Electron-nuclear magnetic resonance in the inverted state

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An analysis is made of the susceptibility of the electron-nuclear system of a ferromagnet in the case when the nuclear magnetization is inverted relative to the hyperfine field direction. A weak radiofrequency signal can be amplified subject to an additional inequality relating the interaction frequency to the electron and nuclear relaxation parameters; the gain may be greater than in the case of an inverted nuclear system in a magnetically ordered material. In the region of strong interaction between ferromagnetic and nuclear resonances the double resonance spectrum has a fine structure which is the reverse of that obtained earlier by the present authors (1975) for the double resonance spectrum in the normal state. The initial transient process which appears in an electron-nuclear system under the action of a high-frequency field is briefly considered.

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INTRODUCTION

In our earlier paper [1] we studied the susceptibility of an electron-nuclear magnetic system of a ferromagnet in the region of strong interaction between ferromagnetic (FMR) and nuclear (NMR) resonances and we demonstrated that the behavior of the susceptibility maxima does not agree even qualitatively with the behavior of the eigenfrequencies of the system. The spectrum of such an electron-nuclear magnetic resonance (ENMR) has a fine structure: a narrow "reverse resonance" peak should be observed against a wide FMR maximum. The fine structure of the ENMR spectrum was observed experimentally in [2] and this has made it possible to study NMR in thin magnetic films by a simple continuous method.

We shall consider the susceptibility of an electron-nuclear system in an inverted state. The term "inverted state" is understood to be a situation in which the nuclear magnetization is rotated through an angle π relative to its equilibrium position and the electron magnetization is in its equilibrium (relative to the external magnetic field) position. The possibility of producing an inverted state in a thin magnetic film outside the range of strong interaction between NMR and FMR was demonstrated experimentally by Salanskiĭ, Lyapunov, and Mal'tsev [3] by inverting the nuclear magnetization with a radiofrequency pulse and by pulse switching of the electron magnetization (the latter effect was predicted by Ignatchenko and Kudenko [4]). To our knowledge, no attempts have yet been made to establish an inverted state of an electron-nuclear magnetic system in a ferromagnet in the range of strong interaction between NMR and FMR.

The purpose of the present paper is to draw the attention of experimentalists to certain features of the ENMR spectrum of an inverted state. We shall consider, as in [1], the example of a thin plate which is magnetized in its own plane. We recall that the shape of the sample has a considerable influence on the electron-nuclear interaction in a ferromagnet [5] because the magnitude of this interaction depends very strongly on the magnetic asymmetry in a plane perpendicular to static magnetic field. The asymmetry can be produced by the demagnetizing field resulting from the shape of the sample or by the magnetic anisotropy field; since the shape of the demagnetizing field for a plate is much stronger than the anisotropy field in ferromagnets, the dynamic electron-nuclear magnetic interaction should be strongest in a plate. Moreover, the strongest interaction between FMR and NMR corresponds to physically attainable frequencies and fields only for certain shapes of samples. [5]

The eigenfrequencies of coupled homogeneous electron-nuclear oscillations in an inverted state have been considered earlier. [6] The nuclear relaxation Γ_n has been ignored compared with the electron relaxation Γ_e because $\Gamma_n \ll \Gamma_e$. However, the nuclear relaxation is as important as the electron relaxation in the description of the ENMR line profile (this applies also to ENMR in the normal state [1]), so that in the first section we shall consider briefly the eigenfrequencies of the system allowing for Γ_e and Γ_n.

In the second section we shall analyze in detail the ENMR line profile in an inverted state for the case of such a weak external high-frequency field that the relaxation regime can be regarded as quasisteady; we shall consider the possibility of amplification of a high-frequency field by an inverted electron-nuclear system. In the third section we shall analyze briefly the initial (linear) stages of a stimulated transient process which appears in an electron-nuclear system under the action of a strong high-frequency field.

1. EIGENFREQUENCIES OF THE SYSTEM

Inversion of the nuclear magnetization μ or the electron magnetization $\mathbf{M}$ alters the sign in front of the square of the frequency of interaction between the electron and nuclear subsystems, because the square of this frequency is proportional to the product $M_z\mu_z$:

$$\omega_q^2=-\gamma_e^2 4\pi A M_z \mu_z \qquad (1.1)$$

(all the notation used here is the same as in our previous paper [1]).

