

Nonlinear spin dynamics in nuclear magnets

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A method is developed for solving nonlinear systems of differential, or integrodifferential, equations with stochastic fields. The method makes it possible to give an accurate solution for an interesting physical problem: What are the peculiarities of nonlinear spin dynamics in nonequilibrium nuclear magnets coupled with a resonator? Evolution equations for nuclear spins are derived based on a Hamiltonian with dipole interactions. The ensemble of spins is coupled with a resonator electric circuit. Seven types of main relaxation regimes are found: free induction, collective induction, free relaxation, collective relaxation, weak superradiance, pure superradiance, and triggered superradiance. The initial motion of spins can be originated by two reasons, either by an imposed initial coherence or by local spin fluctuations due to nonsecular dipole interactions. The relaxation regimes caused by the second reason cannot be described by the Bloch equations. Numerical estimates show good agreement with experiment.

I. INTRODUCTION

Spin dynamics in polarized nonequilibrium systems is usually described by using the Bloch equations for the components of uniform magnetization. The derivation of the Bloch equations from the evolution equations for a spin model can be found, for example, in ter Haar.¹ The solution of the Bloch equations for the case of small deviations from a stationary state is straightforward and well known in the theory of magnetic resonance.² The situation becomes more complicated when the spin system is coupled with a resonator. Then there appears an essential nonlinearity due to the action of resonator feedback field. The system of the coupled Bloch and resonator-field equations is typical of the theory of maser amplifiers and generators.³

The nonlinear system of the Bloch and resonator-field equations can be slightly simplified by invoking the slowly varying amplitude approximation.³ However, this does not help much, since the resulting equations are, as before, nonlinear. To achieve further simplification, one resorts to the adiabatic approximation which leads to the proportionality of the feedback field to transverse magnetization, that is, to the static coupling.⁴⁻⁷ The adiabatic approximation, as is known,⁸ works well only at the final stage of relaxation processes when different variables adiabatically follow each other, but it cannot correctly describe intermediate stages where transient phenomena occur. The incorrectness of the static-coupling approximation is physically evident, as only moving spins, but not immovable, are able to induce a field in resonator. More accurate is the dynamic-coupling approximation⁹ in which the feedback field is proportional to the time derivative of transverse magnetization. But both these, static- as well as dynamic-coupling, approximations do not take into account retardation effects that may be important for transient phenomena.

Moreover, the Bloch equations themselves may be inappropriate for explaining some kinds of relaxation processes. This concerns, for example, the interpretation of the recent series of experiments¹⁰⁻¹⁵ observing nuclear spin superradi-

ance. In these experiments a nonequilibrium system of polarized nuclear spins is placed inside a coil of a resonance electric circuit. The initial polarization is directed opposite to an external magnetic field. If this polarization is sufficiently high and the coupling with a resonator is enough strong, then the power of the current, as a function of time, after some delay, displays a sharp burst with a damping time much shorter than the dephasing time T_2 . This time behavior of the current power is analogous to that of the radiation intensity of atoms or molecules in the case of optical superradiance. Because of this analogy, the corresponding coherent phenomenon in spin systems has also been called superradiance or, more concretely, spin superradiance. Friedberg and Hartmann¹⁶ pointed out that the whole process of interaction of a spin system and a resonance coil, in fact, involves no radiation into free space but merely nonradiative transfer of energy from the sample to the coil, where the energy is dissipated Ohmically. Nevertheless, the term "spin superradiance" has become commonly used. The excuse for this is not solely the formal analogy of temporal behavior of current power, for spin systems, and of radiation intensity, for atomic and molecular systems, but also a deep physical similarity: Spin superradiance, as well as optical superradiance, is a collective process of coherent self-organization, although the self-organized coherence of spin motion develops not because of a common radiation field, as in atomic and molecular systems, but owing to a resonator feedback field. In addition, coherent motion of spins inevitably produces coherent magnetodipole emission with properties completely analogous to superradiance of optical systems, though the magnetodipole radiation intensity is too weak to be measured as easy as the power of current.¹⁷

In the same way as for optical systems,¹⁸ one has to distinguish the pure from triggered spin superradiance. *Pure spin superradiance* is a purely self-organized process starting from an absolutely incoherent state when the average transverse magnetization is strictly zero. *Triggered spin superradiance* is a process in which self-organization also plays an important role but whose beginning is triggered by an initial

coherence imposed onto the spin system, that is, by assuming that the mean transverse magnetization is not zero.

The interpretation of pure spin superradiance cannot be based on the Bloch equations because of the following. If the initial transverse magnetization is zero, then, in the content of these equations, the relaxation of an inverted spin system can be due only to two reasons: either to spin-lattice interactions characterized by a relaxation time T_1 or to thermal damping caused by the Nyquist noise of resonator. At very low temperature, typical of experiments¹⁰⁻¹⁵ with polarized nuclear spins, the spin-lattice relaxation time T_1 is much longer than the dephasing time T_2 ; therefore this mechanism cannot develop coherence. The resonator thermal damping, as shown by Bloembergen and Pound,¹⁹ is negligibly small for macroscopic systems, the thermal relaxation time being proportional to the number of spins, N , in the sample, and so being much longer than not only T_2 but even T_1 . Thus, the resonator Nyquist noise can never produce the initial thermal relaxation. The radiation field in the coil does not provide a microscopic thermal relaxation mechanism, but the inhomogeneous internal, or local, fields are essential.¹⁹

The Bloch equations cannot, in principle, describe pure spin superradiance and, in general, any other relaxation regimes in which no initial coherence is imposed on the spin system. To treat all possible relaxation regimes for a non-equilibrium spin system, coupled with a resonator, it is necessary to take into account local spin fluctuations. This can be done by considering a microscopic model with realistic dipole interactions between nuclear spins. But since the local fields are essential, we cannot invoke for a microscopic model a homogeneous approximation. The latter would immediately return us to the Bloch equations with the lost information on local spin fluctuations.

If the number of spins, N , is not too large, say, between 10 and 10^3 , then one can resort to a numerical solution of the corresponding evolution equations. Such a computer simulation, whose mathematical details can be found in Ref. 20, has been accomplished¹⁷ and confirmed the crucial importance of local spin fluctuations. These are sufficient for describing pure spin superradiance, with no influence of the resonator thermal noise.

Computer simulations, however, can give only a qualitative picture, as the number of spins involved is incomparably smaller than what one has in real samples with N of the order of 10^{23} . In addition, such simulations provide no analytical formulas, making it very difficult, if possible, to classify all possible relaxation regimes occurring when varying the numerous parameters of the system.

The aim of the present paper is to untangle two mutually interrelated problems: first, to formulate a method allowing an analytical solution for a system of nonlinear equations, taking into account local fluctuating fields, as well as dynamic coupling and retardation effects, and second, to analyze various relaxation regimes of nonequilibrium nuclear magnets coupled with a resonator.

II. METHOD OF SOLUTION

The method to be presented here may be used not only for the particular problem discussed in the Introduction, but for a wide variety of evolution equations for different systems. In

this section we will preserve the generality of the presentation. All necessary specifications related to the spin dynamics in nuclear magnets will be expounded in the following sections. To better understand the principal ideas of the method, it is convenient to divide it into several steps.

A. Separation of variables

Suppose that in the problem under consideration there is a set

$$\varepsilon = \{\varepsilon_i | i = 1, 2, \dots; |\varepsilon_i| \ll 1\}$$

of small parameters. Depending on the way in which these parameters enter into the evolution equations, we may distinguish fast and slow variables. The terms describing local fluctuating fields can be treated as random, or stochastic, variables

$$\varphi = \{\varphi_i | i = 1, 2, \dots; \mu_\varphi\},$$

with a probability measure μ_φ .

