Chaotic dynamics in NMR

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The dynamics of nuclear magnetization in NMR with a nonlinearity consisting of a dynamic shift of the precession frequency is analyzed. The conditions for the transition from regular behavior to chaotic behavior are studied analytically and numerically. The effect of nonuniform broadening on the nonlinear dynamics of nuclear spins is studied. The physical consequences of chaotic dynamics in NMR and the conditions for observing them experimentally are discussed.

1. INTRODUCTION

Studies of chaotic dynamics[1,2] have been appearing with increasing frequency in problems of the physics of condensed matter in recent years.[3–16] Several basic directions of the research in this field can be distinguished somewhat arbitrarily: chaotic-attractor regimes in a spin-wave turbulence,[3,4] the dynamics of NMR masers,[5,6] the chaotic dynamics of spin clusters,[7–9] and classical and quantum chaos in models of nonlinear spin systems when periodic external fields are applied.[10–13] Research on dynamic chaos in nonlinear NMR can also be classified in the last of these directions.[14–16] Such studies deal with one of the simplest and comparatively well-studied types of nonlinearities in NMR: the dynamic shift of the precession frequency of nuclear spins,[17] which is proportional to the longitudinal component of the nuclear magnetization. The dynamic frequency shift of NMR is associated with a electron-nucleus hyperfine interaction and arises in magnetically ordered crystals (ferro- and antiferromagnets) at low temperatures.[17]

That chaotic dynamics might arise in nonliner NMR was first pointed out by Buishvili and Ugulava,[14] who studied the dynamics of the interaction of nuclear spins with a periodic train of short, small-area rf pulses. The dynamic NMR frequency shift was taken into account. Their estimates[14] show that chaotic dynamics can arise in this system if the product of the NMR nonlinearity constant (the parameter of the dynamic frequency shift) and the pulse repetition period is much larger than the reciprocal of the area under the pulse. Ugulava[16] showed that a weak longitudinal sinusoidal field near the separatrix corresponding to an aperiodic motion of the magnetization in nonliner NMR could give rise to a narrow stochastic layer. In those studies, the nonlinear dynamics of the nuclear magnetization was studied over times shorter than the nuclear relaxation times, $t \ll T_{1,2}$ (the Hamiltonian approximation).

In the present paper we use the Hamiltonian approximation to study the nonlinear dynamics of the nuclear magnetization for NMR with a dynamic-frequency-shift nonlinearity. The envelope of the oscillatory magnetic field acting on the nuclear spins is given as a periodic train of pulses of arbitrary area. This formulation of the problem arises in a study of various transient effects in NMR, e.g., the attenuation of free induction and the spin echo.[17]

Our basic goals in this study were to determine the conditions for a transition from regular behavior to chaotic behavior for various nonlinearity levels and for various values of the area under the pulses and to study the effect of nonuniform broadening on the chaotic dynamics.

The results below show that a transition to chaos in this system can occur by a more complicated path than would follow from the simple estimates of Refs. 14 and 15. In particular, there are several order-chaos-order transitions, and regularity windows form as the nonlinearity or the area under the pulses is varied. We will see that at small pulse areas the chaotic regime of motion has two time scales: a fast one (global chaos) and a slow one (weak chaos and intermittency). We will also show that the regularity windows are metastable because of the presence of weak chaos.

The effect of nonuniform broadening on the chaotic dynamics in NMR is studied here for the first time. It is shown that under certain conditions nonuniform broadening can smear the threshold for the transition to chaos and intensify the stochastic behavior. We discuss the basic physical consequences of chaotic dynamics in NMR, taking into account nonuniform broadening. We estimate the values of the physical parameters which would be required for an experimental observation of these consequences.

2. BASIC EQUATIONS

We will start with the equations of motion of nuclear magnetization in magnetically ordered materials under conditions such that there is a large dynamic NMR frequency shift as a result of an indirect interaction of nuclear spins through electron spins.[17] We choose a coordinate system whose z axis runs along the magnetic field **H** which is acting on the nuclear spins (**H** is the sum of the static external field $\mathbf{H}_0$ and the hyperfine internal field $\mathbf{H}_n$, which is produced by the magnetic moments of electrons at nuclei). The x axis of the rotating coordinate systems runs antiparallel to the oscillatory external field $H_+ = H_1(t)\exp(-i\omega t)$. The equations of motion in the rotating coordinate system are[17]

$$\begin{aligned} \dot{u} &= v(\Delta-\omega_p m), \\ \dot{v} &= -u(\Delta-\omega_p m)+\omega_1(t)m, \\ \dot{m} &= -\omega_1(t)v, \end{aligned} \tag{1}$$

where $\Delta = \omega_n - \omega$, $\omega_n = \gamma(H_n - H_0)$ is the NMR frequency in the absence of a dynamic frequency shift (γ is the gyromagnetic ratio for a nucleus); ω_p is a nonlinearity parameter ($\omega_p \ll \omega$), specifically, the maximum dynamic frequency shift at the equilibrium value of the magnetization; $\omega_1(t) = \gamma A \chi H_1(t)$; A is the dimensionless hyperfine-interaction constant; and χ is the static transverse susceptibility of the electron magnetic system. The variables u, v, m are components of the vector nuclear magnetization (normalized to the equilibrium value of the magnetization) in the rotating coordinate system. Equations (1) hold when irre-

