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Parametrical Instability in Nuclear Spin Systems with Dipole–Dipole Interactions

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It is shown that when the values of the radiofrequency magnetic field amplitude are more than the critical value, quasi-thermodynamical description of the dynamics of a nuclear spin system with dipole–dipole interaction is impossible because of the unlimited growth of a number of parametrically excited magnons. The critical values of the radiofrequency field and the energies of the first excited magnons are calculated. It is important that in contrast to electron spin systems, in nuclear spin systems the probability of excitation of resonant parametric magnons vanishes if detuning is equal to zero, but the magnons from the top or bottom of the spectrum zone are excited. It is mentioned that such an instability can be a reason for NMR chaos. © 1998 Academic Press

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The quasi-thermodynamical theory of nuclear magnetic resonance (NMR) saturation is usually very successful in describing the influence of radiofrequency (RF) magnetic fields on a nuclear spin system (NSS) with dipole–dipole interaction in both high (*1–3*) and ultralow spin temperature (*4, 5*) cases.

In all the works mentioned above the influence of the RF magnetic field was considered a reason for the thermodynamic equilibrium setting in a rotating frame (with a frequency of the oscillating magnetic field). On the other hand, in the articles (*6, 7*), the parametric instability in electron spin systems was investigated, and it was shown that for values of a varying magnetic field greater than the critical value, unlimited growth of a number of parametric excitations (magnons) takes place. Hence in this case the quasi-thermodynamic description of a spin system is impossible.

Moreover, recent publications about NMR transient chaos (*8, 9*) propose that such an instability may cause structural (magnon) chaos under specific conditions.

In the present paper we will investigate the conditions for the emergence of parametric instability in a NSS ($I = \frac{1}{2}$) with dipole–dipole interaction, situated in a strong static magnetic field H_0 directed along the z axis and undergoing the influence of an RF magnetic field (directed normal to the z axis). Then the Hamiltonian of NSS has the form (*1*)

$$\mathcal{H} = -\hbar\omega_0 \sum_i^N I_i^z - \sum_{i,j}^N \epsilon_{ij}(2I_i^z I_j^z - I_i^+ I_j^-) - \hbar\omega_1 \sum_i^N (I_i^+ e^{i\omega t} + I_i^- e^{-i\omega t}) + \mathcal{H}_{\rm ns}, \qquad [1]$$

where $\omega_0 = gH_0$, g is the gyromagnetic ratio for nuclei, N is the number of spins in a sample, $\mathbf{I}_i$ is a spin operator disposed in the ith lattice site, ω_1 and ω are the amplitude (in frequency units) and frequency of the RF magnetic field, respectively, $I_i^{\pm} = I_i^x \pm I_i^y$, and $\mathcal{H}_{\rm ns}$ denotes the nonsecular terms of the Hamiltonian [1],

$$\epsilon_{ij} = -g^2\hbar^2 \frac{1 - 3\cos^2\theta_{ij}}{4|r_i - r_j|^3}. \qquad [2]$$

$\mathbf{r}_i$ and $\mathbf{r}_j$ are radius vectors of spins disposed in the positions i and j, respectively, and θ_{ij} is the angle between the vector $\mathbf{r}_i - \mathbf{r}_j$ and the z axis.

Let us suppose that the RF magnetic field is switched on, the NSS has been in thermodynamic equilibrium with a lattice, and the polarization of NSS has been close to unity ($1 - p \ll 1$, e.g., low spin temperatures are considered). Hence we can use Dyson's spin-wave approximation approach (*10*). Substituting

$$I_i^+ \Rightarrow a_i,\ I_i^- \Rightarrow a_i^+ - a_i^+ a_i^+ a_i,\ I_i^z \Rightarrow \tfrac{1}{2} - a_i^+ a_i, \qquad [3]$$

and using the impulse presentation

$$a_k^+ = \frac{1}{\sqrt{N}} \sum_{\rm i}^N a_{\rm i}^+ \exp(-ikr_i),$$

$$a_k = \frac{1}{\sqrt{N}} \sum_i^N a_i \exp(ikr_i),$$

$$\epsilon_k = \sum_j^N \epsilon_{ij} \exp(-ik(r_i - r_j)),$$

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we write the Hamiltonian [1] in the form[1]