The fast variables

$$u = \{u_i(\varphi, t) | i = 1, 2, \dots; t \geq 0\}$$

and slow variables

$$s = \{s_j(\varphi, t) | j = 1, 2, \dots; t \geq 0\}$$

differ from each other by the properties of their evolution equations

$$\frac{du}{dt} = f(u, s, \varphi, t, \varepsilon) \quad (1)$$

and

$$\frac{ds}{dt} = \varepsilon g(u, s, \varphi, t, \varepsilon), \quad (2)$$

whose right-hand sides are such that the limit

$$\lim_{\varepsilon \rightarrow 0} f(u, s, \varphi, t, \varepsilon) \neq 0 \quad (3)$$

is not zero, while

$$\lim_{\varepsilon \rightarrow 0} \varepsilon g(u, s, \varphi, t, \varepsilon) = 0. \quad (4)$$

Here and in what follows the matrix form of notation is used, according to which $f = \{f_i\}$, $g = \{g_i\}$, and the product $\varepsilon g = \{\sum_j c_{ij} \varepsilon_j g_j\}$ is to be understood as a column of linear combinations with coefficients c_{ij} . All parameters, variables, functions, and coefficients can be complex except $t \geq 0$ representing time. The limit $\varepsilon \rightarrow 0$ means that all $\varepsilon_i \rightarrow 0$. The right-hand sides of (1) and (2) can contain integral operators, provided that the limits (3) and (4) hold. For brevity, the dependence of the fast, u , and slow, s , variables on the parameters ε is not explicitly written. Equations (1) and (2) are to be complimented by initial conditions

$$u(\varphi, 0) = u_0, \quad s(\varphi, 0) = z_0. \quad (5)$$

The limiting properties (3) and (4) explain why the evolution equations of the form (1) correspond to fast variables, as compared to the evolution equations of the type (2) describing slow variables.

The fast and slow variables are not necessarily simply defined for each given problem, but the aim of this step is to introduce such variables by using the information on the existence of small parameters and by choosing the appropriate changes of variables, so that finally they could be distinguished in the above sense.

B. Quasi-integrals of motion

As far as the slow variables, by definition, vary with time much slower than the fast variables, the former may be considered as quasi-integrals of motion for the latter. Then we can try to solve the equations for fast variables under slow variables kept as fixed parameters. With the renotation

$$u \rightarrow X, \quad s \rightarrow z, \tag{6}$$

where z is fixed, from (1) we have

$$\frac{\partial X}{\partial t} = f(X, z, \varphi, t, \varepsilon), \tag{7}$$

which defines

$$X = X(z, \varphi, t). \tag{8}$$

The art of choosing variables is to get for (7) as simple equation as possible. In many cases this can be done so that (7), under fixed z , becomes a system of linear equations. The quasi-integrals of motion play here a role similar to the guiding centers in the guiding-center approach.²¹

C. Method of averaging

For the fast variable (8) we define the asymptotic period T_0 by the condition

$$\lim_{\varepsilon \rightarrow 0} |X(z, \varphi, t + T_0) - X(z, \varphi, t)| = 0. \tag{9}$$

If (9) gives several solutions for T_0 , the smallest of them is to be taken. And if (9) has no solution for T_0 , we put $T_0 \rightarrow \infty$.

To find the time evolution of quasi-integrals of motion, we substitute (8) into the right-hand side of (2) and introduce the averaged function

$$\bar{g}(z, \varepsilon) \equiv \int \left[\frac{1}{T_0} \int_0^{T_0} g(X(z, \varphi, t), z, \varphi, t, \varepsilon) dt \right] d\mu_\varphi. \tag{10}$$

Then the equation

$$\frac{dz}{dt} = \varepsilon \bar{g}(z, \varepsilon) \tag{11}$$

gives the sought time evolution.

The foundation for this step is the Krylov-Bogolubov method of averaging.^{22,23} The major difference in our case is that the Krylov-Bogolubov vector field (10) is defined as an average with respect to time and, in addition, with respect to the stochastic variable φ .

D. Basic approximation

The basic approximations for slow and fast variables are defined as follows. For the slow variables this is given by the solution

$$z = z(t) \tag{12}$$

of Eq. (11), with the initial condition

$$z(0) = z_0. \tag{13}$$

Substituting (12) into (8), we have

$$x = x(\varphi, t) = X(z(t), \varphi, t) \tag{14}$$

for fast variables. The integration constant appearing when solving (7) is to be found from the initial condition

$$x(\varphi, 0) = u_0. \tag{15}$$

Note that (11) is, generally, a nonlinear equation taking into account the characteristic features of the initial system (1) and (2) in the way typical of the averaging methods. Actually, the basic approximation of this subsection is nothing but the corrected first approximation of the method of averaging.²²⁻²⁵ The solutions to Eqs. (7) and (11) are close to the solutions of the initial Eqs. (1) and (2) in the same asymptotic (with respect to $\varepsilon \ll 1$) sense as is customary for all variants of the averaging method.²²⁻²⁵ Thus, for the norm of a vector function $z = \{z_i | i = 1, 2, \dots\}$ given, e.g., by the so-called octahedral form $\|z\| \equiv \sum_i |\langle z_i \rangle|$, where $\langle z_i \rangle \equiv \int z_i d\mu_\varphi$, we have $\lim_{\varepsilon \rightarrow 0} \|z - s\| = 0$. Similarly, $\lim_{\varepsilon \rightarrow 0} \|u - x\| = 0$. For finite values of $\varepsilon \ll 1$, it is possible to find estimates for the norm of the difference $\|z - s\| \leq \varepsilon \times \text{const}$, and analogously, for $\|u - x\| \leq \varepsilon \times \text{const}$, valid in the time interval $0 \leq t \leq \text{const}/\varepsilon$. These estimates can be easily constructed by using the inequality

$$\frac{d}{dt} \|z\| \leq \left\| \frac{dz}{dt} \right\|.$$

All details of such constructions are expounded in numerous works.²²⁻²⁵

E. Generalized expansion

Corrections to the basic approximation can be found by using the generalized asymptotic expansion,

$$u = x(\varphi, t) + \sum_{n=1}^{\infty} x_n(\varphi, t) \varepsilon^n, \\ s = z(t) + \sum_{n=1}^{\infty} z_n(\varphi, t) \varepsilon^n, \tag{16}$$

about (12) and (14).

The right-hand sides of (1) and (2) are also to be expanded in a similar manner, as

$$f(u, s, \varphi, t, \varepsilon) = f(x, z, \varphi, t, \varepsilon) + \sum_{n=1}^{\infty} f_n(\varphi, t, \varepsilon) \varepsilon^n. \quad (17)$$

For example, in the first two orders we have

$$f_1 = x_1 f'_x + z_1 f'_z,$$

$$f_2 = x_2 f'_x + z_2 f'_z + x_1 z_1 f''_{xz} + \frac{1}{2} (x_1^2 f''_{xx} + z_1^2 f''_{zz}),$$

where the notation

$$f'_x \equiv \frac{\partial}{\partial x} f(x, z, \varphi, t, \varepsilon), \quad x = x(\varphi, t), \quad z = z(t)$$

is used.