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versible relaxation effects are ignored and in the case of spatial homogeneity (there is no nonuniform broadening). We assume that the envelope of the oscillatory magnetic field $H_1(t)$ is a periodic train of square pulses of height h in which the length of an individual pulse is T_0, and the pulse repetition period is T. Ignoring the effect of the dynamic frequency shift and the frequency deviation Δ during the pulse, we can rewrite Eqs. (1) as a mapping:

$$\begin{aligned} u_{n+1}&=u_n\cos\varphi_n+\tilde{v}_n\sin\varphi_n,\\ v_{n+1}&=\tilde{v}_n\cos\varphi_n-u_n\sin\varphi_n,\\ m_{n+1}&=\tilde{m}_n, \end{aligned} \tag{2}$$

$$\begin{aligned} \tilde{v}_n&=v_n\cos\alpha+m_n\sin\alpha,\\ \tilde{m}_n&=m_n\cos\alpha-v_n\sin\alpha,\\ u_n^2&+v_n^2+m_n^2=1,\\ \varphi_n&=\Delta T-\omega_p T\tilde{m}_n,\quad \alpha=\varepsilon T_0, \end{aligned} \tag{3}$$

where u_n, v_n, m_n are the components of the nuclear spin just before the application of the nth pulse, and $\varepsilon=\gamma A\chi h$. It can be shown that the transformations (2) and (3) conserve phase volume:

$$|\partial(u_{n+1},\ v_{n+1},\ m_{n+1})/\partial(u_n,\ v_n,\ m_n)|=1.$$

The mapping (2), (3) is a combination of two rotations: one through a constant angle α (equal to the area under the pulse) in the vm plane and one through an angle φ_n in the uv plane which depends on the value of $\tilde{m}_n$. Mappings of the form (2) were also found in Ref. 13, in an analysis of the interaction of a quantum top with a periodic external field.[11] The nonlinear dynamics generated by this mapping was not analyzed in Ref. 13, however.

What are the conditions for the applicability of mapping (2)? The effect of the dynamic frequency shift and of the frequency deviation Δ can be ignored during the pulse T_0 if $\omega_p,\ \Delta\ll\varepsilon,\ T_0^{-1}$. The condition for the applicability of the mappings (2), (3) take a simpler form, namely,

$$\alpha^3\ll\varepsilon/\omega_p, \tag{4}$$

in the case $\Delta=\omega_p$, $\alpha\ll 1$, $u(0)=v(0)=0$, $m(0)=1$ (these conditions are typical of most experiments). To demonstrate the validity of (4), we note that Eqs. (1) tell us that the dynamic frequency shift can be ignored under the condition

$$m_\perp\ll\frac{m}{1-m}\frac{\varepsilon}{\omega_p},$$

where $m_\perp=(u^2+v^2)^{1/2}$. For $\alpha\ll 1$ it follows from (2) that we have $m_\perp\sim\alpha$, $1-m\sim\alpha^2$, and $m\sim 1$. Putting these relations together, we find (4).

3. CHAOTIC DYNAMICS IN NMR

Let us analyze the nonlinear dynamics generated by the mapping (2). We introduce the new variables I_n, θ_n:

$$u_n=I_n^{1/2}\cos\theta_n,\quad v_n=-I_n^{1/2}\sin\theta_n.$$

In terms of these variables, relations (2) and (3) can be rewritten as follows:

$$\begin{aligned} \theta_{n+1}&=\operatorname{arctg}\left[\cos\alpha\operatorname{tg}\theta_n+\frac{m_n\sin\alpha}{I_n^{1/2}\sin\theta_n}\right]+\varphi_n(\theta_n,I_n),\\ I_{n+1}&=I_n+(\cos^2\alpha-1)I_n\sin^2\theta_n\\ &\quad-I_n^{1/2}m_n\sin(2\alpha)\sin\theta_n+m_n^2\sin^2\alpha\\ m_{n+1}&=m_n\cos\alpha+I_n^{1/2}\sin\theta_n\sin\alpha. \end{aligned} \tag{5}$$