$$\begin{aligned} \mathcal{H} = &-\frac{1}{2}\epsilon_0 N + \sum_k \hbar\omega_k a_k^+ a_k \\ &+ \frac{1}{4N}\sum_{k,p,q} \Gamma_{kp}^q a_{k-q}^+ a_{p+q}^+ a_k a_p \\ &- \hbar\omega_1\sqrt{N}(a_0 \mathrm{e}^{i\omega t} + a_0^+ \mathrm{e}^{-i\omega t}), \end{aligned} \qquad [4]$$

where[2]

$$\begin{aligned} \hbar\omega_k &= \hbar\omega_0 + \epsilon_k, \\ \Gamma_{kp}^q &= -2(\epsilon_{p+q} + \epsilon_{q-k} + 2\epsilon_q + 2\epsilon_{p+q-k}). \end{aligned} \qquad [5]$$

It should be noted that we do not take into account the nonsecular terms in the Hamiltonian [1]; hence expression [4] does not contain three-magnon terms. But as the gap in the spectrum of magnons caused by the Zeeman splitting is much more than the width of the spectrum zone $\epsilon_k^{\max} - \epsilon_k^{\min}$, the probability of parametric excitations by three-magnon processes is negligible (see also (*7*)).

Furthermore, we make the standard simplification (*6, 7*) taking into account that the number of magnons of the uniform precession is much more than the number of thermal magnons. Thus we can write [4] in the form

$$\begin{aligned} \mathcal{H} = &\sum_k \omega_k a_k^+ a_k + \sum_k T_{0k} a_k^+ a_k a_0^+ a_0 \\ &- \omega_1\sqrt{N}(a_0 e^{i\omega t} + a_0^+ e^{-i\omega t}) + \sum_k S_{0k} a_k^+ a_{-k}^+ a_0 a_0 \\ &+ \sum_k S_{k0} a_0^+ a_0^+ a_k a_{-k}, \end{aligned} \qquad [6]$$

where $T_{0k} = -(6/N)\epsilon_k$, $S_{0k} = -(3/N)\epsilon_k$, $S_{k0} = -(2/N)\epsilon_k$. Hamiltonian [6] is not Hermitian ($S_{0k} \neq S_{k0}$) because of the nonunitarity of transformation [3]. Hence, as in the case of the electron spin system, we would write the equations for a_k and a_k^+ by using the relations (*12*) $i\partial a_k/\partial t = [\mathcal{H}, a_k]$ and $i\partial a_{-k}^+/\partial t = [\mathcal{H}, a_{-k}^+]$. After quantum-statistical averaging we get

$$\begin{aligned} &\left[\frac{d}{dt} + \gamma_k + i(\omega_k + T_{0k} a_0^+ a_0)\right] \\ &\quad \times \langle a_k \rangle + 2i \cdot S_{0k}\langle a_{-k}^+ a_0 a_0 \rangle = 0 \\ &\left[\frac{d}{dt} + \gamma_k - i(\omega_k + T_{0k} a_0^+ a_0)\right] \\ &\quad \times \langle a_{-k}^+ \rangle - 2i \cdot S_{k0}\langle a_0^+ a_0^+ a_k \rangle = 0 \end{aligned} \qquad [7]$$

for $k \neq 0$, and

$$\left[\frac{d}{dt} + \gamma_0 + i\omega_0\right]\langle a_0 \rangle - i\omega_1\sqrt{N} \cdot e^{-i\omega t} = 0 \qquad [8]$$

for $k = 0$. Here (*1, 4*) $\gamma_k \approx (1 - p)\omega_d$ is the value of the linear decrement of the k wave and ω_d is a characteristic frequency of dipole–dipole interaction. From [8] one can obtain the stationary solution

$$\langle a_0 \rangle = \langle a_0^+ \rangle^* = \frac{\omega_1\sqrt{N}}{\Delta - i\gamma_0} e^{-i\omega t}, \qquad [9]$$

where $\Delta = \omega_0 - \omega$ and $\langle \cdots \rangle$ denotes the quantum-statistical average. Furthermore, one may uncouple the nonlinear terms in expression [7] and get