The expansions (16) and (17) are to be substituted into the evolution equations (1) and (2). In doing this, we notice that, since because of (14)

$$\frac{dx}{dt} = \left(\frac{\partial X}{\partial t} \right)_z + \left(\frac{\partial X}{\partial z} \right)_t \frac{dz}{dt},$$

then invoking (7) and (11), we get

$$\frac{dx}{dt} = f(x, z, \varphi, t, \varepsilon) + \varepsilon \bar{g}(z, \varepsilon) X'_z(\varphi, t),$$

where

$$X'_z(\varphi, t) \equiv \frac{\partial}{\partial z} X(z, \varphi, t), \quad z = z(t).$$

After substituting the above expansions into Eqs. (1) and (2), we need to compare the left- and right-hand sides of the obtained series. This comparison is made in the way standard for dealing with generalized asymptotic expansions, whose coefficients, unlike those of simple asymptotic series, depend themselves on small parameters. Comparing the generalized asymptotic expansions, one has to keep their coefficients fixed, thus making it possible to equate similar terms with respect to the powers of the expansion parameters. This approach, suggested by Lindstedt, has been substantiated and developed by Poincaré for the problems of celestial mechanics.²⁶ When one equates similar terms of generalized asymptotic expansions, one, of course, gets sufficient, but not necessary, conditions. However, this does not give any trouble, since it merely reflects the known fact that the same function can be presented by different forms of generalized asymptotic series. The mutual transformations between these different forms correspond to the so-called resummation procedure. Generalized asymptotic expansions nowadays are not only widely used but are the basic practical tool of perturbation theory for nonlinear differential equations.²⁷⁻³⁰

Equating similar terms with respect to the power of ε , we obtain the equations for the corrections of arbitrary order. It is important to stress that all these equations are linear; thus, there is no principal difficulty in solving them. To exemplify this, at the same time avoiding cumbersome formulas, let us think of ε as of one parameter. Then for the first-order corrections we find the equations

$$\frac{dx_1}{dt} = f_1(\varphi, t, \varepsilon) - \bar{g}(z, \varepsilon) X'_z(\varphi, t),$$

$$\frac{dz_1}{dt} = g(x, z, \varphi, t, \varepsilon) - \bar{g}(z, \varepsilon). \quad (18)$$

The initial conditions, in compliance with (13) and (15), are

$$x_1(\varphi, 0) = 0, \quad z_1(\varphi, 0) = 0. \quad (19)$$

For all subsequent orders we have

$$\frac{dx_n}{dt} = f_n(\varphi, t, \varepsilon),$$

$$\frac{dz_n}{dt} = g_n(\varphi, t, \varepsilon) \quad (n \geq 2), \quad (20)$$

with the initial conditions

$$x_n(\varphi, 0) = 0, \quad z_n(\varphi, 0) = 0. \quad (21)$$

The first equation of (18) can be reduced to the form

$$\frac{dx_1}{dt} = x_1 f'_x + \Delta_1 - \bar{g} X'_z,$$

in which

$$\Delta_1 \equiv f_1 - x_1 f'_x = z_1 f'_z.$$

As we see, the equation for x_1 is really linear, since

$$z_1(\varphi, t) = \int [g(x, z, \varphi, t, \varepsilon) - \bar{g}(z, \varepsilon)] dt \quad (22)$$

immediately follows from the second equation of (18). The solution for this linear equation is

$$x_1 = e^p \int e^{-p} (\Delta_1 - \bar{g} X'_z) dt, \quad (23)$$

where

$$p = p(\varphi, t, \varepsilon) \equiv \int f'_x(\varphi, t, \varepsilon) dt.$$

For the second-order corrections, from (20), we find

$$x_2 = e^p \int e^{-p} \Delta_2 dt, \quad z_2 = \int g_1 dt, \quad (24)$$

with

$$\Delta_2 \equiv f_2 - x_2 f'_x = z_2 f'_z + x_1 z_1 f''_{xz} + \frac{1}{2} (x_1^2 f''_{xx} + z_1^2 f''_{zz}).$$

Similarly, for the n th-order corrections we obtain the general formulas

$$x_n = e^p \int e^{-p} \Delta_n dt,$$

$$z_n = \int g_{n-1} dt \quad (n \geq 2), \quad (25)$$

in which

$$\Delta_n \equiv f_n - x_n f'_x.$$

The simplicity of obtaining the higher-order corrections, satisfying linear equations, is a considerable advantage of the suggested generalized asymptotic expansion, as compared to the guiding-center approach²¹ or higher-order averaging methods^{22,23} in which each subsequent approximation order invokes more and more complicated nonlinear equations. Here we meet nonlinear equations only once, at the third step, when solving (11), which corresponds to the first-order averaging method.

The use of the averaging method only at one step makes it possible, from one side, to include the characteristic nonlinearity into our basic approximation and, from another side, to define all corrections by simple formulas. The idea of dividing solutions onto their principal parts, including essential nonlinearities, and perturbative corrections, defined by linear equations, greatly helps in solving complicated nonlinear problems.³¹ This idea, actually, goes back to the Struble technique^{32,33} employed for solving the Mathieu equation. Note that the nonlinear principal part could be also defined by other techniques known in the theory of singular perturbations, for instance, by using the methods of strained coordinates, multiple scales, nonlinear renormalizations, matched expansions, variation of parameters, and so on.²⁷⁻³⁰ However, these methods, as is discussed in Refs. 27-30, are more ambiguous, more cumbersome, and less general than the method of averaging.

Finally, we need to remember that, in our case, the solutions of nonlinear equations (1) and (2) contain the stochastic variable φ . As far as observable quantities should not depend on that variable, this means that the former are to be averaged with respect to the random φ with a given probability measure. The solutions themselves are not necessarily such quantities that can be measured directly, but usually, the observables are some functions or functionals of these solutions. This especially concerns the fast variables, while the slow variables are often directly measurable.

III. NUCLEAR MAGNET

The system of nuclear spins can be modeled, as is accepted in the theory of nuclear magnetic resonance,² by the Hamiltonian

$$\hat{H} = \frac{1}{2} \sum_{i \neq j}^N H_{ij} - \mu \sum_{i=1}^N \vec{B} \cdot \vec{S}_i \quad (26)$$

with the dipole interaction energy

$$H_{ij} = \frac{\mu^2}{r_{ij}^3} [\vec{S}_i \cdot \vec{S}_j - 3(\vec{S}_i \cdot \vec{n}_{ij})(\vec{S}_j \cdot \vec{n}_{ij})], \quad (27)$$

in which μ is a nuclear magneton, $\vec{S}_i = \{S_i^x, S_i^y, S_i^z\}$ is a spin operator, and

$$\vec{n}_{ij} \equiv \frac{\vec{r}_{ij}}{r_{ij}}, \quad \vec{r}_{ij} \equiv \vec{r}_i - \vec{r}_j, \quad r_{ij} \equiv |\vec{r}_{ij}|.$$

The total magnetic field

$$\vec{B} = \vec{H}_0 + \vec{H}, \quad \vec{H}_0 = H_0 \vec{e}_z, \quad \vec{H} = H \vec{e}_x \quad (28)$$

consists of two parts: The first is an external magnetic field H_0 directed along the z axis; the second, H , is a field of the coil of a resonance electric circuit, the coil axis being directed along the axis x . The sample is inserted into the coil.