We will restrict the discussion below to initial conditions under which the nuclear magnetization is close to the equilibrium position, $m_0\approx 1$, $I_0\ll 1$. In the other limit ($m_0\to 0$, $I_0\to 1$ with $\alpha\ll 1$), a mapping as in (2) can be approximated well by a standard mapping.[10,13] The condition derived by the phase-stretching model[1] for a transition to global chaos is

$$K_n=|d\varphi_{n+1}/d\varphi_n|=\omega_p T|u_n|\sin\alpha>1.$$

As a condition for stochastic behavior we can adopt $K_1>1$. This condition means that the phase θ is in a stochastic regime of motion after the very first pulse, i.e.,

$$K\equiv K_1=\omega_p T\sin\alpha|u_0\cos\varphi+(v_0\cos\alpha+m_0\sin\alpha)\sin\varphi|>1,$$

$$\varphi\equiv\varphi_0=\Delta T-\omega_p T(m_0\cos\alpha-v_0\sin\alpha). \tag{6}$$

In particular, with $u_0=v_0=0$, $m_0=1$, and $\alpha\ll 1$, condition (6) becomes

$$K=K_0|\sin\varphi|>1,\quad K_0=\omega_p T\alpha^2,\quad \varphi=(\Delta-\omega_p)T+K_0/2. \tag{7}$$

The condition $K_0>1$ was used in Ref. 14 as a characteristic condition for a transition to chaos. For $K<1$, the nonlinear oscillations of the nuclear magnetization are regular [Fig. 1(a)], and their spectrum contains several harmonics [Fig. 2(a)]. For $K>1$, the magnetization oscillations are chaotic [Fig. 1(b)], and the Fourier spectrum is broadened by an amount $\sim\omega_p$ [Fig. 2(b)]. An interesting aspect of the global-chaos condition (6), (7) is the nonlinear (oscillatory) dependence on the dimensionless parameters of the nonlinearity, $\omega_p T$, and the perturbation, α. As a consequence, even under the condition $\omega_p T\sin\alpha>1$ there should be no chaotic motion for

$$u_0\cos\varphi-(v_0\cos\alpha+m_0\sin\alpha)\sin\varphi\approx 0, \tag{8}$$

where φ is given by (6). Condition (8), for the formation of regularity windows, is determined by both the initial conditions u_0, v_0, m_0 and the dependence on the area α under the pulse and the nonlinearity parameter $\omega_p T$. In the particular case $u_0=v_0=0$, $m_0=1$, $\alpha\ll 1$ condition (8) becomes

$$\varphi=l\pi,\quad l=1,\ 2,\ 3,\ldots, \tag{9}$$

where φ is given by (7). Equation (9) determines the center of a region whose width at the base is $\varphi\sim 1/K_0$ [see (7)].

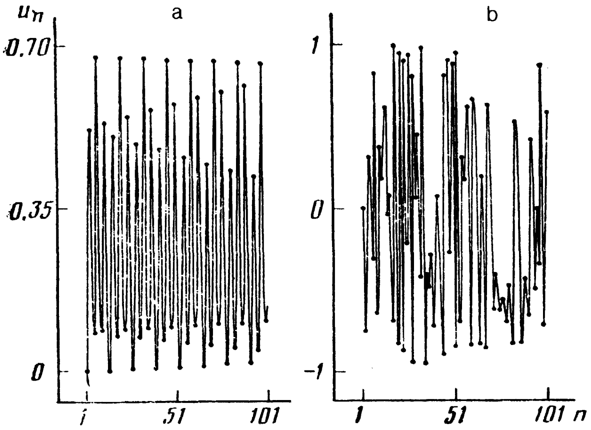

FIG. 1. Nonlinear dynamics of nuclear magnetization in the case $\Delta=\omega_p$, $u_0=v_0=0$, $m_0=1$. a—Regular oscillations ($\omega_p T=5.5$; $\alpha=0.15\pi$), b—chaotic oscillations ($\omega_p T=4$; $\alpha=0.5\pi$).

