points (circles) are the experimental values. Curve 2 is the theoretical dependence for the low-frequency signal and the crosses are the experimental values.

obtained earlier in monochromatic field experiments.[5]

Curve 1 in Fig. 2 shows the dependence of the spectral density $I_\perp(\varepsilon)$ at the line center [plotted using Eq. (2) and assuming that $l = 3.57$, i.e., assuming that $\varepsilon = 0$] on the dimensionless intensity of the exciting field x^2. The ordinate is plotted in units of $I_\perp = I_{\max}$ at the maximum of the curve. The points represent the experimental values of the intensity of the high-frequency signal normalized by the same method, but the values of x^2 are calculated using the above relaxation time T_2. It is clear from this figure that the agreement between the theory and experiment is satisfactory, especially if we bear in mind that no fitting parameters have been used in this comparison.

Curve 2 in Fig. 2 shows the spectral density $I_z(\varepsilon)$ for $\varepsilon = 0$ calculated from Eq. (3). The curve is also normalized to the value of I_z at the maximum. The experimental data are represented by crosses. Once again the agreement between the theory and experiment is satisfactory.

Figure 3 gives the dependences of the line width for the high-frequency (curve 1) and low-frequency (curve 2) signals of the intensity of the exciting field. The abscissa is x^2 and the ordinate is the line half-width referred to the half-width of the signal with the minimum amplitude. The increase in the line width under the influence of the noise field can be regarded as proportional to the intensity of this field, as expected on the basis of the theory. Some discrepancy between the theory and the experiment may be due to the fact that the Bloch equations with phenomenologically in-

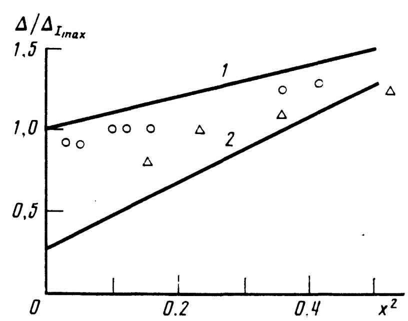

FIG. 3. Dependences of the half-widths of the lines of the observed signal on the intensity of the exciting field: 1) $I_\perp$; 2) I_z (theory); O),△) experimental results for I and I_z, respectively. The ordinate gives the half-width in units of $1/T_2$.

troduced relaxation do not generally describe well the profile of a magnetic resonance observed under optical pumping conditions. [Naturally, the agreement with the experimental results can be improved if we regard T_2 as a fitting parameter and do not use the value given by Eq. (4).] The Bloch equations predict in particular a Lorentzian profile, whereas the observed signal profile is closer to Gaussian.

There is another feature observed in our experiments. Excitation of a resonance by a wide-band noise field cannot be used to observe a signal corresponding to transitions between the sublevels with $F = 3$ In the case of monochromatic excitation such a signal is readily observed, although it is of lower intensity than that associated with transitions in the hyperfine state $F = 4$. This cannot be explained by the theory which utilizes just one Bloch equation.

[1)] The same authors gave in Ref. 6 only the general method of calculation of the influence of Gaussian noise on a multilevel system, but no specific applications are considered.

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