Chaotic regime in nonlinear NMR

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The conditions under which the response of a nuclear spin system in an antiferromagnet to the application of periodic pulses becomes random are studied. A phase diagram constructed through a one-dimensional mapping which models the spin dynamics contains regions of periodic and random motion.

Several problems of NMR in magnetic materials with a large dynamic frequency shift are related to questions of stability. In particular, the conditions for the onset of stochastic dynamic regimes of pulsed NMR were analyzed in the dissipationless limit in Refs. 1. In the present letter we take up a qualitatively different situation, in which dissipation cannot be ignored. We examine the nonlinear dynamics of an acoustic nuclear mode in easy-plane and cubic antiferromagnets as they are excited by a steady-state periodic train of square rf pulses.

1. Let us consider the case of transverse pumping, in which the gain values reach their maximum, $\eta = H_n/|\vec{H}|$, where H_n is the hyperfine field at the nucleus. The static magnetic field $\vec{H}$, which determines the equilibrium orientation of the magnetic moments of the sublattices, and the rf field $\vec{H}_1(t) = 2\vec{h}\cos\omega t$, which has an amplitude $2|\vec{h}| \ll |\vec{H}|$ and a frequency ω and which is perpendicular to the static field, both lie in a plane with a small magnetic anisotropy. The equations of motion of the nuclear magnetization $\vec{m}$ of a sublattice take their simplest form in the rotating proper coordinate system[2]

$$\begin{aligned} \dot{m}_x &= -(\omega - \omega_n^0 + \omega_p m_z/m_0)m_y - (m_x - \chi\eta h)/T_2, \\ \dot{m}_y &= (\omega - \omega_n^0 + \omega_p m_z/m_0)m_x + \omega_1 m_z - m_y/T_2, \\ \dot{m}_z &= -\omega_1 m_y - (m_z - m_0)/T_1, \end{aligned} \qquad (1)$$

where $\omega_n^0 \simeq \gamma H_n$ is the unshifted NMR frequency, ω_p is the equilibrium value of the dynamic frequency shift, $\omega_1 = \gamma\eta h$ is the amplitude of the circularly polarized resonant component of the amplified rf field, expressed in frequency units, χ and $m_0 \simeq \chi H_n$ are the static magnetic susceptibility and the equilibrium magnetization of the nuclei of the sublattice, T_2 and $T_1 \gg T_2$ are the transverse and longitudinal relaxation times, and γ is the nuclear gyromagnetic ratio.

2. Since the inhomogeneous broadening of an NMR line in a real sample is caused primarily by the scatter in the values of ω_p (Ref. 2), we will use a single-parameter family of oscillators to model the spin nuclear system. We impose some restrictions on the pulse length T_i and the pause length T_p, specifically, $T_i \ll T_{1,2}$ and $T_p \gg T_2$. It thus becomes possible to eliminate the variables $m_{x,y}$. In order to avoid dealing with the diffusion of the nuclear spin temperature, which plays out over a time scale T_d, we consider two limiting cases.

a) In the limit of slow diffusion, $T_d \gg T_i$, T_p, T_1, we are dealing with an ensemble of noninteracting oscillators with various values of ω_p. The solution of Eqs. (1) in the conservative approximation is well known.[3] Introducing the new variable $z = m_z/m_0$, and allowing for the relaxation during the pause, we find a one-dimensional mapping which relates the values of z at the leading edges of two successive pulses:

$$z_{n+1} = 1 - a + a z_n \left\{1 - \frac{2\tilde{\omega}_1^2}{\rho(Y; g_2^n, g_3^n) + [\tilde{\omega}_1^2 + (z_n + d)^2]/3}\right\}, \tag{2}$$

where $\rho(Y;\ g_2^n, g_3^n)$ is the Weierstrass function,[4] with the invariants $g_j^n = g_j(z_n)$, $g_3(z) 4\tilde{\omega}_1^4 z^2 - \frac{8}{3}\tilde{\omega}_1^2 z(z+d)[\tilde{\omega}_1^2 + (z+d)^2]^2 + (\frac{2}{3})^3[\tilde{\omega}_1^2 + (z+d)^2]^3$, $g_2(z) = -8\tilde{\omega}_1^2 z(z+d) + \frac{4}{3}[\tilde{\omega}_1^2 + (z+d)^2]^2$; $a = \exp(-T_p/T_1)$, $\tilde{\omega}_1 = \omega_1/\omega_p$, $d = (\omega - \omega_n^0)/\omega_p$, and $Y = \omega_p T_i/2$. For simplicity we restrict the analysis to a relatively slight saturation, $\Delta z = 1 - z \ll 1$, in the approximation $\tilde{\omega}_1 \ll 1$, in which we can approximate (2) by the mapping

