Nuclear spin motion with allowance for Suhl-Nakamura interaction

E. A. Turov, M. I. Kurkin, and V. V. Nikolaev
Institute of Metal Physics, USSR Academy of Sciences
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It is shown that the NMR dynamic frequency shift (DFS or pulling) in ferro- and antiferromagnetic substances caused by Suhl-Nakamura interaction of nuclear spins at low temperatures may pronouncedly affect the nonlinear oscillations of nuclear magnetic moments in a varying radio-frequency field near the resonance frequency. Equations are derived which define coupled uniform motion of electron and nuclear spins at NMR frequencies under nonlinear resonance conditions. The equations can be employed for studying the peculiarities of free precession and of spin echo signals in both ferromagnetic and antiferromagnetic substances (of the "easy plane" type). It is shown that pulling may lead to aperiodic solutions for the motion of nuclear magnetization in a rotating coordinate system (with the frequency of the varying field). This signifies, in particular, that under conditions when the solutions are realized there should be no periodic dependence of the free precession signal amplitude on duration of the radio-frequency pulse. The equations of motion are solved for some particular cases. The period of motion and maximal deviation of nuclear magnetization from equilibrium are found. It is shown that the effect of pulling, which impedes the appearance of large deviations, can be avoided by simple sinusoidal modulation of the radio-frequency pulse frequency.

1. FORMULATION OF PROBLEM

In ferro- and antiferromagnets with sufficiently high concentration of the nuclei that possess magnetic moments, the hyperfine coupling of the nuclei with the electrons can lead to a strong indirect interaction between the nuclear spins (the interaction after Suhl and Nakamura[1], see also[2]). There are two known effects due to these interactions: the broadening of the NMR line, and the dynamic shift of the resonant frequency (pulling)[3,2]. The latter is observed only at sufficiently low temperature, since it is proportional to the nuclear magnetization $m = \text{const}/T_S$, where T_S is the temperature of the nuclear spin system. This temperature can change when NMR is saturated, so that pulling causes also a number of nonlinear phenomena observed in the stationary regime[4,5]. However, NMR in magnets is investigated in many cases by a nonstationary (pulsed) procedure, such as the spin-echo procedure, the free-precession damping procedure, etc. Spin echo was also observed recently in certain antiferromagnets under strong pulling conditions[6].

It is quite obvious that in those cases when the pulling is large, it should influence strongly the indicated quasistationary phenomena. Yet theoretical calculations of the motion of nuclear magnetization in strong pulsed fields of resonant frequency, with allowance for pulling, have as yet not been published. In our opinion, this hinders also the experimental research in this field to a certain degree.

The present paper is the first attempt to take into account the influence of the Suhl-Nakamura interaction on the motion of nuclear magnetization in strong resonant-frequency fields, when the deviation of the nuclear moment from the equilibrium orientation can be arbitrarily large. The duration of the resonant-field pulse will be assumed small in comparison with the times of nuclear magnetic relaxation, so as to be able to neglect relaxation processes. Such a situation corresponds to the conditions of free-precession-damping and spin-echo signals.

Before we proceed to a direct calculation for concrete case of ferro- and antiferromagnets, let us formulate the general limitations under which the problem is solved. First, we assume that the resonant-frequency field and the hyperfine interaction have little influence on the behavior of the electronic magnetization in the sense that the motion of the latter can be described by using the formulas of the theory of the linear response, both with respect to the external resonant-frequency field, and with respect to the hyperfine field due to the nuclei. Strictly speaking, only in this case can the interaction of the transverse component of the electronic and nuclear spins be reduced to an effective (Suhl-Nakamura) interaction between the nuclear spins. This will also mean that the pulling should be small in comparison with the unshifted NMR frequency:

$$\omega_n = \gamma_n H_n, \tag{1}$$

where γ_n is the gyromagnetic ratio of the nuclei and H_n is the static hyperfine field acting on them.

It must also be recognized that the dynamic interaction of the electronic and nuclear magnetizations can be regarded as a small perturbation if the difference between the natural frequency of the oscillations of these magnetizations is much larger than the hyperfine interaction. Since we are interested in the behavior of the system near the NMR frequency, the motion of the electronic subsystem at such frequencies can be regarded in the quasistatic approximation, by replacing its dynamic-susceptibility tensor by the corresponding static tensor χ_0.

We are interested in ferro- and antiferromagnets with large hyperfine fields at the nuclei and with large gains of the resonant frequency field. The direct interaction between the nuclear magnetization and the external fields can then be neglected and we can assume that these fields act only on the electronic magnetization.

Finally, we consider only homogeneous and spatial oscillations of the magnetization and neglect also the quadrupole splitting of the nuclear spins. Under these conditions, the problem is easiest to solve on the basis of the classical equation of motion for the magnetic moments in the corresponding effective fields.

2. EQUATIONS OF MOTION IN THE CASE OF A FERROMAGNET

We consider first an arbitrary ferromagnet magnetized to saturation in the direction of the z axis. We need to know only the components of its magnetic susceptibility tensor relative to the transverse resonance-frequency field in the quasistatic approximation, i.e., χ_{xx} and χ_{yy}. The calculation presented below pertains to practically all known concrete cases for different (ellipsoidal) sample shapes and different crystal-lattice symmetries.