In an equilibrium state the magnetizations M_z and μ_z have opposite signs and $\omega_q^2 > 0$, whereas in an inverted state $\mathbf{M}$ and μ are directed parallel to one another and $\omega_q^2 < 0$. Therefore, the eigenfrequencies of the system in the normal and inverted states can be found by equating to zero the denominator of Eq. (4) in [1]:

$$(\omega_e^2+2i\Gamma_e\omega-\omega^2)(\omega_n^2+2i\Gamma_n\omega-\omega^2)-\omega_n^2\omega_q^2=0. \qquad (1.2)$$

There are two complex frequencies in the solution of this equation. The changes in the position of these frequencies due to inversion of the system are particularly striking if we ignore the damping: $\Gamma_e = \Gamma_n = 0$. Then,

the real parts of the frequencies are pushed apart in a normal state [5] and merge over the whole frequency interval in question in an inverted state; [6] the imaginary parts vanish in a normal state and differ from zero in the same frequency interval in an inverted state. [6] However, in the case of real ferromagnets the value of Γ_e is large, being of the order of the NMR frequency ω_n, and graphs of the solutions of Eq. (1.2) are quite different. In this case one oscillation mode remains also near the point of the maximum interaction of electron-like modes; the corresponding frequency will be denoted by $\omega_e' + i\omega_e''$. A different oscillation mode is nuclear-like; we shall denote its frequency by $\omega_n' + i\omega_n''$.

Far from the strong interaction region the first frequency becomes $(\omega_e^2 - \Gamma_e^2)^{1/2} + i\Gamma_e$, and the second reduces to $\omega_n + i\Gamma_n$ in the normal and inverted states. In the strong interaction region the real parts of the eigenfrequencies now intersect both in the normal [7] and inverted [6] states. The imaginary parts of the eigenfrequencies at the point of coincidence of the NMR and FMR, $\omega_e = \omega_n$, are given by the expression

$$\omega_e'' \approx \Gamma_e - \omega_q^2/4\Gamma_e, \quad \omega_n'' \approx \Gamma_n + \omega_q^2/4\Gamma_e. \tag{1.3}$$

Since Γ_e is of the order of ω_n, the electron-like oscillations are damped out very rapidly—in a time of the order of one oscillation period. The reduction in Γ_e by an amount $|\omega_q^2/4\Gamma_e|$ in the normal state and an increase in the inverted state are negligible compared with Γ_e. The situation is different for the nuclear-like oscillations: in this case we have $\omega_n'' \ll \omega_n'$; Γ_n and $|\omega_q^2/4\Gamma_e|$ can be of the same order of magnitude. Therefore, a change in the sign of ω_q^2 as a result of inversion may alter considerably ω_n'' including a change in its sign if $|\omega_q^2| > 4\Gamma_e\Gamma_n$; then, the damping is replaced by the growth of nuclear-like oscillations.

2. CHANGE IN THE ENERGY OF A WEAK RADIOFREQUENCY FIELD BY AN INVERTED ELECTRON-NUCLEAR SYSTEM

Real relationships between the parameters of the electron and nuclear magnetic subsystems in ferromagnets are such that the nonlinear effects in the motion of μ may correspond to solutions which are still linear in $M_{x,y}$. This situation is described by the following system of equations of motion

$$\begin{aligned}
&\dot M_x + \omega_2 M_y + \xi \dot M_y + \gamma_e A M \mu_y = 0,\\
&\dot M_y - \omega_1 M_x - \xi \dot M_x - \gamma_e A M \mu_x = -\gamma_e M h_x,\\
&\dot\mu_x + \omega_n \mu_y + \Gamma_n \mu_x - \gamma_n A \mu_z M_y = 0,\\
&\dot\mu_y - \omega_n \mu_x + \Gamma_n \mu_y + \gamma_n A \mu_z M_x = +\gamma_n \mu_z h_x,\\
&\dot\mu_z - \gamma_n A (M_x \mu_y - M_y \mu_x) + (\mu_z + \mu)/T_1 = 0,
\end{aligned} \tag{2.1}$$

where $\omega_1 = \gamma_e(H - H_c)$ and $\omega_2 = \gamma_e 4\pi M$.

If initially the nuclear magnetization goes over to an inverted state, $\mu_z = \mu$, it returns subsequently to a normal state $\mu_z = -\mu$, i.e., the z projection changes. The terms coupling M_i to μ_z in the third and fourth equations of the system (2.1) make the problem nonlinear.