$$\begin{aligned} &\left[\frac{d}{dt} + \gamma_k + i\tilde{\omega}_k\right]\langle a_k \rangle + 2iS_{0k}\langle a_0 \rangle^2 \langle a_{-k}^+ \rangle = 0 \\ &\left[\frac{d}{dt} + \gamma_k - i\tilde{\omega}_k\right]\langle a_{-k}^+ \rangle - 2iS_{k0}\langle a_0^+ \rangle^2 \langle a_k \rangle = 0 \\ &\qquad \text{for } k \neq 0, \end{aligned} \qquad [10]$$

where $\tilde{\omega}_k = \omega_k + T_{0k}|\langle a_0 \rangle|^2$.

Let us search for a solution of system of Eqs. [10] in the form $\langle a_k(t) \rangle = \langle a_k(0) \rangle \cdot \exp(\nu_k t + i\omega t)$, $\langle a_{-k}^+(t) \rangle = \langle a_{-k}^+(0) \rangle \cdot \exp(\nu_k t - \mathrm{i}\omega \mathrm{t})$. Substituting these expressions into [10] we get the condition for increment ν_k:

$$\nu_k = \sqrt{4S_{0k}S_{k0}|\langle a_0 \rangle|^4 - (\tilde{\omega}_k - \omega)^2} - \gamma_k. \qquad [11]$$

If $\nu_k > 0$, exponential growth of the number of parametrical magnons takes place. Thus we obtain from [11] the condition of parametric instability,

$$\begin{aligned} &\frac{\Delta + \epsilon_k}{2\epsilon_k}\left[1 - \left(\frac{2}{3} - \frac{\gamma_k^2}{3(\Delta + \epsilon_k)^2}\right)^{1/2}\right]\Bigg|_{\min} \\ &\quad < \frac{\omega_1^2}{\Delta^2 + \gamma_0^2} \\ &\quad < \frac{\Delta + \epsilon_k}{2\epsilon_k}\left(1 + \left(\frac{2}{3} - \frac{\gamma_k^2}{3(\Delta + \epsilon_k)^2}\right)^{1/2}\right)\Bigg|_{\max}. \end{aligned} \qquad [12]$$

Moreover, the conditions $(\Delta + \epsilon_k)/\epsilon_k > 0$ and $|\Delta + \epsilon_k| > |\gamma_k|/\sqrt{2}$ must be satisfied. The minimum and maximum

[1] a_i^+ and a_i (as a_k^+ and a_k) are not complex conjugate operators, but they satisfy Bose–Einstein commutation relations (*10, 11*).

[2] In the case of a spherical sample with a cubic lattice (*1*), $\epsilon_0 = 0$. For simplicity we will consider only this case.

values in expression [12] should be taken for constant Δ and for ϵ_k situated within the limits $\epsilon_k^- < \epsilon_k < \epsilon_k^+$, where (*1, 4*) $\epsilon_k^- < 0$ and $\epsilon_k^+ > 0$ (ϵ_k^- and ϵ_k^+ denote minimum and maximum values of ϵ_k, respectively). The upper limit in expression [12] does not have a physical sense because (as the calculations show) the upper limit is always greater than or an order of unity, but the condition of the applicability of spin-wave approximation imposes the stronger restriction

$$n_0/N = \langle a_0^+ a_0 \rangle / N = \omega_1^2/(\Delta^2 + \gamma_0^2) \ll 1. \qquad [13]$$

The lower limit[3] in expression [12] determines the critical amplitude of the RF magnetic field ω_1^{cr}. Minimizing [12] one can obtain the values of energy ϵ_k^{F} of the first excited magnons (magnons are excited when $\omega_1 = \omega_1^{\mathrm{cr}}$) for the various Δ assuming $\gamma_k = \gamma_0 \equiv \gamma$:

$$\epsilon_k^{\mathrm{F}} = -\gamma \frac{|\Delta|}{\Delta} \left(\frac{3}{2} \left[\frac{\gamma^2}{\Delta^2} + 1 \right] \right)^{1/2} \times \left\{ 1 + \frac{\Delta}{\gamma} \left(\frac{2}{3} \left[\frac{\gamma^2}{\Delta^2} + 1 \right] \right)^{1/2} \right\}. \qquad [14]$$