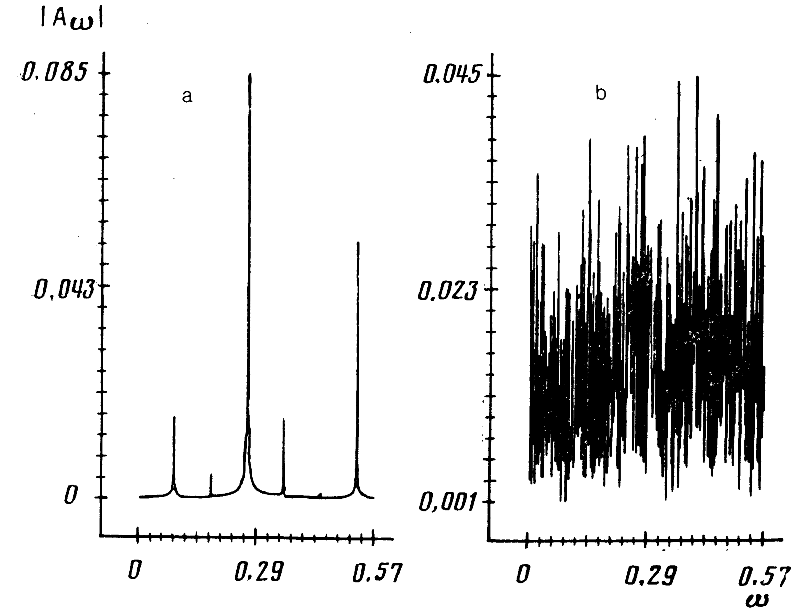

FIG. 2. Fourier spectra for (a) regular and (b) chaotic motion of the magnetization according to calculations for the processes in Figs. 1(a) and 1(b), respectively.

The existence of regularity windows is well known in dissipative systems.[2] They were recently studied numerically in a continuous Hamiltonian system.[18] An analysis[12] of the behavior as a function of the parameters of fixed points and of periodic trajectories has shown that regularity windows can exist in a mapping which arises when a train of δ-function pulses is applied to a spin system.

As the dimensionless nonlinearity $\omega_p T$ or the area α under the pulses varies, there can be several order-chaos-order transitions in the dynamic system (2). Such transitions have indeed been observed in numerical simulations (Figs. 3 and 4).

The picture drawn of the behavior of the mapping (2) as a function of the parameters by the phase-stretching condition (6), (7) is not complete, however. Under certain conditions, chaos can also exist in the case $K<1$ ($\alpha \ll 1$, $\omega_p T > 1$). Let us examine the mapping (5) in the limit $\alpha \ll 1$:

$$\begin{aligned} \theta_{n+1} &= \theta_n + \varphi_n, \\ I_{n+1} &= I_n - 2\alpha m_n I_n^{1/2} \sin\theta_n + \alpha^2 m_n, \\ m_{n+1} &= m_n + \alpha I_n^{1/2} \sin\theta_n - \tfrac{1}{2}\alpha^2 m_n, \end{aligned} \qquad (10)$$

$$\varphi_n = \Delta T - \omega_p T [m_n(1-\alpha^2/2) + \alpha I_n^{1/2} \sin\theta_n + O(\alpha^3)]. \qquad (11)$$

It follows from (10) and (11) that θ is a fast variable, while I and m are slow ($|\delta\theta/\delta I| \sim |\delta\theta/\delta m| \sim K/\alpha^2 \sim \omega_p T > 1$). In the case $K<1$, the change in the phase between pulses, (11), is small, $\varphi_n \ll 1$, at long times. The result is an adiabatic change in the action I and in m. The motion of the nuclear magnetization is regular in this case. Because of the slow variation of m and I, however, there can be a phase buildup, and for a certain time we may have $\varphi_n \sim 2\pi$. In this case the motion is no longer adiabatic; abrupt changes in the phase lead to the onset of a stochastic behavior and to growth in I (a decrease in m) on the average. The dynamics of the mag-

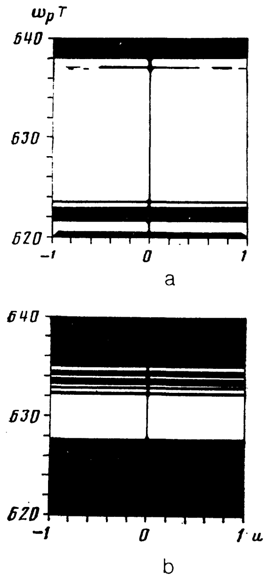

FIG. 3. Regularity window in the case $\Delta = \omega_p$, $u_0 = v_0 = 0$, $m_0 = 1$, $\alpha = 0$. a—The length of the trajectories is $N = 10^3$ iterations; b—3×10^3.

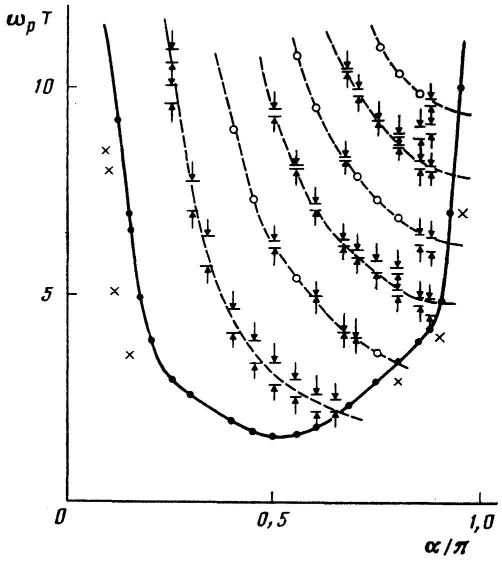

FIG. 4. Dependence of the regime of motion (regular or chaotic) on the dimensionless nonlinearity $\omega_p T$ and on the perturbation α. These results were found in calculations for $N = 10^3$ iterations.