In accordance with the statements made above, we describe the motion of the nuclear magnetization m by the equation

$$d\mathbf{m}/dt=\gamma_n[\mathbf{m}\mathbf{H}_{\text{eff}}],\qquad \mathbf{H}_{\text{eff}}=A\mathbf{M},\tag{2}$$

where $\mathbf{H}_{\text{eff}}$ is the effective hyperfine field acting on the nuclei as a result of the electronic magnetization M (A is the dimensionless hyperfine coupling constant). The electronic magnetization moves under the influence of a resonant-frequency field h(t) that is perpendicular to the z axis, and an effective hyperfine field Am produced by the nuclei. In other words

$$M_x=\chi_{xx}(h_x+Am_x),$$
$$M_y=\chi_{yy}(h_y+Am_y),\qquad M_z\approx M_0,\tag{3}$$

where M_0 is the saturation magnetization. The appearance of pulling is precisely connected with the reaction of the nuclear magnetization on the electronic magnetization, a reaction accounted for by the Am terms in (3).

After substituting (3), Eq. (2) in terms of the components takes the form of the following system:

$$\begin{aligned}\dot m_x&=\gamma_n(H_n-A\eta_2m_z)m_y-\gamma_n\eta_2h_ym_z,\\ \dot m_y&=-\gamma_n(H_n-A\eta_1m_z)m_x+\gamma_n\eta_1h_xm_z,\\ \dot m_z&=\gamma_nA(\eta_2-\eta_1)m_xm_y+\gamma_n\eta_2h_ym_x-\gamma_n\eta_1h_xm_y.\end{aligned}\tag{4}$$

Here $H_n = AM_0$ is the dc component of the hyperfine field and acts on the nuclear spin along the z axis, while $\eta_1 = A\chi_{xx}$ and $\eta_2 = A\chi_{yy}$ are the gains of the resonant-frequency field in the directions x and y, respectively. In the approximation linear in the resonance-frequency field h(t), we obtain from the system (4) $m_z \approx m$ = const, and for the NMR frequency with allowance for the pulling we obtain

$$\Omega_n=\gamma_n[(H_n-A\eta_1m)(H_n-A\eta_2m)]^{1/2}\tag{5}$$

or approximately $\Omega_n = \omega_n - \omega_p$, where

$$\omega_p=\frac{1}{2}(\eta_1+\eta_2)\frac{m}{M_0}\omega_n\tag{6}$$

is precisely that change of the NMR which is defined as pulling.

We now assume, for concreteness, that

$$h_x=2H_1\cos\omega t,\quad h_y=0,$$

and transform the equations in (4) in a coordinate system that rotates about the z axis, keeping in mind the fact that the effective field ω/γ_n connected with this rotation almost cancels out the longitudinal field H_n. In this coordinate system the equations in (4) contain terms that do not depend explicitly on the time, and also terms proportional to sin 2ωt and cos 2ωt. When considering nonlinear resonance at frequencies $\omega \sim \omega_n \sim \Omega_n$, we should discard the terms of the latter type, since they result only in small rapidly-oscillating (with frequency 2ω) corrections to the fundamental "slow" motion of interest to us, with angular frequencies of the order of $\omega_1 = \gamma_n H_1\eta_1$ or ω_p, which is registered by the nonstationary NMR methods. As a result, Eqs. (4) take in the rotating coordinate system the form

$$\begin{aligned}\dot m_x&=m_y\left(\Delta\omega-\omega_p\frac{m_z}{m}\right),\\ \dot m_y&=-m_x\left(\Delta\omega-\omega_p\frac{m_z}{m}\right)+\omega_1m_z,\qquad \dot m_z=-\omega_1m_y,\end{aligned}\tag{7}$$

where $\Delta\omega = \omega_n - \omega$. We did not introduce new symbols for the rotating coordinate axes, since the coordinate frame referred to will be clear from the text.

Before we proceed to solve the nonlinear system (7), we derive the corresponding equations of motion for an antiferromagnet of the "easy plane" type since, as will be shown later, they also reduce to a system of the form (7).

3. EQUATIONS OF MOTION IN THE CASE OF AN ANTIFERROMAGNET OF THE "EASY PLANE" TYPE

Assume that the z axis of a uniaxial antiferromagnet of the "easy plane" type is the symmetry axis, the equilibrium vector of the antiferromagnet is directed along the z axis, and the constant magnetic field H_0 is directed along the x axis (Fig. 1). In this case, as is well known,[2] the greatest gain will occur in a resonance-frequency field parallel to the z axis. We therefore assume

$$h_z=2H_1\cos\omega t,\qquad h_x=h_y=0.\tag{8}$$

The energy of such an antiferromagnet (per unit volume), with allowance for the hyperfine interaction, can be written in the form

$$\begin{aligned}\mathscr{H}=&\frac{J}{M_0^2}\mathbf{M}_1\mathbf{M}_2+\frac{K}{M_0^2}(M_{1y}^2+M_{2y}^2)+\frac{D}{M_0^2}(M_{1x}M_{2z}-M_{1z}M_{2x})\\ &-H_0(M_{1x}+M_{2x})-h_z(M_{1z}+M_{2z})-A(\mathbf{m}_1\mathbf{M}_1+\mathbf{m}_2\mathbf{M}_2).\end{aligned}\tag{9}$$

Here $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetizations of the sublattices while $\mathbf{m}_1$ and $\mathbf{m}_2$ are the corresponding nuclear magnetizations. The first and second terms in (9) are respectively the exchange and anisotropy energies, while the third takes into account the weak ferromagnetism (which appears frequently in antiferromagnets of the "easy plane" type). The meaning of the remaining terms is evident without explanation.