The initial state of the spin system is assumed to be non-equilibrium and characterized by a statistical operator $\hat{\rho}(0)$. So the average spin

$$\langle \vec{S}_i \rangle \equiv \text{Tr} \hat{\rho}(0) \vec{S}_i(t) = \text{Tr} \hat{\rho}(t) \vec{S}_i(0)$$

is a function of time. The evolution equations for averages can be obtained by using either the Liouville equation for the statistical operator $\hat{\rho}(t)$ or the Heisenberg equations of motion for operators. We prefer the latter based on the Heisenberg equations.

The resonance electric circuit, coupled with the spin sample, is characterized by resistance R , inductance L , and capacity C . The coil, in which the sample is immersed, has n turns of cross section A_0 over a length l . The magnetic field inside the coil,

$$H = \frac{4\pi n}{cl} j, \quad (29)$$

is formed by an electric current satisfying the Kirchhoff equation

$$L \frac{dj}{dt} + Rj + \frac{1}{C} \int_0^t j(\tau) d\tau = - \frac{d\Phi}{dt} + E_f, \quad (30)$$

in which E_f is an electromotive force of external fields, if any, and of the thermal Nyquist noise; the magnetic flux

$$\Phi = \frac{4\pi}{c} n A_0 \eta \rho M_x$$

is due to the x component of the magnetization

$$M_x = \frac{\mu}{N} \sum_{i=1}^N \langle S_i^x \rangle,$$

and the filling factor η and spin density ρ are

$$\eta \equiv \frac{V}{V_0}, \quad \rho \equiv \frac{N}{V} \quad (V_0 \equiv l A_0),$$

respectively.

The resonance electric circuit will be called, for brevity, the resonator, and the internal coil field (29), the resonator field. For the latter, the Kirchhoff equation (30) can be rewritten as

$$\frac{dH}{dt} + 2\gamma_3 H + \omega^2 \int_0^t H(\tau) d\tau = -4\pi\eta\rho \frac{dM_x}{dt} + \frac{cE_f}{nA_0},$$

where

$$\omega \equiv \frac{1}{\sqrt{LC}} \quad \left(L \equiv \frac{4\pi n^2 A_0}{c^2 l} \right)$$

is the resonator natural frequency and

$$\gamma_3 \equiv \frac{R}{2L} = \frac{\omega}{2Q} \left(Q \equiv \frac{\omega L}{R} \right)$$

is the resonator damping.

It is convenient to introduce the dimensionless resonator field

$$h \equiv \frac{\mu H}{\hbar \gamma_3}, \quad (31)$$

driving force

$$f \equiv \frac{c \mu E_f}{n A_0 \hbar \gamma_3^2}, \quad (32)$$

and the dimensionless average magnetization

$$s_\nu \equiv \frac{M_\nu}{\mu} = \frac{1}{N} \sum_{i=1}^N \langle S_i^\nu \rangle, \quad (33)$$

in which $\nu = x, y, z$. Define the coupling constant

$$\alpha_0 \equiv \pi \eta \frac{\rho \mu^2}{\hbar \gamma_3}, \quad (34)$$

characterizing the strength of coupling between the spin system and resonator. Then the Kirchhoff equation acquires the form

$$\frac{dh}{dt} + 2\gamma_3 h + \omega^2 \int_0^t h(\tau) d\tau = -4\alpha_0 \frac{ds_x}{dt} + \gamma_3 f. \quad (35)$$

The resonator field h , as seen from (35), can be induced by a driving force f and by moving, but not static, transverse magnetization.

The statistical averaging of a spin operator $S_i^\alpha = S_i^\alpha(t)$, with $\alpha = x, y, z$, is given by $\langle S_i^\alpha \rangle \equiv \text{Tr} \hat{\rho}(0) S_i^\alpha(t)$. We shall use the notation

$$u_i \equiv \langle S_i^- \rangle, \quad s_i \equiv \langle S_i^z \rangle. \quad (36)$$

The statistical operator $\hat{\rho}(0)$ defines the initial values of (36), that is, $u_i(0)$ and $s_i(0)$.

To obtain the evolution equations for the transverse, u_i , and longitudinal, s_i , magnetizations, we have to average the Heisenberg equations of motion. The dipole interactions are of long-range type; therefore the double-spin correlations can be decoupled in the mean-field approximation

$$\langle S_i^\alpha S_j^\beta \rangle \rightarrow \langle S_i^\alpha \rangle \langle S_j^\beta \rangle \quad (i \neq j).$$

Although this decoupling is well justified for long-range forces, it has a deficiency that is important for nonequilibrium processes: It does not take into account the attenuation due to spin-spin interactions. This attenuation appears in the higher-order corrections to the mean-field approximation. The derivation of the spin-spin damping γ_2 in the second-order perturbation theory can be found, e.g., in ter Haar.¹ This damping has to be retained for a correct description of relaxation process, though γ_2 is much smaller than the Larmor frequency

$$\omega_0 \equiv \frac{\mu H_0}{\hbar} > 0.$$

At the same time the small second-order corrections to the oscillation frequency ω_0 can be neglected; alternatively, they can be included into the definition of ω_0 . The mean-field decoupling with corrections leading to the appearance of the spin-spin relaxation parameter γ_2 can be called the corrected mean-field approximation.

Define the arithmetic averages

$$u \equiv \frac{1}{N} \sum_{i=1}^N u_i, \quad s \equiv \frac{1}{N} \sum_{i=1}^N s_i \quad (37)$$

for the transverse and longitudinal magnetizations, respectively, and also for a stationary magnetization

$$\zeta \equiv \frac{1}{N} \sum_{i=1}^N \zeta_i.$$

Introduce the notation for the local fields

$$\delta_i \equiv \frac{1}{N} \sum_{j(\neq i)} \left(\frac{3}{2} a_{ij} s_j + c_{ij} u_j^* + c_{ij}^* u_j \right), \quad (38)$$

which is a real quantity, and

$$\varphi_i \equiv -\frac{2}{\hbar} \sum_{j(\neq i)} (b_{ij} u_j^* + c_{ij} s_j), \quad (39)$$

which is complex. The coefficients a_{ij} , b_{ij} , and c_{ij} are defined in Appendix A.

For the averages in (37) we find

$$i \frac{du}{dt} = -(\omega_0 + i\gamma_2)u + \gamma_3 h s + \frac{1}{N} \sum_{i=1}^N (\delta_i u_i + \varphi_i s_i) \quad (40)$$

and

$$i \frac{ds}{dt} = \frac{1}{2} \gamma_3 h (u - u^*) - i\gamma_1 (s - \zeta) + \frac{1}{2N} \sum_{i=1}^N (\varphi_i^* u_i - \varphi_i u_i^*). \quad (41)$$

The derivation of (40) and (41) is given in Appendix A.

The quantities (38) and (39) are local fluctuating fields,¹ whose existence is due to the inhomogeneity of the spin distribution. If one would resort to a homogeneous approximation, in which u_j and s_j do not depend on the index j , then δ_i and φ_i would be zero, since for the dipole interactions we have

$$\sum_{j(\neq i)}^N a_{ij} \approx \sum_{j(\neq i)}^N b_{ij} \approx \sum_{j(\neq i)}^N c_{ij} \approx 0$$

when $N \rightarrow \infty$ and the spin sample is macroscopic in all three dimensions. The above sums can be nonzero if the number of spins is not high ($N < 10$) or if the sample has a specially prepared irregular shape. Then the nonzero values of these sums are defined by a nonuniformity in the space distribution of spins in the vicinity of the sample surface. Such a boundary nonuniformity for small, at least in one of the dimensions, samples can lead to unisotropic effects in relaxation processes.^{16,34} This kind of inhomogeneity of a sample inside a coil can be explicitly taken into account in the definition of the effective filling factor.¹⁹

It is worth emphasizing that even when the spin sample is macroscopic and has a regular shape, so that the above sums over the dipole interactions are nullified, nevertheless, the local fields (38) and (39) are nonzero if one does not invoke a uniform approximation for the magnetizations u_j and s_j . The local nonuniformities contribute to the inhomogeneous dipole broadening.³⁵ What is the most important is that without taking into account such local fluctuating fields it is impossible, as has been stressed by Bloembergen and Pound,¹⁹ to provide a correct description of relaxation in spin systems.