TABLE I.

α	$\omega_p T$	$(\omega_p T)_{cr}$	α	$\omega_p T$	$(\omega_p T)_{cr}$
0,03	117÷125	216	$0,125\pi$	5,2	9,3
0,1	72,8÷82,4	139	0.1275π	4.9÷5,0	9
0,12	52÷56	97	$0,1333\pi$	4,5÷4,6	8,2
$0,1\pi$	7,8÷8	14,5			

netization is thus very nonuniform: Over a comparatively brief time interval ($\lesssim 10$ iterations) the motion can be fast and random, the trajectory will then "get stuck" for a long time ($\sim 10^2$ iterations), a fast motion will then resume quickly, etc. (an intermittency regime of motion).

The possible occurrence of chaos when the adiabatic-motion conditions are violated has also been demonstrated recently for measure-preserving mappings which arise in problems in plasma physics[19] and hydrodynamics.[20]

Table I shows values of the dimensionless nonlinearity parameter $\omega_p T$ for which, at the given value of α, we observed a slow stochastic motion at $K_0 < 1$ in the numerical calculations. The critical value of the nonlinearity, $(\omega_p T)_{cr}$, which corresponds to the boundary of the transition to global chaos ($K \approx 1$), is given in the third column of this table. For all values in this table we have $u_0 = v_0 = 0$, $m_0 = 1$, and $\Delta = \omega_p$, and the computation time is $N = 10^3$–10^4 iterations. The boundaries of the regions with weak chaos shown in this table (in the second column) are approximate. These boundaries actually have a complex fine structure because of the pronounced sensitivity of the regime of weak chaos to even slight changes in the parameter values (see also Sec. 4 and Fig. 7). The mechanism for the onset of chaos described here in the case $K_0 < 1$ may also operate at comparatively large values $\alpha \lesssim 0.2\pi$ (see also Fig. 4).

The existence of a slow onset of chaos in the case $K < 1$ results in gradual destruction of the regularity windows. Figure 3 illustrates this destruction, for regularity windows with $l = 1$ and at the parameter values $\alpha = 0.1$, $\Delta = \omega_p$, $u_0 = v_0 = 0$, and $m_0 = 1$. Inside a regularity window there exists an additional integral of motion (of the adiabatic-invariant type), $u(t = nT) = 0$. As time elapses this integral is destroyed stochastically. The boundary of a regularity window is a complex hierarchical structure which consists of narrow alternating regions of stochastic and regular motion. The window in Fig. 3 is destroyed completely over times corresponding to $N \sim 5 \times 10^3$ iterations. At the same values of the parameters, the regularity window with $l = 2$ is essentially unobservable, while the narrow window with $l = 3$ is destroyed completely over a time corresponding to $N \sim 1.5 \times 10^3$.

Figure 4 shows the regime of motion (regular or chaotic) as a function of the parameters $\omega_p T$ and α (in the region $\alpha \sim 1$). The solid curve in Fig. 4 separates regions of regular behavior and global chaos in the plane of the variables $\omega_p T$ and α. The crosses show the positions of some separate small regions of chaotic behavior below the boundary of global chaos. The arrows show regularity windows, while the symbol O means that the given window is essentially indistinguishable. The dashed lines show the positions of the centers of regularity windows as calculated from (9). We see from Fig. 4 that near $\alpha \sim \pi/2$ the regularity windows are slightly distorted in comparison with the estimates which follow from (7) and (9) [the first window ($l = 1$) agrees to within a few percent with the theoretical estimate with $\alpha = \pi/2$]. As $\alpha \to \pi$ and for $\alpha \ll 1$, the widths of all the windows seriously depart from their estimates according to (7). The even-numbered windows are usually distorted to a much greater extent than the odd-numbered windows. At $\alpha \sim 1$, the lower windows are destroyed very slowly. The window with $l = 1$ in the case $\alpha = 0.3\pi$ ($\Delta = \omega_p$, $u_0 = v_0 = 0$, $m_0 = 1$), for example, is covered completely by "stochastic blinds" over times corresponding to $N \sim 10^6$ iterations.