For the calculations that follow, it is convenient to express the energy (9) in terms of other variables, defined by the transformation

$$\begin{aligned}M_{1x}&=M_{1x_1}\cos\psi+M_{1z_1}\sin\psi,\\ M_{1z}&=-M_{1x_1}\sin\psi+M_{1z_1}\cos\psi,\\ M_{2x}&=-M_{2x_2}\cos\psi+M_{2z_2}\sin\psi,\\ M_{2z}&=-M_{2x_2}\sin\psi-M_{2z_2}\cos\psi,\end{aligned}\tag{10}$$

where ψ is the angle by which the equilibrium vectors $\mathbf{M}_1$ and $\mathbf{M}_2$ are deflected from the z axis under the influence of the field H_0. This is actually a rotation transformation (about the y axis) to the proper coordinate system of each of the sublattices, the axis z_i (i = 1, 2) being the equilibrium directions of the magnetizations of the corresponding sublattices. The vectors $\mathbf{m}_1$ and $\mathbf{m}_2$ will also be expressed in the same system.

Transforming $\mathscr{H}$ in accordance with (10) and minimizing the equilibrium energy with respect to the angle ψ, we have

$$\sin\psi\approx\frac{H_0+H_D}{H_E};\qquad H_E=\frac{2J}{M_0},\qquad H_D=\frac{D}{M_0}.\tag{11}$$

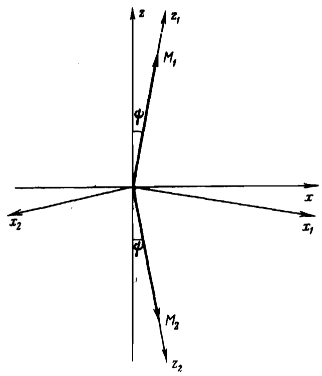

FIG. 1. Coordinate axes of the laboratory system (x, y, z) and of the systems (x_1, y_1, z_1) and (x_2, y_2, z_2) connected respectively with the sublattice magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$.

We assume here that the exchange field $H_E \gg (H_0 + H_D)$, therefore $\sin\psi \ll 1$. With this relation taken into account, the energy (9) in terms of the new variables assumes the approximate form

$$\mathscr{H} = \frac{H_E}{2M_0}(-M_{1x_1}M_{2x_2} + M_{1y}M_{2y} - M_{1z_1}M_{2z_2}) + \frac{H_A}{2M_0}(M_{1y}^2 + M_{2y}^2) + \\ + \frac{H_0^2 - H_D^2}{H_E M_0}(M_{1x_1}M_{2x_2} + M_{1z_1}M_{2z_2}) - A(\mathbf{m}_1\mathbf{M}_1 + \mathbf{m}_2\mathbf{M}_2) \\ + 2H_1\cos\omega t\frac{H_0 + H_D}{H_E}(M_{1x_1} + M_{2x_2}), \tag{12}$$

where $H_A = 2K/M_0$.

We can next write down the equations of motion for $\mathbf{M}_1$ and $\mathbf{M}_2$ in the corresponding effective fields:

$$H^{(i)} = -\partial\mathscr{H}/\partial M_i, \quad i = 1, 2.$$

After linearizing these equations, and recognizing that $M_{1z_1} \approx M_{2z_2} \approx M_0$, and M_{1x_1}, M_{3x_2}, M_{1y}, $M_{2y} \ll M_0$, we obtain a system of four equations. If we introduce new variables of the type

$$M_\pm^\alpha = M_{1\alpha_1} \pm M_{2\alpha_2}, \quad m_\pm^\alpha = m_{1\alpha_1} \pm m_{2\alpha_2}, \tag{13}$$

then this system breaks up into two independent subsystems corresponding to two branches of the natural oscillations of the magnetic moments of the antiferromagnet. The first system

$$\frac{1}{\gamma}\dot{M}_+^x = (H_E + H_A)M_+^y - H_n m_+^y, \\ \frac{1}{\gamma}\dot{M}_+^y = -\frac{H_0(H_0 + H_D)}{H_E}M_+^x - 4\frac{H_0 + H_D}{H_E}M_0 H_1\cos\omega t + H_n m_+^x \tag{14}$$

corresponds to the low-frequency branch of the oscillations with frequency

$$\omega_+ \approx \gamma[H_0(H_0 + H_D)]^{1/2}. \tag{15}$$

The second subsystem

$$\frac{1}{\gamma}\dot{M}_-^x = \left[H_A + \frac{D_D(H_0 + H_D)}{H_E}\right]M_-^y - H_n m_-^y, \\ \frac{1}{\nu}\dot{M}_-^y = -H_E M_-^x + H_n m_-^x \tag{16}$$

corresponds to the high-frequency branch of the oscillations with frequency

$$\omega_+ = \gamma[H_E H_A + H_D(H_0 + H_D)]^{1/2}.$$

The resonance-frequency field does not enter in the subsystem (16), so that the oscillations of the second branch are not excited by a field parallel to the z axis. And if the initial conditions are such that