Moreover, the condition

$$\epsilon_k^- < \epsilon_k^{\mathrm{F}} < \epsilon_k^+ \qquad [15]$$

must be satisfied. Expression [14] is useful only in the ranges $\Delta_2^- < \Delta < \Delta_1^-$ and $\Delta_1^+ < \Delta < \Delta_2^+$, where $\Delta_1^\pm \equiv -\gamma^2(1 + \sqrt{3/2})/\epsilon_k^\mp$ and $\Delta_2^\pm \equiv -\epsilon_k^\mp - \sqrt{3/2}\gamma$ (it can be shown that $(\epsilon_k^{\mathrm{F}})^{\min} > 0$ for $\Delta < 0$ and $(\epsilon_k^{\mathrm{F}})^{\max} < 0$ for $\Delta > 0$). In the other regions of Δ, restriction [15] is not valid and expression [14] becomes useless, but after some analytical consideration one obtains that for $\Delta_1^- < \Delta < \Delta_0$

$$\left(\Delta_0 \equiv \frac{-\gamma^2}{4(\sqrt{3/2} - 1)} \cdot \frac{\epsilon_k^- + \epsilon_k^+}{\epsilon_k^- \epsilon_k^+} \approx 0 \right) \epsilon_k^{\mathrm{F}} = \epsilon_k^+$$

and for $\Delta_0 < \Delta < \Delta_1^+ \epsilon_k^{\mathrm{F}} = \epsilon_k^-$. Moreover, for $\Delta_3^- < \Delta < \Delta_2^-$ and $\Delta_2^+ < \Delta < \Delta_3^+$ ($\Delta_3^\pm \equiv -\epsilon_k^\mp - \gamma/\sqrt{2}$), $\epsilon_k^{\mathrm{F}} = \epsilon_k^+$ and $\epsilon_k^{\mathrm{F}} = \epsilon_k^-$, respectively; for $\Delta < \Delta_3^-$ $\epsilon_k^{\mathrm{F}} = \epsilon_k^-$ and for $\Delta > \Delta_3^+$ $\epsilon_k^{\mathrm{F}} = \epsilon_k^+$. After substituting these values of the energy ϵ_k^{F} of the first excited magnons (Fig. 1b) in the lower limit of expression [12], one obtains the critical values of the amplitude ω_1^{cr} of the RF magnetic field (Fig. 1a). It is easy to see that for $\Delta = 0$ $\omega_1^{\mathrm{cr}}(0) = \gamma(\frac{1}{2}(1 - \sqrt{\frac{2}{3}}))^{1/2} \simeq 0,3 \cdot \gamma$. At the points $\Delta = \Delta_3^\pm$ the function ω_1^{cr} jumps in value. Furthermore, in the ranges $\Delta < \Delta_3^-$ and $\Delta > \Delta_3^+$, ω_1^{cr} grows so rapidly that condition [13] is not satisfied, and we cannot use the spin-wave approximation approach in these ranges.

[3] Taking into consideration expression [13] the lower limit (left term in condition [12]) also should be much less than unity.