These results on the position and width of the regularity windows agree well with data on the local stability (or instability) of the motion: At parameter values corresponding to global chaos we always observe local instability, while within a metastable window the local instability does not occur during the "stability time." In these numerical simulations, the transition to chaos is observed only through intermittency. If the conditions for stochastic instability, (7), or the conditions for the onset of weak chaos are satisfied, the magnetization vector will deviate from its equilibrium position on the average. A stochastic excitation of this sort could apparently be observed experimentally by comparing the values of the longitudinal magnetization in the regular and chaotic regimes of motion. Although the stochastic excitation in the case $K < 1$ is quite slow, it is important to keep the existence of such chaotic regimes in mind. The presence of even a slight external noise could accelerate the diffusion in the way that an external noise accelerates slow Arnol'd diffusion in multidimensional Hamiltonian systems.[2] The regime of weak chaos ($K_0 < 1$) also plays an important role in stochastic excitation when nonuniform broadening is taken into account.

4. EFFECT OF NONUNIFORM BROADENING ON NONLINEAR DYNAMICS IN NMR

Nonuniform broadening has an important effect on the behavior of NMR with a dynamic frequency shift in most experimental situations.[17,21] The basic physical reason for nonuniform broadening is generally a variation of the susceptibility χ, which causes a spread in the values of the dynamic-frequency-shift parameter, $\omega_p = \overline{\omega}_p + \delta$, where $\overline{\omega}_p$ is the average value of ω_p (Ref. 21). Observable quantities are the average values

$$\langle B \rangle = \int_{-\infty}^{+\infty} g(\delta) B(\overline{\omega}_p, \delta)\, d\delta, \qquad (12)$$

where B is one of the functions $u(\delta)$, $v(\delta)$, $m(\delta)$; and $g(\delta)$ is a distribution function. We assume below that it is Gaussian with a standard deviation σ:

$$g(\delta)=(2\pi\sigma^2)^{-1/2}\exp(-{}^1/_2\delta^2/\sigma^2).$$

Typical values of the relative standard deviation are $\sigma/\omega_p \sim 10^{-2}-10^{-1}$

How does nonuniform broadening affect the conditions for global chaos? The condition for a transition to chaos for an individual isochromate with a dynamic-frequency-shift parameter $\omega_p+\delta(-\infty<\delta<+\infty)$ is

$$K_\delta>1,\ K_\delta=|(\overline{\omega}_pT+\delta T)\sin\alpha[u_0\cos\varphi_\delta+(v_0\cos\alpha+m_0\sin\alpha)\sin\varphi_\delta]|, \tag{13}$$

$$\varphi_\delta=\Delta T-(\omega_pT+\delta T)(m_0\cos\alpha-v_0\sin\alpha). \tag{14}$$

Since isochromates with $|\delta|$ less than or on the order of σ and with $\sigma\ll\overline{\omega}_p$ dominate the calculation of the averages in (12), the primary distinction between (6) and the condition for chaos in the case of nonuniform broadening is in the behavior of the terms containing $\sin\varphi_\delta$ and $\cos\varphi_\delta$. The condition under which nonuniform broadening has only a slight effect on the condition for transition to global chaos is

$$|\varphi_\delta-\varphi|=\sigma T\cos\alpha\ll\pi, \tag{15}$$

$$\left|\frac{\varphi_\delta-\varphi}{\varphi}\right|=\left|\frac{\sigma T\cos\alpha}{\Delta T-\omega_pT\cos\alpha}\right|\ll1 \quad (u_0=v_0=0,\ m_0=1). \tag{16}$$

If conditions (15) and (16) are violated, many isochromates with $K_\delta>1$ may contribute to the calculation of the averages in (12). Let us examine some particular cases.

1. For $\alpha\sim\pi/2$, with arbitrary Δ, inequalities (15) and (16) essentially always hold, and the nonuniform broadening has only a slight effect on the nonlinear dynamics of the system.

2. Under the conditions $\Delta=\overline{\omega}_p$, $\alpha\ll1$, relation (16) becomes the inequality

$$2\sigma/\overline{\omega}_p\alpha^2\ll1. \tag{17}$$

Under the condition $\sigma T>1$, conditions (15)–(17) are violated, with the possible result that the regularity windows disappear in an analysis of the dynamics of the averages in (12).

How does the nonuniform broadening affect the nonlinear dynamics of the averages in (12) under the conditions $K<1$, $\alpha\ll1$? In this case, an individual isochromate may acquire chaotic dynamics if the adiabatic condition $\varphi_n(\delta)\ll1$ is violated, where $\varphi_n(\delta)$ is found from (11) and the substitution $\omega_p\to\overline{\omega}_p+\delta$. Comparing (11) for $\varphi_n(\delta=0)$ and $\varphi_n(\delta)$ for $m\sim1$, we find that the nonuniform broadening causes essentially no change in the nonlinear dynamics in the case $K_\delta<1$ if inequalities (15) and (16) hold. In the opposite case, in which these inequalities do not hold [in particular, under the condition $\varphi\ll1$, condition (17) may be violated even in the case $\sigma T\ll1$], the integral in (12) receives contributions from many isochromates with chaotic dynamics, even if $K<1$. The result may be enhancement of the stochastic excitation.