$$M_-^\alpha = m_-^\alpha = 0, \quad \alpha = x, y, z, \tag{17}$$

then these variables remain equal to zero both when a resonance-frequency pulse is applied, and when the nuclear spins move in the interval between the pulses[1)]. In accordance with our program, we go over in the subsystem (14) to a quasistatic approximation, putting $M_+^x = M_+^y = 0$, which is possible in this case if $\omega \sim \omega_n \ll \omega_+$. As a result we get

$$M_+^x \approx -4\frac{M_0}{H_0}H_1\cos\omega t + \frac{\gamma^2 H_E H_n}{\omega_+^2}m_+^x, \tag{18}$$

$$M_+^y \approx \frac{H_n}{H_E}m_+^y, \tag{19}$$

from which we see that $M_+^y \ll M_+^x$, and we can set with good approximation $M_+^y = 0$. This together with (17) yields

$$M_{1x_1} = M_{2x_2} = {}^1/_2 M_2^x, \quad M_{1y} = M_{2y} \approx 0, \tag{20}$$

$$m_{1\alpha_1} = m_{2\alpha_2} = {}^1/_2 m_+^\alpha. \tag{21}$$

We consider now the motion of the nuclear magnetizations $\mathbf{m}_1$ and $\mathbf{m}_2$ in the effective fields $A\mathbf{M}_1$ and $A\mathbf{M}_2$. If we write the corresponding equations of motion in terms of the variables (13) with allowance for (20) and (21), then after going over to a rotating coordinate system[2)] and discarding the small rapidly oscillating terms, we again obtain a system of the form (7), in which now $m_\alpha \equiv m_+^\alpha = 2m_{1\alpha_1} = 2m_{2\alpha_2}$ ($m = [m_x^2 + m_y^2 + m_z^2]^{1/2}$), $\omega_1 = \gamma_n H_1 \eta$, where $\eta = H_n/H_0$ is the gain and

$$\omega_p = \frac{1}{4}\omega_n\eta\frac{H_E}{H_0 + H_D}\frac{m}{M_0}$$

is the maximum pulling. Comparing the last formula with (6), we see that in this case the pulling is many times larger than in the case of a ferromagnet (at the same values of the other parameters).

We note further that for the considered type of oscillations, according to (21), the motion of the nuclear magnetizations of the sublattice relative to the proper (rotating) coordinate systems is quite identical, so that m_α in the system (7) can also be taken to mean the components of the nuclear magnetizations of each of the sublattices (in the proper coordinate system).

After solving (7) with definite initial conditions, we can use (18) and (19) (at $H_1 = 0$) to find the amplitude of the free-precession signals amplified by the electronic subsystem. As already noted, the largest response to the motion of the nuclear magnetization can be exhibited by the variable M_+^x.. According to (10), however, this means that in the common coordinate system connected with the constant magnetic field ($H_0 \parallel x$) the precession of the nuclear magnetizations will induce the largest summary electronic magnetization along the z axis:

$$M_{1z} + M_{2z} = -(M_{1x_1} + M_{2x_2})\sin\psi \approx -\frac{H_0 + H_D}{H_E}M_+^x = -\eta m_+^x. \tag{22}$$

After determining the maximum value of m_+^x for each of the solutions of the system (7) of interest to us we can use (22) to estimate the amplitude of the signal registered at the frequency $\omega \sim \omega_n$. Just as in the case of a ferromagnetic (see formula (3)), the gain η for the free-precession signal coincides with the gain for the resonant-frequency field. (The direct signal would be the largest from the component m_+^y of the nuclear magnetization.)

4. PERIODIC SOLUTIONS

Thus, in both cases (ferromagnet and antiferromagnet) the problem has been reduced to the solution of the nonlinear system (7). Since $m^2 = m_x^2 + m_y^2 + m_z^2$ is an integral of the motion, it is convenient to solve this problem by changing over to the polar coordinates $m_x = m \sin\theta\cos\varphi$, $m_y = m\sin\theta\sin\varphi$ and $m_z = m\cos\theta$. We then obtain in place of (7) the two equations

$$\dot\theta = \omega_1 \sin\varphi, \tag{23}$$

$$\dot\varphi \sin\theta = \omega_1\cos\theta\cos\varphi - (\Delta\omega - \omega_p\cos\theta)\sin\theta. \tag{24}$$

It is easy to see that this system has one more integral of motion:

$$\lambda = -\omega_1\sin\theta\cos\varphi - \Delta\omega\cos\theta + {}^1/_2\omega_p\cos^2\theta, \tag{25}$$

which represents (apart from a constant factor) the energy of the system. The concrete value of λ is determined from the initial conditions of the problem. If we obtain $\sin\varphi$ as a function of $\cos\theta$ from (25) and substitute it in (23), the latter can be easily integrated:

$$\pm\int\limits_{\cos\theta_0}^{\cos\theta}\frac{d\xi}{\sqrt{P_4(\xi)}} = \omega_p(t-t_0), \tag{26}$$

where

$$P_4(\xi) = \left(\frac{\omega_1}{\omega_p}\right)^2(1-\xi^2) - \left(\frac{1}{2}\xi^2 - \frac{\Delta\omega}{\omega_p}\xi - \frac{\lambda}{\omega_p}\right)^2 \tag{27}$$

is a polynomial of fourth degree, and θ_0 and θ are respectively the polar angles at the initial instant of time t_0 and at the instant of time t. The sign in the left-hand side of (26) is determined by the initial value of φ.