At the same time, if (38) and (39) depend on the index i showing their local position, then Eqs. (40) and (41) are not closed, but for the case of N spins we need to deal with a system of $3N$ equations. For a macroscopic sample with $N \sim 10^{23}$, to deal with such a number of nonlinear differential equations is a task that is not affordable even for a computer.

A way out of this trouble is as follows. We may treat (38) and (39) as random fluctuating fields with a distribution given by a probability measure μ_φ . That is, we may put into correspondence to the local fields (38) and (39) stochastic fields

$$\{\varphi_0\} \leftrightarrow \{\delta_i\}, \quad \{\varphi\} \leftrightarrow \{\varphi_i\},$$

in which φ_0 is real, representing the real δ_i , and φ is complex representing the complex φ_i . At the present stage an explicit form of the probability measure μ_φ is not important and will be considered later.

With the stochastic representation of local fields in mind, Eqs. (40) and (41) are reduced to

$$\frac{du}{dt} = i(\omega_0 - \varphi_0 + i\gamma_2)u - i(\gamma_3 h + \varphi)s \quad (42)$$

and

$$\frac{ds}{dt} = \frac{i}{2}(\gamma_3 h + \varphi)u^* - \frac{i}{2}(\gamma_3 h + \varphi^*)u - \gamma_1(s - \zeta). \quad (43)$$

Since u is complex, the third equation, additional to (42) and (43), can be the equation for u^* or for $|u|^2$. For the latter we have

$$\frac{d}{dt}|u|^2 = -2\gamma_2|u|^2 - i(\gamma_3 h + \varphi)su^* + i(\gamma_3 h + \varphi^*)su. \quad (44)$$

These equations are to be complemented by initial conditions

$$u(0) = u_0, \quad s(0) = z_0. \quad (45)$$

Equations (42)–(44) for the magnetizations plus Eq. (35) for the resonator field form the basic system of equations permitting a correct description of relaxation processes for a spin sample coupled with a resonator. The physical meaning of all terms in these equations is quite transparent: The real random field φ_0 shifts the oscillation frequency; and the term $\gamma_3 h + \varphi$ plays the role of an effective field acting on spins, h being the resonator field and φ the stochastic field caused by local fluctuations. If in (42)–(44) we put φ_0 and φ zero, then we would return to the Bloch equations; however, the presence of these random fields, as discussed above and as will be demonstrated in what follows, provides a crucial re-

laxation mechanism. Note that the stochastic local fields interconnect the transverse and longitudinal components of magnetization, but do not change the absolute value of the latter whose time variation

$$\frac{d}{dt}(|u|^2 + s^2) = -2\gamma_2|u|^2 - 2\gamma_1 s(s - \zeta)$$

is caused only by spin-spin dephasing collisions and spin-lattice interactions.

If we would decide to invoke the adiabatic approximation, in the way one usually does, then we should put $du/dt \rightarrow 0$ in (42) which immediately results in a linear relation between h and u , that is, in the static approximation. However, as is discussed in the Introduction, such an approximation could be reasonable only at the final stage of relaxation, but cannot correctly describe transient phenomena.

IV. SEPARATION OF VARIABLES

To solve the system of equations (42)–(44) and (35), we use the method developed in Sec. II. To this end, we need to separate fast from slow variables by defining the appropriate small parameters. Usually, the widths γ_1 and γ_2 are small as compared to ω_0 and γ_3 is small as compared to ω . The stochastic fields φ_0 and φ are also to be considered as small, since the corresponding local fields (38) and (39), as is evident from their definition, are of the order of the local dipole interactions, that is, of the order of γ_* which is a part of the inhomogeneous dipole broadening, γ_* being much smaller than ω_0 . Thus, there are four small parameters

$$\frac{\gamma_1}{\omega_0} \ll 1, \quad \frac{\gamma_2}{\omega_0} \ll 1, \quad \frac{\gamma_*}{\omega_0} \ll 1, \quad \frac{\gamma_3}{\omega} \ll 1. \quad (46)$$

An additional small parameter appears in the quiresonance situation when the resonator natural frequency is close to the Larmor frequency of spins. Then the detuning from the resonance Δ gives another small parameter,

$$\frac{|\Delta|}{\omega_0} \ll 1 \quad (\Delta \equiv \omega - \omega_0). \quad (47)$$

The quantities inverse to the corresponding widths define the characteristic times

$$T_1 \equiv \frac{1}{\gamma_1}, \quad T_2 \equiv \frac{1}{\gamma_2}, \quad T_2^* \equiv \frac{1}{\gamma_*}, \quad T_3 \equiv \frac{1}{\gamma_3}, \quad (48)$$

among which T_1 is the spin-lattice relaxation time, T_2 the spin-spin dephasing time, T_2^* the inhomogeneous dephasing time, and T_3 the resonator ringing time. To be more cautious, it is worth noting that, in our case, the width γ_* is due to local spin fluctuations which is only one of the possible mechanisms of inhomogeneous broadening. The latter arises also owing to crystalline defects, hyperfine interactions, and other inhomogeneities³⁵ that are not included in our consideration. Therefore, here T_2^* is of the order of T_2 , both of them being related to dipole interactions, and so $\gamma_* \sim \gamma_2$. The existence of the small parameters (46) means that the oscillation period

$$T_0 \equiv \frac{2\pi}{\omega_0} \ll \min\{T_1, T_2, T_2^*, T_3\} \quad (49)$$

is the shortest time as compared to the characteristic times (48).

To check the properties (3) and (4), we have to take the limit in Eqs. (35) and (42)–(44) by putting zero all small parameters (46) and (47) and, respectively, φ_0 and φ . This procedure yields the limits

$$\frac{dh}{dt} \rightarrow -\omega^2 \int_0^t h(\tau) d\tau - 2i\alpha_0\omega_0(u - u^*),$$

$$\frac{du}{dt} \rightarrow i\omega_0 u, \quad \frac{ds}{dt} \rightarrow 0, \quad \frac{d}{dt}|u|^2 \rightarrow 0,$$

which shows that u and h are to be treated as fast, while s and $|u|^2$ as slow variables. The second of these limits also shows that the adiabatic approximation is not appropriate when u is not zero.

At the next step we have to consider the slow variables as quasi-integrals of motion for fast variables. The corresponding equations (42) and (35), with the notation

$$u = x - iy, \quad s = z, \quad (50)$$

where z is kept as a fixed parameter, can be written in the form

$$\frac{dx}{dt} = -\gamma_2 x + \omega_\varphi y - \varphi_2 z,$$

$$\frac{dy}{dt} = -\omega_\varphi x - \gamma_2 y + (\gamma_3 h + \varphi_1) z, \quad (51)$$

$$\frac{dh}{dt} = -2\gamma_3 h - \omega^2 \int_0^t h(\tau) d\tau - 4\alpha_0 \frac{dx}{dt} + \gamma_3 f,$$

in which

$$\omega_\varphi \equiv \omega_0 - \varphi_0$$

is the shifted frequency and the stochastic field

$$\varphi = \varphi_1 - i\varphi_2$$

is separated into its real and imaginary parts. The initial conditions to (51) are

$$x(0) = x_0, \quad y(0) = y_0, \quad h(0) = 0.$$

It is remarkable that the system of three integro-differential equations (51), under fixed z , is linear, and thus can be solved exactly by employing, e.g., the method of the Laplace transforms. Equivalently, differentiating the last of the equations in (51), we may convert (51) into a linear system of five ordinary differential equations, which is again exactly solvable by means of either the method of the Laplace transforms or the matrix methods.