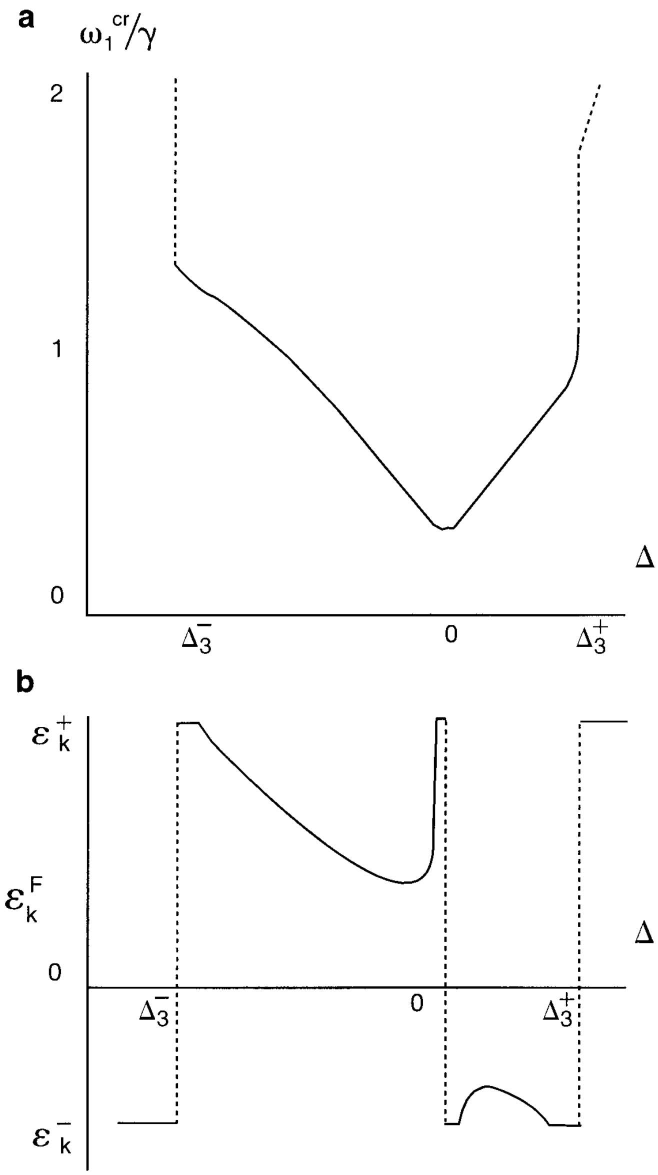

FIG. 1. (a) Dependence of ω_1^{cr} (in γ units) on the detuning Δ. (b) Dependence of first excited magnon's energy on the detuning Δ. In calculations it was taken to be $\epsilon_k^+ = -2\epsilon_k^- = 10\gamma$.

In contrast to the case of electron spin systems in magnets (*6, 7*) where the energies of the first excited magnons satisfy the parametric resonance condition

$$\tilde{\omega}_k = \omega, \qquad [16]$$

in the case of NSS with dipole–dipole interaction another situation develops. Indeed, for $\Delta = 0$ the condition [16] gives $\epsilon_k = 0$ but in view of the form of the Hamiltonian [6] one can see that the probability of the excitation of parametric magnons with such energies is equal to zero. Unlike this, in NSS with dipole–dipole interaction, as concluded from the above, the magnons from the bottom or from the top of the spectrum zone ($\epsilon_k^F = |\epsilon_k|^{max}$) are excited.

The results obtained above can be verified experimentally very easily if the imaginary part of the magnetic susceptibility χ'' is measured. In the case $\omega_1 < \omega_1^{cr}$, χ'' may be given by the expression (*7*)

$$\chi'' = g^2\hbar^2 \frac{\gamma}{\gamma^2 + \Delta^2},$$

but if the amplitude of an RF magnetic field ω_1 exceeds the critical value ω_1^{cr}, parametric instability takes place and χ'' becomes strongly dependent on ω_1. This is most distinct (*6, 7*) for the detuning range $\gamma \ll \Delta \sim \omega_d$, where after ω_1 exceeds the critical value (calculated above) χ'' grows rapidly.

In Fig. 1a the values of ω_1^{cr} are given for the different Δ. For example, if in CaF_2 the spin temperature of thermal magnons is $T = 3 \times 10^{-3}$K and if $H_0 = 5 \times 10\ ^4$Oe ($1 - p \simeq 0, 1 \ll 1$), then $\gamma \approx \omega_d(1 - p) \approx 10^3\ s^{-1}$ and $\omega_1^{cr}(0) \approx 3 \times 10^2\ s^{-1}$. So under these conditions in the detuning range $-\epsilon_k^+ < \Delta < -\epsilon_k^-$ the value of ω_1^{cr} does not exceed $\sim 10^3\ s^{-1}$.

As was mentioned initially, the considered instability may cause magnon chaos. In particular, we propose that the RF magnetic field with modulated amplitude close to the critical value can be a reason for chaotic behavior of spin excitations. However, this problem is a subject for further investigations.

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