$$\zeta_{n+1} = a[\zeta_n - \cos(b\zeta_n + \phi)] \equiv f(\zeta_n), \tag{3}$$

where

$$\zeta = \zeta_1 = \Delta z(1+d)^2/\tilde{\omega}_1^2 - a/(1-a), \quad b = b_1 = \omega_p T_i \tilde{\omega}_1^2/(1+d)^2,$$
$$\phi = \phi = -\omega_p T_i (1+d)[1 - a\tilde{\omega}_1^2/(1-a)(1+d)^3], \tag{4}$$

for large frequency deviations $(\tilde{\omega}_1^2/(z+d)^2 \ll 4|z+d|/27z \ll 1)$ or

$$\zeta = \zeta_2 = \Delta z/\tilde{\omega}_1^{2/3} - a/(1-a), \quad b = b_2 = 3^{1/4}\omega_p T_i \tilde{\omega}_1^{2/3}/6,$$

$$\phi = \phi_2 = b_2[(6-5a)/(1-a) - (1+d)/\tilde{\omega}_1^{2/3}] \tag{5}$$

for small frequency deviations $(|z+d| \ll z^{1\setminus 3}\tilde{\omega}^{2/3}|2 \ll 1)$.

b) In the limit of rapid diffusion, $T_i \ll T_d \ll T_p$, T_1, a uniform nuclear spin temperature becomes established over a time $t \gg T_d$ after the pulse. Mappings for a uniform z are found by taking an average over the scatter in ω_p; they are of the form in (3). In the systems under consideration here, $\omega_p = 2\pi\nu_p$ is far higher than the inhomogeneous linewidth $\Delta\nu$; i.e., we have $\Delta\nu|\nu_p^0 \ll 1$, where ν_p^0 corresponds to the center of the shifted NMR line. Under our approximations, it is important to allow for the inhomogeneity of ω_p only in the argument of the cosine in mapping (3). Under the assumption that the line has a Gaussian equilibrium shape we have

$$\zeta = \zeta_j = \zeta_j \exp(\kappa_j), \quad b = b_j^- = b_j \exp(-\kappa_j). \tag{6}$$

Here $j = 1$, 2 labels the regions of large and small deviations; $(\pi\Delta\nu T_i)^2/4 \ln 2$, $k_2 = k_1/12\times 3^{1/2}$; and the quantity ω_p which appears in ζ_j, b_j corresponds to the center of the equilibrium NMR line (i.e., $\omega_p = \omega_p^0 = 2\pi\nu_p^0$).

3. The dissipation is characterized by the slope $0 < a < 1$ of the envelopes of the function $f(\zeta)$. In the iterations, the imaging point is attracted by a finite region of phase space,

$$|\zeta| \le a/(1-a), \tag{7}$$

in which a balance is struck between the energy absorbed by the spins, which depends on the oscillation period $2\pi/b$ and the phase ϕ of the function $f(\zeta)$, on the one hand, and the energy which the spins give up to the lattice, on the other. The results of a study of the number and stability of 1-cycles of mapping (3) can be represented as a

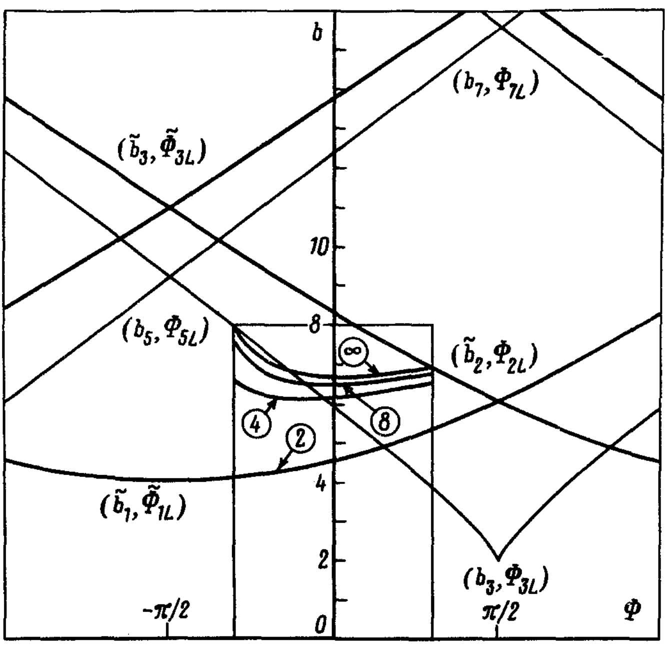

FIG. 1. Elements of the (b, ϕ) phase diagram of mapping (3) with $a = 1/3$. The arrows in the rectangle $0 \le b \le 8$, $-0.9 \le \phi \le 0.9$ show arcs of period-doubling bifurcation curves (according to a numerical simulation). The resultant period is shown in the attached circle.