The exact solution of (51) is so cumbersome that it is not pleasure to write it down explicitly. Fortunately, we can simplify it by using the existence of the small parameters (46) and (47). Such a simplification can be done directly by, first, finding an exact solution of (51) and, second, performing

some expansions in small parameters. However, this direct way is extremely tedious and does not provide insight into the physics of the made simplifications. The same final result can be obtained in another way which is much less wearisome and more physically clear, and which is explained in Appendix B.

For definiteness, we take the electromotive force E_f in the standard form

$$E_f = E_0 \cos \omega t, \quad (52)$$

which gives for the driving force (32)

$$f = f_0 \cos \omega t, \quad f_0 \equiv \frac{c\mu E_0}{nA_0 \hbar \gamma_3^2}. \quad (53)$$

Solving (51), we obtain the fast variable

$$u = u_s + u_\varphi + u_f \quad (54)$$

consisting of three terms. The first,

$$u_s = (c_1 e^{i\Omega_\varphi t} + c_2 e^{-i\Omega_\varphi t}) e^{-\Gamma_\varphi t}, \quad (55)$$

corresponds to spin oscillations. The second,

$$u_\varphi = x_\varphi - iy_\varphi, \quad (56)$$

is caused by the local random fields. And the third,

$$u_f = (d_1 e^{i\omega t} + d_2 e^{-i\omega t})(1 - e^{-\gamma_3 t}), \quad (57)$$

is due to the driving force (53).

The resonator field

$$h = h_s + h_f \quad (58)$$

is given by the sum of the term

$$h_s = \frac{\omega_\varphi}{\gamma_3} \left[i(\alpha_1 + i\alpha_2)u^* - i(\alpha_1 - i\alpha_2)u + \frac{2}{\omega_\varphi}(\alpha_1\varphi_2 + \alpha_2\varphi_1)z \right], \quad (59)$$

induced by spin motion, and of the field

$$h_f = \frac{f_0}{4} \left(1 - i\frac{\gamma_3}{\omega} \right) (e^{i\omega t} + e^{-i\omega t})(1 - e^{-\gamma_3 t}), \quad (60)$$

produced by the electromotive force. All necessary notation for the expressions (54)–(60) is presented in Appendix B.

Note that the factors $(1 - e^{-\gamma_3 t})$ in (57) and (60) describe the retardation in the interaction of spins and the resonator.

At the next step of the method displayed in Sec. II, we have to substitute the fast variables (54) and (58) into Eqs. (43) and (44) for the slow variables,

$$s = z, \quad |u| = v, \quad (61)$$

averaging the right-hand sides of (43) and (44) over the asymptotic period of fast oscillations and also over a distribution of stochastic fields characterized by a probability measure μ_φ . The asymptotic period, according to the definition (9), is just (49). Let us denote the double averaging of a function $F = F_\varphi(t)$, over the asymptotic period and over stochastic fields, as

$$\langle\langle F \rangle\rangle \equiv \int \left[\frac{1}{T_0} \int_0^{T_0} F_\varphi(t) dt \right] d\mu_\varphi. \quad (62)$$

Since φ_0 is real and $\varphi = \varphi_1 - i\varphi_2$ is complex, there are three independent real components of the stochastic fields; thence, the differential measure $d\mu_\varphi$ can be written as the product

$$d\mu_\varphi = d\mu(\varphi_0) d\mu(\varphi_1) d\mu(\varphi_2).$$

It is customary to model the distribution of local dipole fields in spin systems by a Gaussian distribution.^{3,35} Accepting this and assuming, for simplicity, that each distribution of φ_ν , with $\nu=0,1,2$, has the same width γ_* , we get

$$d\mu(\varphi_\nu) = \frac{1}{\sqrt{2\pi}} \exp\left\{-\frac{1}{2}\left(\frac{\varphi_\nu}{\gamma_*}\right)^2\right\} \frac{d\varphi_\nu}{\gamma_*}.$$

Accomplishing the averaging (62), we will take into account the existence of the small parameters (46) and (47). The basic formulas that are met in the course of averaging the right-hand sides of (43) and (44) are assembled in Appendix C. Averaging the coupling functions we have

$$\begin{aligned} \alpha &\equiv \langle\langle \alpha_1 \rangle\rangle = \alpha_0 \left(\frac{\gamma_3}{\omega_0} \right) \frac{\pi(\gamma_2 - \gamma_3)^2}{(\gamma_2 - \gamma_3)^2 + \Delta^2}, \\ \beta &\equiv \langle\langle \alpha_2 \rangle\rangle = \alpha_0 \left(\frac{\gamma_3}{\omega_0} \right) \frac{\pi(\gamma_2 - \gamma_3)\Delta}{(\gamma_2 - \gamma_3)^2 + \Delta^2}. \end{aligned} \quad (63)$$

The average effective frequency and attenuation are, respectively,

$$\begin{aligned} \Omega &\equiv \langle\langle \Omega_\varphi \rangle\rangle = \omega_0(1 + \beta z), \\ \Gamma &\equiv \langle\langle \Gamma_\varphi \rangle\rangle = \gamma_2 + \alpha\omega_0 z, \end{aligned} \quad (64)$$

where an expansion in powers of the small parameters in (63) is used.

To write the evolution equations for the slow variables (61) in a compact form, we shall use some notation. Introduce the effective coupling parameter

$$g \equiv \alpha \frac{\omega_0}{\gamma_2} = \alpha_0 \left(\frac{\gamma_3}{\gamma_2} \right) \frac{\pi(\gamma_2 - \gamma_3)^2}{(\gamma_2 - \gamma_3)^2 + \Delta^2}. \quad (65)$$

Define the damping

$$\begin{aligned} \gamma_s &\equiv \frac{f_0 \gamma_3^2}{8\omega_0} \left\{ x_0 + 2\pi y_0 + \frac{2\omega_0 z}{\Delta^2 + \gamma_2^2} [x_0(\beta\Delta - \alpha\gamma_2) \right. \\ &\quad \left. + y_0(\alpha\Delta + \beta\gamma_2)] \right\} \end{aligned} \quad (66)$$

appearing when calculating the correlator $\langle\langle u_s h_f \rangle\rangle$ and also the attenuation

$$\begin{aligned} \gamma_f &= \frac{f_0^2 \gamma_3^4}{32\omega_0^2(\Delta^2 + \gamma_2^2)} \left\{ \left(1 + \frac{8\pi^2}{3} \right) \gamma_2 - 2\pi\Delta + \frac{\omega_0 z}{\Delta^2 + \gamma_2^2} [(\alpha \right. \\ &\quad \left. - 2\pi\beta)(\Delta^2 - \gamma_2^2) + 2\gamma_2\Delta(\beta + 2\pi\alpha)] \right\} \end{aligned} \quad (67)$$

resulting from the calculation of the correlator $\langle\langle u_f h_f \rangle\rangle$.