Linearization of the system (2.1) for a varying z projection of μ is possible if this variation is sufficiently slow compared with other characteristic times of the system. To satisfy this requirement we shall consider a case in which the transverse nuclear relaxation frequency ω_n'' is positive (transverse oscillations decay) and satisfies $(\omega_n'')^{-1} \ll T_1$. We shall assume that in the course of a transient process the second term in the last equation of the system (2.1) is much smaller than the third term (the necessary inequality will be given later). We then obtain the "quasisteady" relaxation regime in which the variation of μ_z is governed only by the time constant T_1:

$$\mu_z = \mu[2\exp(-t/T_1) - 1]. \tag{2.2}$$

If all the other characteristic times are much shorter than T_1, the first four equations of the system (2.1) can be solved as a linear system on the assumption that μ_z = const and the variation of μ_z can be allowed for in the final result. Then, the general solution for M_x is given by the expression

$$\begin{aligned}
M_x = \chi_{xx} h e^{i\omega t} + \exp(-\omega_e'' t)[C_1 \exp(i\omega_e' t) + C_2 \exp(-i\omega_e' t)]\\
+ \exp(-\omega_n'' t)[C_3 \exp(i\omega_n' t) + C_4 \exp(-i\omega_n' t)]
\end{aligned} \tag{2.3}$$

and by the corresponding expressions for M_y, μ_x, and μ_y. Here, χ_{xx} is the complex susceptibility described by an expression given in [1], except that now the components μ_z occurring in this expression are no longer constant but vary from μ to $-\mu$, in accordance with Eq. (2.2). After a time $t \gg (\omega_n'')^{-1}$ the free nuclear-like oscillations die out (the electron-like oscillations die out much earlier because $\omega_e'' \gg \omega_n''$) and the absorption of energy is described completely by Eqs. (7)—(11) from [1] subject to Eq. (2.2).

We shall consider the energy absorbed at the NMR frequency when ω_e and ω_n are far apart: $\omega \sim \omega_n \ll \omega_e$. Introducing the gain $\eta = AM/(H - H_c)$, we shall rewrite the relevant expression in the form [1]

$$P \approx \frac{M}{H - H_c} \frac{\Gamma_e \omega^2}{\omega_e^2} h^2 - \frac{\mu_z}{H_n} \frac{\Gamma_n \omega^2 \omega_n^2}{(\omega_n^2 - \omega^2)^2 + 4\Gamma_n^2 \omega^2} (\eta h)^2, \tag{2.4}$$

where $H_n = AM$ is the effective field on the nucleus. The first term in the above expression describes the nonresonant electron absorption and the second the resonant nuclear absorption or amplification.

Generally, in an inverted state of μ ($\mu_z > 0$) the expression (2.4) describes a reversed NMR signal on a background of nonresonant electron absorption. If $|\omega_q^2| > 4\Gamma_e\Gamma_n$, then the power is $P < 0$ at $\omega = \omega_n$, i.e., the energy is not absorbed by amplification takes place. If the opposite inequality is satisfied, the nonresonant electron absorption predominates over the resonant nuclear amplification. In fact, such a separation into nuclear amplification and electron absorption is somewhat arbitrary. We recall that in the derivation of (2.4) the nuclear susceptibility is ignored because of its smallness. Both terms in Eq. (2.4) are due to the electron susceptibility which—in its turn—can be separated approximately into two components one of which is the intrinsic electron susceptibility and the other is the nuclear susceptibility multiplied by η.

If we assume that $\Gamma_e = 0$ and $\eta = 1$, the first term disappears and Eq. (2.4) reduces to the well-known formula for the gain in an inverted paramagnet. The predicted amplification of electromagnetic field pulses has been observed experimentally in electron [8-10] and nuclear [11] paramagnetic systems. It follows from Eq. (2.4) that in the case of a ferromagnet it is not sufficient to transfer the nuclear system to an inverted state in order to achieve amplification: we must also satisfy the condition $|\omega_q^2| > 4\Gamma_e\Gamma_n$.

We shall now determine the reciprocal of the Q factor of the system Q^{-1}, which represents the relative gain (the gain is $k = 1 + \pi Q^{-1}$):

$$Q^{-1}=|P|/\omega\varepsilon=|P|T/2\pi\varepsilon. \tag{2.5}$$

Here, $|P|T$ is the energy radiated per period and ϵ is the maximum energy stored in the system:

$$\varepsilon=B_x h_x/8\pi\approx{}^1/_2 M_x h_x={}^1/_2\chi' h^2. \tag{2.6}$$

If $|\omega_q^2| \gg 4\Gamma_e\Gamma_n$, then

$$Q^{-1}\approx\left|\frac{\chi''}{\chi'}\right|=2\pi\frac{|\mu_z|}{H_n}\frac{\omega_n}{\Gamma_n}\frac{A\eta}{4\pi}. \tag{2.7}$$

This expression differs from the corresponding expression for the nuclear system in a nonferromagnetic material by the factor $A\eta/4\pi \gg 1$.