Formula (26) solves our problem in principle, since we can determine for arbitrary initial conditions the variation of the angle θ as a function of the duration of the resonance-frequency pulse $(t - t_0)$, and we can then determine also the angle φ from (25). If the parameters ω_1, $\Delta\omega = \omega_n - \omega$ and ω_p are known, then a successful application of (26) makes it possible to calculate numerically not only the amplitude of the free-precession signal but also the spin-echo signals. We were unable to carry out an analytic investigation of the solution (26) in the general case, since this calls for knowledge of the roots of the polynomial (27), which are of very cumbersome form. We can only state that, with the exception of certain special cases referred to in the next section, $\cos\theta$ is a periodic function of the duration of the resonance-frequency pulse and is expressed in terms of Jacobi elliptic functions. We consider below some of the simplest particular cases of definite practical interest.

1. The Case $\Delta\omega = \omega_n = 0$, $\theta_0 = 0$

Let the frequency of the resonance-frequency field coincide with the unshifted (by pulling) NMR frequency ($\omega = \omega_n$), and let the vector $\mathbf{m}$ begin its motion from an equilibrium position at the instant of time $t_0 = 0$. Under these conditions, according to (25), we have $\lambda = \omega_p/2$ and formula (26) takes the form

$$\int\limits_0^\theta\frac{d\theta}{\sqrt{1-\varepsilon^{-2}\sin^2\theta}} = \omega_1 t, \tag{28}$$

where $\epsilon = 2\omega_1/\omega_p$. From (28) we have[7]

$$\sin\theta = \mathrm{sn}(\omega_1 t;\ \varepsilon^{-1}) \quad \text{if} \quad \varepsilon \geqslant 1, \tag{28$'$}$$

$$\sin\theta = \varepsilon\,\mathrm{sn}({}^1/_2\omega_p t;\ \varepsilon) \quad \text{if} \quad \varepsilon \leqslant 1. \tag{28$''$}$$

Thus, in this case $\sin\theta$ is a periodic function of t with a period

$$T = 4K(\varepsilon^{-1})/\omega_1 \quad \text{if} \quad \varepsilon > 1, \tag{29}$$

$$T = 8K(\varepsilon)/\omega_p \quad \text{if} \quad \varepsilon < 1, \tag{30}$$

where $K(k)$ is a complete elliptic integral of the first kind for the modulus $k = \epsilon^{-1}$ or ϵ, respectively. If $\epsilon^2 = 1$, both solutions coincide and degenerate to the nonperiodic function

$$\sin\theta = \mathrm{th}\,\omega_1 t. \tag{31}$$

This is a new type of motion connected with the pulling, and we shall deal with such aperiodic solutions in greater detail in the next section. As to the periodic motions, their period and amplitude depend essentially on the parameter ϵ. In particular, for an antiferromagnet of the "easy plane" type

$$\varepsilon = 2\frac{\omega_1}{\omega_p} = \frac{H_1}{H_n}\frac{M_H}{m}, \tag{32}$$

where

$$M_H = 2M_0(H_0 + H_D)/H_E \equiv \chi_\perp(H_0 + H_D) \tag{33}$$

is the electronic static magnetization of the antiferromagnet in a transverse field H_0, and

$$m = (\hbar\gamma_n)^2 I(I+1)NH_n/3k_BT \tag{34}$$

is double the nuclear static magnetization of one sublattice at a temperature T in the hyperfine field H_n; N is the number of magnetic nuclei per unit volume in both sublattices.

At low temperatures (in the helium region) the pulling for such antiferromagnets is usually so large that $\epsilon \ll 1$. In this case, according to (28$''$) and (30), the amplitude of the deviation of the nuclear magnetization from the equilibrium position and the period of the motion decrease by a factor ϵ in comparison with their values in the absence of pulling.

2. The Case $\Delta\omega \equiv \omega_n - \omega = \omega_p$, $\theta_0 = 0$

Assume now that at the same initial conditions the frequency of the resonance-frequency field is equal to the NMR frequency with allowance for pulling ($\omega = \Omega_n = \omega_n - \omega_p$). In this case $\lambda = -\omega_p/2$,

$$P_4(\xi) = {}^1/_4[\varepsilon^2(1-\xi^2) - (1-\xi)^4], \tag{35}$$

and the solution (26) takes the form[7]:

$$\sin^2\frac{\theta}{2} = \frac{1}{4}\varepsilon^2\left\{\mathscr{P}\left(\frac{1}{2}\omega_p t;\ g_2, g_3\right) + \frac{1}{12}\varepsilon^2\right\}^{-1}, \tag{36}$$

where $\mathscr{P}$ is the Weierstrauss function with the invariants $g_2 = \epsilon^4/12$ and $g_3 = \epsilon^4/4 + \epsilon^6/216$.