To test these estimates we carried out some numerical calculations with 2.4×10^3 isochromates (Figs. 5–8). A further increase in the number of isochromates has essentially no effect on the results over the times studied. It can be seen from Fig. 5 that a violation of conditions (15)–(17) has the consequences that the stochastic excitation at the values $\alpha=0.1$ and $\overline{\omega}_pT=628$ (curve 1; these values correspond to

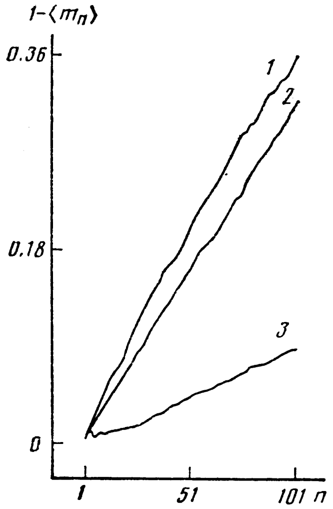

FIG. 5. Stochastic excitation of nuclear magnetization in the case $\Delta=\overline{\omega}_p$, $u_0=v_0=0$, $m_0=1$, $\alpha=0.1$, $\sigma/\overline{\omega}_p=0.1$. An average was calculated in accordance with (12) with 2.4×10^3 isochromates. 1—$\omega_pT=628$; 2—314; 3—90.

the center of a regularity window in the case without nonuniform broadening) is even faster than at the value $\overline{\omega}_pT=314$ (curve 2), which corresponds to the fastest diffusion in the uniform case. The stochastic excitation at the value $\overline{\omega}_pT=90$, which lies below the threshold for global chaos [in the case at hand, $(\omega_pT)_{cr}\approx139$], is shown by curve 3 in Fig. 5. In this case the stochastic excitation is dominated by the isochromates shown in Fig. 7. As can be seen from this figure, these isochromates have a fine structure. This fine structure exists as a result of the pronounced sensitivity to changes in the parameter values in the weak-chaos regime. Figure 6 illustrates stochastic excitation at small values of K_0. In this case the diffusive excitation is nonuniform.

In summary, if inequalities (15) and (16) do not hold, nonuniform broadening spread out the threshold for the transition to chaos. With increasing value of the nonlinearity parameter $\overline{\omega}_pT$, the stochastic excitation is accelerated. Slow excitation may occur even at fairly small values of $\overline{\omega}_pT$ $(K_0\ll1)$.

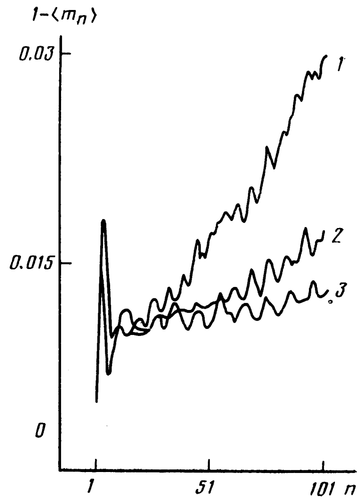

FIG. 6. The same as in Fig. 5, but for small values of K_0. 1—$\omega_pT=60$; 2—50; 3—30.

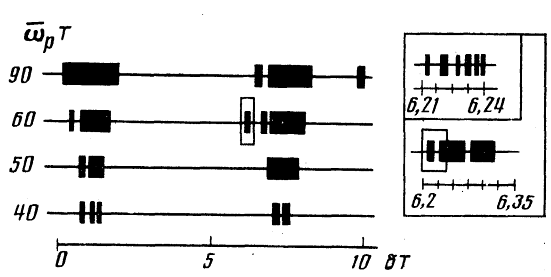

FIG. 7. Isochromates which lie below the boundary of global chaos $[(\omega_p T)_{cr} \approx 139]$ and which exhibit chaotic dynamics in the case $\alpha = 0.1$, $\Delta = \omega_p$, $u_0 = v_0 = 0$, $m_0 = 1$, $N = 10^3$. The inset shows the fine structure of the isochromates with chaos which is manifested as the resolution in terms of the parameter δT increases.