The value of P varies with time because of the corresponding variation of μ_z. Consequently, the amplitude of a reversed NMR signal gradually decreases to zero and then a normal NMR signal is observed and its amplitude reaches its steady-state value after a time $\sim T_1$.

We shall now consider the case when the NMR and FMR frequency coincide: $\omega_e = \omega_n$. The power absorbed in the vicinity of this frequency is described by [1]

$$P\sim F(\omega)=\frac{4\Gamma_e(\omega_n-\omega)^2+\Gamma_n(4\Gamma_e\Gamma_n+\omega_q^2)}{[4(\omega_n-\omega)^2-4\Gamma_e\Gamma_n-\omega_q^2]^2+16\Gamma_e^2(\omega_n-\omega)^2}. \tag{2.8}$$

The function $F(\omega)$ in a normal state is of the form shown in Fig. 1 by a continuous curve: against the background of a wide $\Delta\omega_1 \approx 2\Gamma_e$ FMR maximum of amplitude $F_1 \approx 1/4\Gamma_e$ we can see a narrow "reverse resonance" peak. The value of the function at the point $\omega = \omega_n$ is

$$F_2=\frac{1}{4\Gamma_e+\omega_q^2/\Gamma_n} \tag{2.9}$$

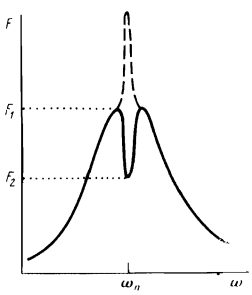
and the half-width of the reverse resonance peak

$$\Delta\omega_2/2=\Gamma_n+\omega_q^2/4\Gamma_e \tag{2.10}$$

is identical with the damping coefficient of nuclear-like oscillations ω_n'' given by Eq. (1.3).

Inversion of the nuclear magnetization produces hardly any changes except in a narrow region near ω_n, where the changes are great. In an inverted state we have, instead of a narrow downward peak, an even narrower upward peak (dashed curve in the figure). During subsequent relaxation the amplitude of the upward peak decreases and it disappears at $\mu_z = 0$ so that only a wide FMR line remains; then, an "reverse resonance" peak appears and begins to grow tending to its steady-state value corresponding to $\mu_z = -\mu$.

It may seem that for $|\omega_q^2|$, close to $4\Gamma_e\Gamma_n$, we may obtain an inverted-state peak as narrow as we please; this is not true because it would violate the condition



$(\omega_n'')^{-1} \ll T_1$ and the case requires special consideration. We shall now give the explicit conditions for the validity of the expressions obtained. During the transient stage before $\Delta\mu_z = \mu_z + \mu$ becomes very small, i.e., as long as $\Delta\mu_z$ is of the order of μ, the second term in the last equation of the system (2.1) should be much smaller than the third term; only then does μ_z relax in accordance with Eq. (2.2). Conversely, the second and third terms become comparable only when

$$\Delta\mu_z/\mu=(\mu_z+\mu)/\mu\ll 1. \tag{2.11}$$

These two conditions are equivalent and can be expressed in the form

$$\gamma_n A(M_x\mu_y-M_y\mu_x)T_1/\mu\ll 1. \tag{2.12}$$

The inequality (2.12) represents the condition for the absence of saturation in the nuclear system. For $\omega = \omega_n \ll \omega_e$, this condition becomes

$$(\gamma_n\eta h)^2 T_1 T_2/2\ll 1, \tag{2.13}$$

and for $\omega = \omega_e = \omega_n$ we have

$$2\Gamma_n\gamma_e^2(4\pi Mh)^2T_1/(\omega_q^2+4\Gamma_e\Gamma_n)^2\ll 1. \tag{2.14}$$

Since an inverted state is not in equilibrium, all the effects considered here can only be observed for a time δt which has upper and lower limits:

$$1/\omega_n''\ll\delta t\ll T_1 \tag{2.15}$$

This gives rise to a frequency indeterminacy $\delta\omega \sim 1/\delta t$. We can see from Eq. (2.15) that this quantity is much smaller than the line width of the fine ENMR structure $\Delta\omega = 2\omega_n''$ so that the limits imposed on δt should not prevent the observation of the effects discussed above.