If $\epsilon \lesssim 1$ then, accurate to quantities of the order of 10^{-2}, the function $\mathscr{P}$ can be expressed in terms of Jacobi elliptic functions[8]

$$\mathscr{P}\left(\frac{1}{2}\omega_p t;\ g_2, g_3\right) \approx \left(\frac{\varepsilon}{2}\right)^{4/3}\left[1 + \sqrt{3}\frac{1 + \mathrm{cn}(3^{1/4}(\varepsilon/2)^{2/3}\omega_p t;\ k)}{1 - \mathrm{cn}(3^{1/4}(\varepsilon/2)^{2/3}\omega_p t;\ k)}\right], \tag{37}$$

where the modulus of the elliptic function is $k = (2 - \sqrt{3})^{1/2}/2 \approx 0.26$. Thus, $\sin^2(\theta/2)$ is a periodic function of t, its period is determined by the period of the function cn in (37) and is therefore equal to

$$T = \frac{4K(k)}{3^{1/4}\omega_p^{1/3}\omega^{2/3}} \approx \frac{2\pi}{3^{1/4}\omega_1}\left(\frac{\varepsilon}{2}\right)^{1/3}. \tag{38}$$

The maximum value of the angle θ corresponds to the condition $\mathrm{cn} = -1$ and is determined by

$$\sin\frac{\theta_{max}}{2} = \left(\frac{\varepsilon}{2}\right)^{1/3}\left[1 + \frac{1}{3}\left(\frac{\varepsilon}{2}\right)^{2/3}\right]^{-1/2}. \tag{39}$$

Consequently, with increasing pulling (with decreasing ϵ) the maximum deviation decreases like $\epsilon^{1/3}$, i.e., more slowly than in the preceding case. For example, at $\omega_1/\omega_p \sim 0.1$ the angle θ_{max} is of the order of 60°.

5. APERIODIC SOLUTIONS

We shall show now that among the solutions (26) there are not only periodic but also aperiodic solutions. This new type of motion of the nuclear moment is connected precisely with the pulling. One particular case of such motion was already encountered above (see formula (31)). Let us investigate the possibility of the appearance of aperiodic motions in more general cases.

From the general form of the solution (26) it follows that the aperiodic motions correspond to the case when the polynomial $P_4(\xi)$ has at least two coinciding real roots $|\xi| \leq 1$. Indeed, in this case the integral (26) diverges if the value of such root lies inside the integration interval. This means that rotation of the vector m through the corresponding angle θ will occur after an infinitely long time, meaning that the nuclear magnetization approaches asymptotically a certain limiting direction[3)].

The physical cause of the possible realization of the aperiodic motion is the following. As seen from the system (7), the role of the pulling reduces to the fact that when the vector m moves a change takes place in the z-projection of the effective field (together with the change of m_z) in which this motion takes place. It may turn out that the trajectory of m assumes a direction that is parallel (or antiparallel) to the effective-field vector. Then

$$\dot{\mathbf{m}} = \gamma_n [\mathbf{m} \times \mathbf{H}_{\text{eff}}] = 0 \qquad (40)$$

and the vector m will approach the indicated direction asymptotically. It can be shown that the requirement (40) and the conditions for the multiplicity of the real roots of the polynomial $P_4(\xi)$ are equivalent.

Let us find the relations that must be satisfied by the parameters $\Delta\omega$, ω_1, and ω_p in order for the motion to be aperiodic, if it again begins from the equilibrium position $m \parallel z$ ($\cos\theta_0 = 1$). In this case

$$P_4(\xi) = (1-\xi) P_3(y); \qquad (41)$$
$$P_3(y) = y^3 - 3py - 2q, \quad y = 1 - \xi - {}^2/_3\beta,$$

$$q = {}^1/_{27}(-\beta^3 - 9\beta\varepsilon^2 + 27\varepsilon^2); \qquad (42)$$
$$p = \frac{-3\varepsilon^2 + \beta^2}{9}, \quad \beta = 2\left(1 - \frac{\Delta\omega}{\omega_p}\right).$$

If we disregard the trivial case when the roots of $P_4(\xi)$ coincide (namely, the roots $\cos\theta = 1$), which is realized when $\omega_1 = 0$, then the multiple roots of interest to us should be the roots of the polynomial $P_3(y)$. The condition for the coincidence of the two real roots of such a polynomial, as is well known, is the equality $p^3 = q^2$, which, taking (42) into account, takes the form

$$\varepsilon^4 + (2\beta^2 - 18\beta + 27)\varepsilon^2 + \beta^3(\beta - 2) = 0. \qquad (43)$$

This relation is satisfied at real ϵ if

$$0 \leq \beta \leq {}^9/_4. \qquad (44)$$

The values of ϵ^2 corresponding to these values of β are determined by (43) and are represented by curve a in Fig. 2. The solution (26) under the condition (43) can be written in the form

$$\int_0 \frac{dx}{(x - u_m)[x({}^2/_3\beta + 2q^{1/3} - x)]^{1/2}} = \frac{\omega_p}{2} t, \qquad (45)$$

where $u = 1 - \cos\theta$ and $u_m = 2\beta/3 = q^{1/3}$. The relation $u_m = 1 - \cos\theta_m$ determines the limiting value of the angle θ as $t \to \infty$. The dependence of $\cos\theta_m$ on the parameter β is shown by curve d of Fig. 2.

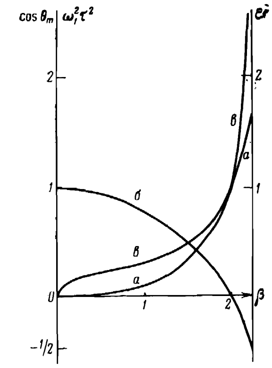

FIG. 2. Plots representing the parameter $\epsilon = 2\omega_1/\omega_p$(a), the limiting deflection angle θ_m (b), and the effective time τ (c) during which the limiting angle θ_m is reached, as functions of the parameter $\beta = 2(1 - \Delta\omega/\omega_p)$ for aperiodic motion of the nuclear magnetization.