Thus, the averaging of the right-hand sides of Eqs. (43) and (44), in compliance with (62), leads to the equations

$$\frac{dz}{dt} = g\gamma_2 w - \gamma_s - \gamma_1(z - \zeta) - \gamma_f z,$$

$$\frac{dw}{dt} = -2\gamma_2 w - 2(g\gamma_2 w - \gamma_s)z + 2\gamma_f z^2 \quad (68)$$

for the slow variables, where

$$w \equiv v^2 - 2\varepsilon_* z, \quad \varepsilon_* \equiv \frac{\gamma_*^2}{\omega_0^2}. \quad (69)$$

The quantities (66) and (67) characterize the relaxation of the magnetization owing to the action of the resonator field (60) formed by the driving force (53). Note that $\gamma_s \equiv 0$ for the incoherent initial condition, when $u_0 \equiv x_0 - iy_0 = 0$. The squared amplitude of the driving force (53), remembering (34), can be written as

$$f_0^2 = \frac{8\alpha_0 E_0^2}{\hbar \gamma_3^2 R N}. \quad (70)$$

This shows that $f_0 \sim 1/\sqrt{N}$. Consequently, for the attenuations (66) and (67) we have $\gamma_s \sim 1/\sqrt{N}$ and $\gamma_f \sim 1/N$. These values for a macroscopic sample with $N \sim 10^{23}$ should be negligibly small.

In particular, if the electromotive force (52) corresponds to a resonance mode of the thermal Nyquist noise of the resonator, then³ for its amplitude we have

$$E_0^2 = \frac{\hbar \omega}{2\pi} \gamma_3 R \coth \frac{\hbar \omega}{2k_B T}, \quad (71)$$

where k_B is the Boltzmann constant and T the temperature. For ω in the radio-frequency region, typical of spin systems, (71) simplifies to

$$E_0^2 \approx \frac{\gamma_3}{\pi} R k_B T \left(\frac{\hbar \omega}{k_B T} \ll 1 \right).$$

Whence, for the amplitude in (70) we get

$$f_0^2 = \frac{8\alpha_0 k_B T}{\pi \hbar \gamma_3 N} \quad (\text{Nyquist noise}). \quad (72)$$

Substituting (72) into (66) and (67), we again come to the conclusion that these attenuations for a macroscopic sample are negligible. We shall exemplify this by numerical estimates in Sec. VI.

The conclusion that the radiation field of the coil does not provide a microscopic relaxation mechanism, so that γ_s and γ_f can be neglected in the equations for slow variables, is in complete agreement with the statement of Bloembergen and Pound¹⁹ that a homogeneous magnetic field, such as exists in the coil, will never produce the initial thermal relaxation in a macroscopic sample.

Let us acknowledge that γ_s and γ_f are negligibly small as compared to γ_2 . In addition, at low temperatures, characteristic of experiments,¹⁰⁻¹⁵ the spin-lattice damping is also much smaller than the spin-spin dephasing parameter. Thus, we have

$$\frac{\gamma_s}{\gamma_2} \ll 1, \quad \frac{\gamma_f}{\gamma_2} \ll 1, \quad \frac{\gamma_1}{\gamma_2} \ll 1. \quad (73)$$

Taking into consideration (73), the slow-variable equations in (68) can be contracted to

$$\frac{dz}{dt} = g\gamma_2 w, \quad \frac{dw}{dt} = -2\gamma_2 w(1 + gz). \quad (74)$$

The equations in (74) can be solved exactly in the following way. Notice that the effective attenuation Γ , with notation (65), acquires the form

$$\Gamma = \gamma_2(1 + gz). \quad (75)$$

Using (75) in (74), we obtain

$$\frac{d\Gamma}{dt} = (g\gamma_2)^2 w, \quad \frac{dw}{dt} = -2\Gamma w.$$

Differentiating here the first equation, we come to $\ddot{\Gamma} + 2\Gamma\dot{\Gamma} = 0$, which yields

$$\frac{d\Gamma}{dt} + \Gamma^2 = \gamma_0^2,$$

where γ_0 is an integration constant. This is the Riccati equation whose solution is

$$\Gamma = \gamma_0 \tanh\left(\frac{t-t_0}{\tau_0}\right) \quad \left(\tau_0 \equiv \frac{1}{\gamma_0}\right),$$

where t_0 , having the meaning of a delay time, is another integration constant. From (75) we have

$$z = \frac{\gamma_0}{g\gamma_2} \tanh\left(\frac{t-t_0}{\tau_0}\right) - \frac{1}{g}, \quad (76)$$

and from the first equation in (74) we find

$$w = \left(\frac{\gamma_0}{g\gamma_2}\right)^2 \operatorname{sech}^2\left(\frac{t-t_0}{\tau_0}\right).$$

The functions z and w are the exact solutions of (74). For the slow variable v , the relation (69) gives

$$v^2 = \left(\frac{\gamma_0}{g\gamma_2}\right)^2 \operatorname{sech}^2\left(\frac{t-t_0}{\tau_0}\right) + 2\varepsilon_* z. \quad (77)$$

As is seen, τ_0 is an effective relaxation time.

The integration constants γ_0 and t_0 are to be found from the initial conditions

$$z(0) = z_0, \quad v(0) = v_0. \quad (78)$$

From (76), (77), and (78) we obtain

$$\begin{aligned} \gamma_0^2 &= \Gamma_0^2 + (g\gamma_2)^2 (v_0^2 - 2\varepsilon_* z_0), \\ \Gamma_0 &\equiv \gamma_2(1 + gz_0), \quad \gamma_0\tau_0 = 1, \end{aligned} \quad (79)$$

and the delay time

$$t_0 = \frac{\tau_0}{2} \ln \left| \frac{\gamma_0 - \Gamma_0}{\gamma_0 + \Gamma_0} \right|. \quad (80)$$

So all integration constants in the solutions (76) and (77) for

the slow variables are defined. The corresponding solutions for the fast variables are obtained by substituting (76) and (77) into the sums (54) and (58).

V. RELAXATION REGIMES

Depending on the initial conditions and system parameters, one can distinguish several qualitatively different relaxation regimes. The advantage of dealing with analytical solutions, as compared to numerical solutions, is that there are explicit formulas allowing direct investigation. When the problem contains many parameters, as in the considered case, the detailed numerical analysis of the solutions by varying the numerous parameters becomes excessively laborious if not impossible. At the same time it may happen that not all parameters are equally important, but only some of them or some their combinations. A striking example of this kind is presented by the problem considered here. Really, despite the great number of various parameters, characterizing the spin system coupled with a resonator, the solutions of the evolution equations contain only several constants, the main one of which is the effective coupling parameter (65). The general qualitative classification of different relaxation regimes can be done by varying only three quantities: the coupling parameter g , the initial polarization z_0 , and the initial transverse magnetization v_0 . The latter defines the level of initial coherence imposed on the system.

First of all, one can easily observe that if there is neither initial polarization nor initial coherence, then (74) has only the trivial solution

$$z = v = 0 \quad (z_0 = v_0 = 0). \quad (81)$$

Therefore, the necessary and sufficient condition for the existence of nontrivial solutions is a nonzero initial magnetization

$$m_0^2 \equiv z_0^2 + v_0^2 > 0. \quad (82)$$

The relation between the effective relaxation time τ_0 and the spin-spin dephasing time T_2 depends on the value of gm_0 . Namely,

$$\begin{aligned} \tau_0 &\approx T_2 \quad (gm_0 \leq 1), \\ \tau_0 &< T_2 \quad (gm_0 > 1), \end{aligned} \quad (83)$$

which follows from (79) under the assumption that $g\varepsilon_* \ll 1$. The latter inequality is justified owing to the definition of ε_* in (69) as of a small parameter of second order with respect to (46).