3. INITIAL TRANSIENT PROCESSES IN ENMR

If a high-frequency field h is greater than the value given by the inequality (2.12), the relaxation of μ_z ceases to be of the quasisteady type. Then, the system (2.1) becomes strongly nonlinear. However, it can still be always linearized in a narrow time interval Δt, beginning at the moment of application of the high-frequency field in a normal ($\mu_z = -\mu$) or inverted ($\mu_z = \mu$) states. In fact, the transverse projections of M and μ are then small and the change in μ_z is proportional to their product. Therefore, we can always find an interval Δt during which we can assume that μ_z = const and consider the first four equations of the system (2.1) as a system of linear equations.

During this time interval the general solution of the system (2.1) is given by expressions of the (2.3) type, where the constants are found from the initial conditions: at t = 0, we have $M_x = M_y = \mu_x = \mu_y = 0$. Assuming that $t \ll T_1$ and $t \ll |\omega_n''|^{-1}$, we expand $\exp(-\omega_n''t)$ as a series retaining the term linear in t and assuming that $\omega = \omega_n$, i.e., that the external frequency is tuned to NMR. We shall first consider the case when the FMR and NMR frequencies are far apart: $\omega_n \ll \omega_e$.

The expression for M_x is

$$M_x\approx-\frac{Mh}{H-H_\kappa}\exp(-\Gamma_e t)\cdot\left(\frac{\Gamma_e}{\omega_e'}\sin\omega_e' t+\cos\omega_e' t\right)$$
$$+\frac{Mh}{H-H_\kappa}\left(\frac{2\Gamma_e\omega_n}{\omega_e^2}\sin\omega_n t+\cos\omega_n t\right)-\frac{1}{2}\mu_z\gamma_n\eta^2 ht\sin\omega_n t. \tag{3.1}$$

The first term describes electron-like oscillations. These oscillations are damped out rapidly and the amplitude M_x then increases to $M_x^{(0)}$, corresponding to the

steady state in the absence of the nuclear subsystem [second term in Eq. (3.1)]. The third term in (3.1) describes the initial transient processes associated with the nuclear-like oscillations.

We shall now consider the absorption of energy in such a transient process after a time $t \gg \Gamma_e^{-1}$. Since the amplitude of M_x varies slowly with time, we shall introduce the average energy absorption per period:

$$P(t) \approx \frac{1}{T}\int_t^{t+T} h_x(t')\frac{d}{dt'}M_x(t')dt' \approx \frac{M}{H-H_к}\frac{\Gamma_e\omega_n^2}{\omega^2}h^2 - \frac{1}{4}\omega_n\mu_z\gamma_n(\eta h)^2 t. \tag{3.2}$$

The absorbed power P reaches rapidly its value $P^{(0)}$ corresponding to the amplitude $M_x^{(0)}$ and then it increases slowly if $\mu_z = -\mu$ or decreases if $\mu_z = \mu$.

We shall now consider the case when the FMR and NMR frequencies coincide: $\omega_e = \omega_n$. The expression for M_x is of the form

$$M_x \approx +e^{-\Gamma_e t}\frac{\omega_z^2 h}{8\pi\Gamma_e\omega_e'}\sin\omega_e' t + \frac{h\omega_z^2}{8\pi\Gamma_e\omega_n}\left(1-\frac{\omega_q^2 t}{4\Gamma_e}\right)\sin\omega_n t. \tag{3.3}$$

The average power absorbed per period after a time $t \gg \Gamma_e^{-1}$ is given by

$$P(t) \approx \frac{\pi(\gamma_e M h)^2}{\Gamma_e}\left(1-\frac{\omega_q^2 t}{4\Gamma_e}\right). \tag{3.4}$$

Since we are considering time intervals $t \ll |\omega_n''|^{-1}$, $t \ll T_1$, the sign and value of ω_n'' are not restricted in any way. The condition of validity of expressions obtained above is the inequality $\mu_{xy}/\mu \ll 1$; for $\omega_n \ll \omega_e$ and $\omega_n = \omega_e$ this condition assumes, respectively, the forms

$$^1/_2\gamma_n\eta h t \ll 1,\ \gamma_e^2\pi M h t/\Gamma_e \ll 1. \tag{3.5}$$

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