The integral in formula (45) can be easily calculated, and the result is as follows:

$$u = u_m \left(\text{ch}\frac{t}{\tau} - 1\right) \Big/ \left(\text{ch}\frac{t}{\tau} + a\right), \qquad (46)$$

where $a = (2u_m - \beta)/(u_m - \beta)$ and the parameter

$$\tau = [{}^1/_2\omega_p \sqrt{u_m(2\beta - 3u_m)}\,]^{-1} \qquad (47)$$

has the meaning of the effective time after which the limiting angle θ_m is actually reached. Such a parameter can be introduced, since, according to (46), $u \to u_m$ exponentially when $t \gg \tau$.

The possible values of τ as functions of β are shown in Fig. 2 by curve c. For $\beta \ll 1$ we have approximately from (47), taking (42) into account,[4)]

$$\tau \approx \frac{1}{\omega_1} \frac{(2\varepsilon)^{1/3}}{\sqrt{3}} \ll \frac{1}{\omega_1}.$$

Here $1 \cos\theta_m \approx {}^1/_3\beta \approx {}^1/_2(2\epsilon)^{2/3}$. With increasing β (and accordingly ϵ), the time τ increases, and at $\beta = 2$ ($\epsilon = 1$) it reaches in accordance with (31) the value $1/\omega_1$. Simultaneously, the formula (46) goes over into (31). The time τ approaches infinity as $\beta \to {}^9/_4$, meaning a transition from an exponential law to a power law. Indeed, by calculating in (46) the limit as $\beta \to {}^9/_4$, corresponding to $(2\beta - 3u_m) \to 0$ we get

$$u = \frac{3}{2} \frac{\omega_1^2 t^2}{\omega_1^2 t^2 + 3}.$$

6. POSSIBILITY OF INCREASING THE NMR INTENSITY BY FREQUENCY MODULATING THE RESONANCE-FREQUENCY FIELD

According to (39), we cannot obtain large changes of the angle θ at sufficiently low values of the ratio $\epsilon = 2\omega_1/\omega_p$, even if the frequency of the resonance-frequency field coincided with the NMR frequency at the initial instant of time. This case corresponds precisely to the experimental situation in[6]. Since the variation of the angle θ under the influence of the resonance-frequency pulses governs the intensity of the echo signals[9], the question arises whether the variation of this angle can be increased by modulating the frequency of the resonance-frequency field. Indeed, were it possible to vary the frequency of the resonance-frequency field so as to keep it resonance on any section of the trajectory of the vector m, then it would be possible to obtain any change of the angle θ. The practical realization of such a possibility depends, however, on two circumstances: first, on whether the indicated resonance conditions can be realized for the entire volume of the sample, and second on the complexity of such a modulation.

Let us assume first an inhomogeneous broadening $\delta\Omega \ll \omega_1$. Then, as follows from (7) or from (23) and (24), the law governing the modulation and determined by the condition for resonance at all the points of the trajectory of motion m, is given by

$$\omega(t) = \omega_n - \omega_p \cos\theta(t), \quad (48)$$

where the function $\cos\theta(t)$ should satisfy the system (23, 24). When the conditions (48) is satisfied, this system describes the precession of the vector m about the x axis with frequency ω_1,

$$\cos\theta(t) = a\cos(\omega_1 t + \theta_0), \quad (49)$$

where the quantities a and θ_0 are given by the initial conditions.

We can now assess the modulation needed for the frequency ω in order to maximize the intensity of a two-pulse echo signal. To this end, as is well known[9], it is necessary that the first pulse be as close as possible to 90°, and the second to 180°. Before the first pulse, the vector m was in equilibrium, i.e., it was oriented along the z axis. For such initial conditions we have from (48) $\theta = \omega_1 t$. Thus, the first pulse will be a 90° pulse for all nuclear spins, if the frequency of the resonant-frequency field varies like

$$\omega(t) = \omega_n - \omega_p \cos\omega_1 t, \quad (50)$$

and the pulse duration is $t_1 = \pi/2\omega_1$.

The analysis of the second pulse is a more complicated task. The point is that in the interval between the pulses the transverse component of m decays because of the inhomogeneity of the NMR frequency in different points of the sample. In other words, the vector m becomes essentially inhomogeneous over the sample. Yet Eqs. (7), on which the entire analysis was based, were obtained under the assumption that m is homogeneous.

It is easy to see that the role played by pulling during the time of the second pulse depends strongly on the ratio of the spatial scale of the inhomogeneity of m to the radius of the Suhl-Nakamura interaction. We shall demonstrate this using a ferromagnet as an example and writing for this purpose one of the equations of the system (4) (e.g., the first), without assuming that m is homogeneous:

$$\dot{m}_x(\mathbf{r}) = \omega_n m_y(\mathbf{r}) - \gamma_n \eta h_y m_z(\mathbf{r}) - \gamma_n A^2 m_z(\mathbf{r}) \int \chi(\mathbf{r}-\mathbf{r}') m_y(\mathbf{r}') d\mathbf{r}'. \quad (51)$$

Here

$$\chi(\mathbf{r}-\mathbf{r}') = \frac{\gamma M_0}{V} \sum_{\mathbf{k}} \omega_{\mathbf{k}}^{-1} e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')}$$

is the local electronic magnetic susceptibility with respect to the homogeneous field, such that the integral

$$\int \chi(\mathbf{r}-\mathbf{r}') d\mathbf{r}' = \chi_\perp = \chi_{xx} = \chi_{yy} = \gamma M_0/\omega_0 \quad (52)$$

gives the total susceptibility (per unit volume) in a homogeneous field. As seen from (51), $\chi(\mathbf{r} - \mathbf{r}')$ is simultaneously the kernel of the Suhl-Nakamura interaction.