(b, ϕ) phase diagram with a period of 2π along ϕ. A family of bifurcation curves (the light lines in Fig. 1), defined by $\phi_L(b) = 2\pi(L + 1/4) \pm \Delta\phi(b)$, $\Delta\phi(b) = \sqrt{[(ab/1-a)]^2 - 1} - \arctan\sqrt{[(ab/1-a)]^2-1}$, where $b \geqslant 1/a - 1$, and L is an integer, divides the (b, ϕ) parameter plane into regions in which mapping (3) has $k = 2n + 1$ fixed points ($n = 1, 2, 3,...$). These curves intersect at threshold points with the coordinates $b_k = (1+\varphi_k^2)^{1/2}[1/(a-1)]$, $\phi_{kL} = \pi[4L + (-1)^{(k+1)/2}]/2$, where $\varphi_k < \varphi_{k+2}$ is the set of nonnegative solutions of the equation $\varphi = \tan\varphi$, with $\varphi_3 = 0$. The stability of the 1-cycles changes when the control physical parameters change. As a result, the trajectory in the (b, ϕ) plane intersects the bifurcation curves (the heavy lines in Fig. 1), which are defined by the equations $\tilde{\phi}_L(b) = 2\pi(L - 1/4) \pm \Delta\tilde{\phi}(b)$, $\Delta\tilde{\phi}(b) = 1 + a/1 - a\sqrt{[(ab/1+a)]^2-1} - \arctan\sqrt{[(ab/1+a)]^2-1}$, where $b \leqslant 1/a + 1$, and L is an integer. These curves are the boundaries of the regions which contain $n = 1, 2, 3,...$ unstable 1-cycles on nondecaying regions of $f(\zeta)$. They intersect at the threshold points $\tilde{b}_n = (1+\tilde{\varphi}_n^2)^{1/2}(1/a+1)$, $\tilde{\phi}_{nL} = \pi[4L + (-1)^n]/2$, where $\tilde{\varphi}_n < \tilde{\varphi}_{n+1}$ is the set of nonnegative solutions of the equation $\tilde{\varphi} = -\tan[(1+a/1-a)\tilde{\varphi}]$, where $\tilde{\varphi}_1 = 0$. Under the condition $[(ab/1+a)]^2 \gg 1$, stable 1-cycles can be observed only in narrow intervals at the boundaries of attractive region (7). The width of these intervals, $(1+a)^2/2ab^2(1-a)$, goes through a minimum at $a = 1/3$.

More complex internal bifurcations of limiting sets and combinations thereof have been studied numerically. Shown in the box in Fig. 1 is a portion of the phase diagram for a single oscillator in the limit of slow diffusion, in which a Feigenbaum transition is observed.[5] It was found at $a = 1/3$ for mapping (3), with the equilibrium value $\zeta = -1/2$ as the initial value. Figure 2 shows an example of a bifurcation diagram and the behavior of the Lyapunov exponent λ, found through an increase in the parameter b at $\phi = \text{const} = 0$. In the b regions in which we have $\lambda > 0$, there is a chaos, interrupted by windows of periodicity. In the region $b > 1/a + 1$ the amplitude of the variations in ζ can approach (in magnitude) the width of the attractive region, (7).

4. Analysis reveals the following.

a) In the limit of slow diffusion, oscillators with various attractors coexist as the result of inhomogeneous broadening. The dynamic spread in the values of m_z caused by this situation modifies the shape of an NMR line (ultimately splitting it) and generally makes it time-dependent (or dependent on the number of rf pulses applied). Under the conditions $\tilde{\omega}_1^2, {}_1^{2/3} > \Delta\nu(1-a)/2\nu_p^0 a$, the dynamic spread in the frequencies of the oscillators may be significantly greater than the equilibrium inhomogeneous width at large and small frequency deviations, respectively. In this case the observed shift, the observed width, and the observed shape of the NMR line are of purely dynamic, nonequilibrium nature. From definitions (4) and (5) and from the threshold values of b we can find the amplitudes which the rf fields must have if the effects described above are to appear. Our estimates for $CsMnF_3$, $RbMnF_3$, and $MnCO_3$, on the basis of the data of Refs. 6–9, with pulse lengths $T_i \sim 1$–$10\ \mu s$ and pause lengths $T_p \sim T_1$, yield values on the order of 10^{-1}–10^{-2} Oe.