We now consider the nonlinear dynamics of the nuclear magnetization in the case $\alpha \sim 1$ with nonuniform broadening [Figs. 8(a) and 8(b)]. It follows from this figure that the motion of the magnetization is different from the parameter values which correspond to global chaos (solid line) and those which correspond to a regularity window (dashed line). The rapid damping (over a few iterations) of the oscillations of the longitudinal component [Fig. 8(a)] and the transverse component [Fig. 8(b)] of the nuclear magnetization might be called "stochastic saturation," since it is caused by a rapid splitting of the correlations in the motion of individual isochromates in a regime of dynamic chaos. The stochastic saturation described here occurs in the absence of any irreversible relaxation. This point distinguishes the case at hand from the stochastic saturation described in Ref. 15, which can occur when diffusion is canceled by relaxation of the longitudinal magnetization component.

5. CONCLUSION

We have examined the transition to chaos in nonlinear NMR in magnetically ordered media. Depending on the area α under the existing pulse and also on the dimensionless nonlinearity parameter $\omega_p T$, regularity windows may form, and there may be several order-chaos-order transitions. At small values of α the regularity windows become covered by stochastic blinds in all cases (although the process is usually slow), so these windows are metastable. The occurrence of nonuniform broadening may also, under certain conditions, close the regularity windows and intensify the stochastic behavior. In addition to the stochastic excitation, the broadening of the Fourier spectrum of the NMR signal by an amount on the order of ω_p and the stochastic saturation which is seen at $\alpha \sim 1$, as a result of the combined effects of nonuniform broadening and dynamic chaos, can be classified as manifestations of chaotic dynamics in NMR with a dynamic frequency shift.

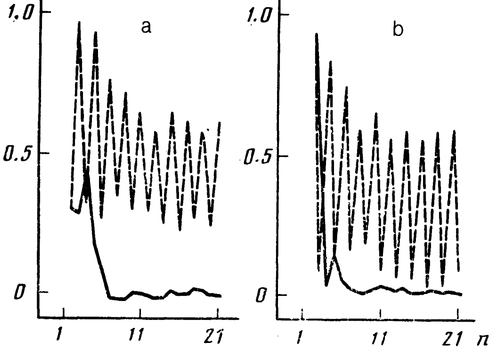

FIG. 8. Nonlinear dynamics of (a) the longitudinal component $m_\parallel$ and (b) the transverse component $m_\perp = [u_2 + v_2]^{1/2}$ of the nuclear magnetization in the case with nonuniform broadening with the values $\alpha = 0.4\pi$, $\Delta = \omega_p$, $\sigma/\bar{\omega}_p = 0.1$, $u_0 = v_0 = 0$, $m_0 = 1$. Solid line—$\bar{\omega}_p T = 6.5$; dashed line—4.45. The average was taken over 2.4×10^3 isochromates.

These effects could apparently be observed experimentally in ferro- and antiferromagnets at liquid-helium temperatures at the following values of the physical parameters: $T_2 \sim 10^{-4}$–10^{-3} s, $T_1 \sim 10^{-3}$–10^{-1} s, $T \sim 10^{-6}$–10^{-4} s, $\varepsilon \sim 10^6$ Hz, $\omega_p \sim 10^5$–10^7 Hz, $T_0 \sim 10^{-7}$–10^{-6} s, and $\alpha \sim 0.1$–1.

We propose the following experiment for observing stochastic excitation: At a time t ($T_2 \lesssim t < T_1$) after N short pulses have been applied ($N \sim 5$–10), yet another pulse is applied, and the free-induction signal is observed. The amplitude of this signal depends on the longitudinal component of the magnetization, which in turn has different values in the regular and chaotic regimes of motion during the application of the N pulses.

We conclude with a few comments.

1. Preliminary numerical experiments have shown that the chaotic dynamics persists even when irreversible relaxation is taken into account, up to $T/T_2 \approx 1$ in the case $\alpha \ll 1$ and up to $T/T_2 \approx 0.1$ in the case $\alpha \sim 1$ (under the assumption $T_1 \gg T_2$). It would be interesting to study the possible existence of a steady-state chaotic regime of motion of the strange-attractor type in this problem.

2. The new effects which we have described here (the existence of metastable regularity windows and the existence of stochastic saturation in the case of nonuniform broadening) appear to be quite general. For example, analogs of stochastic saturation should probably be observed in nonlinear systems which can have chaotic dynamics when strongly perturbed (a strong perturbation would cause a trajectory to cover a large part of the phase space) and in the presence of nonuniform broadening (the spread could be in any parameter of the system). In this connection it would be interesting to search for and study effects of this sort in other physical systems.

3. The situation studied in the present paper is very close to that which arises in research on spin echoes, as we have already mentioned. Preliminary numerical calculations have shown that chaotic dynamics of isochromates can affect the behavior of spin-echo signals.

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