We can now separate two cases by introducing the characteristic scale $r_0 = v_0^{1/3}$, which is the linear dimension of the minimal region within which the average value of the transverse component of m (at the instant when the second pulse is turned on) vanishes, i.e.,

$$\int_{v_0} m_x(\mathbf{r}) d\mathbf{r} = \int_{v_0} m_y(\mathbf{r}) d\mathbf{r} = 0. \quad (53)$$

Let $r_0 \ll r_{SN}$, where r_{SN} is the radius of the Suhl-Nakamura interaction. In this case $\chi(\mathbf{r} - \mathbf{r}')$ can be approximately regarded as constant over distances of the order of r_0, so that the integral in the right-hand side of (51) vanishes by virtue of (53). This means that the nuclear spins will move just as if there were no pulling. Thus, at $r_0 \ll r_{SN}$ the second pulse will be 180° for all spins at a constant frequency of the resonance-frequency field $\omega = \omega_n$ and at a pulse duration $t_2 = \pi/\omega_1$.

In the other limiting case $r \gg r_{SN}$, the quantity $m_y(\mathbf{r}')$ can be regarded as constant in a region with dimensions of the order of r_{SN} and taken outside the integral sign in (51). Then, taking (52) into account, we obtain an equation that coincides exactly with the first equation in (4). Then the problem of the motion of the nuclear spins during the time of the second pulse again reduces to the system (7). The initial value of the angle θ_0 can then be regarded the same for all spins ($\theta_0 = \pi/2$), whereas the initial angle φ_0 can assume all values from 0 to 2π. In this case we can attempt to offset the action of the pulling, which leads to a deviation from resonance, just as in the time of the first pulse, by frequency-modulating the resonance-frequency field in accordance with (49). The constant a in this case, however, depends on the initial value of the angle φ, namely $a = \sin\varphi_0$. Therefore the frequency modulation necessary for the resonance should be given by

$$\omega(t) = \omega_n + \omega_p \sin\varphi_0 \sin\omega_1 t, \quad (54)$$

meaning that the resonance conditions cannot be simultaneously satisfied for all points of the sample (since they correspond to different angles φ_0). It can be assumed, however, that in the case of a modulation in accordance with (54) the resonance conditions are approximately satisfied not for some particular concrete value of φ_0, but for a certain angle interval $\delta\varphi$ defined by the inequality

$$\delta\omega < \omega_1, \quad \delta\omega = \omega_p [\sin(\varphi_0 + \delta\varphi) - \sin\varphi_0], \quad (55)$$

where $\delta\omega$ is the maximum frequency deviation for the indicated interval of the angles $\delta\varphi$ in comparison with its exact resonant value (54).

It is easy to show that, at a given value of the detuning (satisfying the inequality (55)), the interval $\delta\varphi$ will be the largest for the angles $\varphi_0 = \pm\pi/2$. For these values we have

$$\sin^2\frac{\delta\varphi}{2} = \frac{|\delta\omega|}{2\omega_p}. \quad (56)$$

In accordance with (54) and (55), the equality (56) means that in the case of frequency modulation of the type

$$\omega(t) = \omega_n \pm \omega_p \sin\omega_1 t$$

some of the nuclear spins, for which φ_0 lies in the angle interval $\delta\varphi$ satisfying the inequality

$$\left|\sin\frac{\delta\varphi}{2}\right| < \left(\frac{\omega_1}{2\omega_p}\right)^{1/2} \quad (57)$$

(near $\varphi_0 = \pm\pi/2$), are approximately at resonance with the resonance-frequency field over the entire trajectory of its motion even at $\omega_1 \ll \omega_p$. At $\epsilon \sim 0.1$, for example, approximately 10% of all the nuclear spins satisfy the condition (57), and consequently contribute to the echo-signal intensity. For these spins, the duration of the 180° pulse is, as usual,

$$t_2 = \pi / \omega_1.$$

As noted above, the foregoing estimates are valid only if the inhomogeneous broadening is $\delta\Omega \ll \omega_1$. When $\delta\Omega > \omega_1$ a contribution to the intensity of the echo signal is made only that fraction of the spins for which the inhomogeneous broadening is smaller than ω_1.

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1) We make here the important assumption that the magnetic sublattices are strictly equivalent.

2) I.e., after a transformation of the type

$$m_{ix_l} = m_{ix_l'} \cos \omega t - m_{iy_l'} \sin \omega t,$$
$$m_{iy_l} = m_{iy_l'} \cos \omega t + m_{ix_l'} \sin \omega t$$

(we then omit the primes).

3) It must be borne in mind, however, that actually there is always a certain spread $\delta\Omega$ of the NMR frequency for different sections of the sample. We can therefore speak of aperiodic motion only in a time interval not larger than $\delta t = (\delta\Omega)^{-1}$.

4) The formula is valid, accurate to 10%, up to the value $\beta = 1$.

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34