b) In the limit of rapid diffusion, the width and shape of the NMR line remain

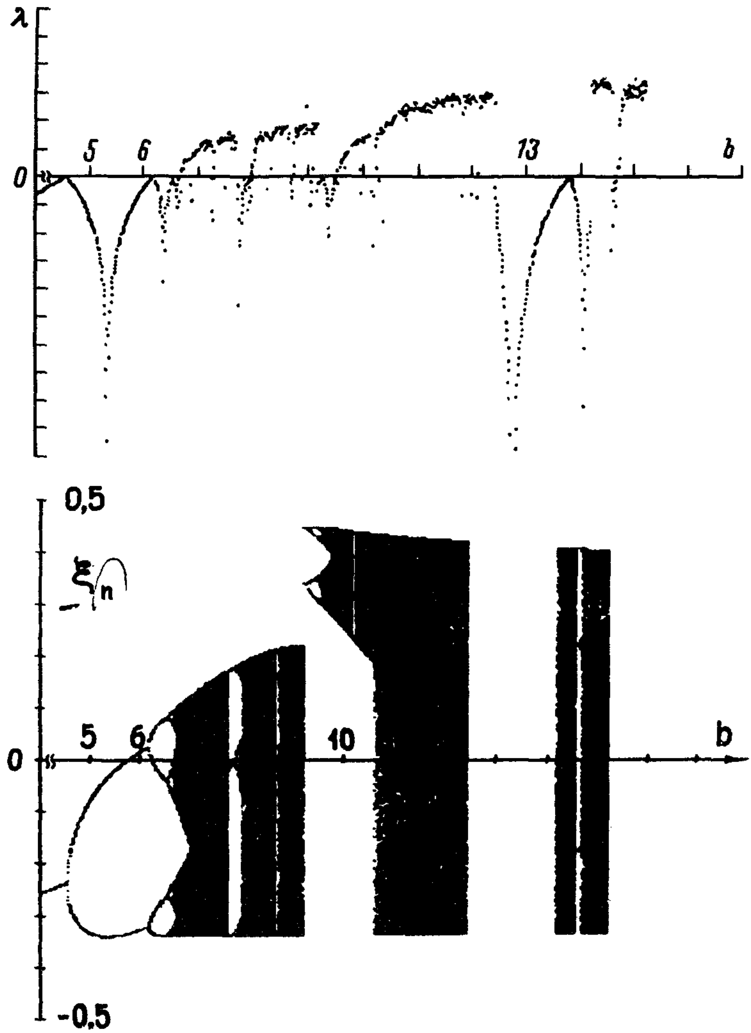

FIG. 2. Top—Lyapunov exponent (in arbitrary units) as a function of the parameter b; bottom—bifurcation diagram. Shown here are 10^3 values of ζ_n for each value of b over the interval $4 \leqslant b \leqslant 15.3$ at steps of 0.02 in the steady state.

the equilibrium width and shape, at the accuracy of this analysis, while the dynamic frequency shift may either be fixed or vary in time, periodically or at random. Since b is a nonmonotonic function of T_i, the efficiency of the excitation by pulses of fixed amplitude reaches a maximum at pulse lengths $T_{i1} \simeq (2 \ln 2)^{1/2}/\pi\Delta\nu$ for large values of the deviation or at $T_{i2} \simeq 6T_{i1}/3^{1/4}$ for small values of the deviation. The corresponding threshold values of the amplitude of the rf pulses, found from $b_1^- \simeq 2(2 \ln 2/e)^{1/2}(\nu_p^0/\Delta\nu)(\omega_1/\omega_p^0)^2$, $b_2^- \simeq b_1^-(\omega_1/\omega_p^0)^{-4/3}$ [see (6)], are of the same order of magnitude as in the limit of slow diffusion.

The spin-echo method[10] might be used to observe the effects described above. In this case, information on the distribution of the longitudinal components of the nuclear magnetization would be embodied in the spectrum of the echo signal. This information could also be found through an analysis of a free-induction signal. In the region in which there is a substantial relative heating of the nuclear spin system, one might use the method proposed by Svistov and Smirnov[11] to signal NMR, on the basis of surges in the microwave power absorption, as proposed in Ref. 11.

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