The delay time (80) can have either negative or positive sign depending on the value of gz_0 :

$$\begin{aligned} t_0 &\leq 0 \quad (gz_0 \geq -1), \\ t_0 &> 0 \quad (gz_0 < -1). \end{aligned} \quad (84)$$

If $t_0 \leq 0$, then the maximum of the transverse magnetization (77) occurs at $t = 0$. In this case, since $gz_0 \geq -1$, then $\Gamma_0 > 0$, which means that the amplitude of the fast variable u decreases with time. When $t_0 > 0$, then the maximum of (77), i.e., the maximum of coherence, occurs at $t = t_0$. In this

situation, as far as $gz_0 < -1$, we have $\Gamma_0 < 0$, which leads to the increase of the amplitude of u . The negative sign of the attenuation Γ_0 means that the system acts as a generator.

Varying the quantities gz_0 and gv_0 , we may distinguish seven qualitatively different relaxation regimes.

(i) *Free induction*:

$$\begin{aligned} g|z_0| < 1, \quad 0 < gv_0 < 1, \\ t_0 < 0, \quad \tau_0 \approx T_2. \end{aligned} \quad (85)$$

This is the standard case of free nuclear induction, with the maximal coherence imposed at $t=0$ and relaxation time T_2 . The coupling with a resonator plays no principal role. Note that the conditions of the upper line and lower line in (85) are not independent, but one line follows from another, in compliance with (83) and (84). However, we write down the relations between effective parameters, as well as those between characteristic times, to make the classification more physically transparent.

(ii) *Collective induction*:

$$\begin{aligned} gz_0 > -1, \quad gv_0 > 1, \\ t_0 < 0, \quad \tau_0 < T_2. \end{aligned} \quad (86)$$

This case differs from the free induction by an essential role of the coupling with the resonator, which is sufficiently strong to develop collective effects leading to the shortening of the relaxation time τ_0 . When $gv_0 \gg 1$, then $\tau_0 \ll T_2$. But as in the previous case, the maximal coherence is that which is imposed at $t=0$.

(iii) *Free relaxation*:

$$\begin{aligned} g|z_0| < 1, \quad v_0 = 0, \\ t_0 < 0, \quad \tau_0 \approx T_2. \end{aligned} \quad (87)$$

The initial polarization z_0 and the coupling parameter g are not sufficiently high for the appearance of self-organized coherence. At the same time, there is no imposed coherence. The relaxation process is mainly incoherent being due to the local random fields.

(iv) *Collective relaxation*:

$$\begin{aligned} gz_0 > 1, \quad v_0 = 0, \\ t_0 < 0, \quad \tau_0 < T_2. \end{aligned} \quad (88)$$

The difference with the previous case is that the positive initial polarization and the coupling parameter now are high, so that collective effects shorten the relaxation time. However, the initial state is close to a stationary one, and the change of v , being again due to the local fields, is too small to yield a noticeable coherence.

(v) *Weak superradiance*:

$$\begin{aligned} -2 < gz_0 < -1, \quad v_0 = 0, \\ t_0 > 0, \quad \tau_0 \approx T_2. \end{aligned} \quad (89)$$

The negative initial polarization corresponds to an inverted system. The value of this polarization and that of the coupling parameter g are sufficient to make the delay time posi-

tive and to develop a weak coherence, as a result of incipient self-organization. But the latter is not yet enough strong to shorten the relaxation time.

(vi) *Pure superradiance*:

$$\begin{aligned} gz_0 < -2, \quad v_0 = 0, \\ t_0 > 0, \quad \tau_0 < T_2. \end{aligned} \quad (90)$$

The system is prepared in a strongly nonequilibrium state with a high negative polarization. The coupling with a resonator is also strong. No initial coherence is imposed on the system. The coherence arises as a purely self-organized process started by local stochastic fields and developed owing to the resonator feedback field.

(vii) *Triggered superradiance*:

$$\begin{aligned} gz_0 < -1, \quad gv_0 > 1, \\ t_0 > 0, \quad \tau_0 < T_2. \end{aligned} \quad (91)$$

The initial polarization is negative and the coupling with a resonator is strong enough, so that the collective behavior of spins, tight with each other through the feedback field, is important. But the relaxation is triggered by an imposed initial coherence. Therefore, this is a collective but not purely self-organized process.

In this classification, three regimes, free induction, collective induction, and triggered superradiance, are triggered by initial coherence thrust upon spins, that is, by setting $v_0 \neq 0$. Local random fields do not play an important role. Such kinds of regimes can be described by the Bloch equations. Other four relaxation regimes, free relaxation, collective relaxation, weak superradiance, and pure superradiance, are initiated solely by local fields. No initial coherence is involved, i.e., $v_0 = 0$. The Bloch equations cannot treat these four regimes.

Organizing the above classification, we separated qualitatively different relaxation types. As is clear, there can be intermediate kinds of relaxation in between these regimes. For example, the case when

$$gz_0 < -1, \quad 0 < gv_0 < 1$$

is between the weak superradiance and triggered superradiance. In principle, everywhere in this classification the condition $v_0 = 0$ can be replaced by $gv_0 < 1$, to include the intermediate regimes. However, it seems reasonable to distinguish, first, different physical reasons causing different relaxation mechanisms.

In the process of relaxation, the polarization (76), starting at $z = z_0$, tends to

$$z \approx \frac{\gamma_0}{g}(T_2 - \tau_0) \quad (t \gg t_0). \quad (92)$$

If the initial polarization z_0 is negative, then (92) shows that a noticeable polarization reversal to a positive value occurs for the case when $\tau_0 < T_2$, that is, for pure and triggered superradiance; also, it may happen at collective induction, though then the initial polarization is not high. The highest initial polarization is needed for pure superradiance. The corresponding polarization threshold is twice as large as that for weak superradiance or triggered superradiance. Equation

(92) shows as well that there can be no essential reversal of polarization from positive to negative values.

It is illustrative to consider more in detail two limiting situations, when the coupling of the spin system with a resonator is either weak or strong. Start with the weak coupling limit $g \ll 1$. Then for the relaxation width and relaxation time, from (79), we get

$$\begin{aligned}\gamma_0 &\approx \gamma_2 \left[1 + gz_0 + \frac{g^2}{2}(v_0^2 - 2\varepsilon_* z_0) \right], \\ \tau_0 &\approx T_2 \left[1 - gz_0 - \frac{g^2}{2}(v_0^2 - 2z_0^2 - 2\varepsilon_* z_0) \right].\end{aligned}\quad (93)$$

For the delay time (80) we have

$$t_0 \approx \frac{\tau_0}{2} \ln \left| \frac{g^2}{4}(v_0^2 - 2\varepsilon_* z_0) \right|. \quad (94)$$

The behavior of polarization is

$$z \approx z_0 + \frac{g}{2}(v_0^2 - 2\varepsilon_* z_0)(1 - e^{-2\gamma_2 t}). \quad (95)$$

When $g|z_0| < 1$ and $gv_0 < 1$, we have the case of free induction (85), if $v_0 \neq 0$. And if $v_0 = 0$, then we have free relaxation (87) with

$$\begin{aligned}t_0 &\approx \frac{\tau_0}{2} \ln \left| \frac{g^2}{2} \varepsilon_* z_0 \right|, \\ z &\approx z_0 - g\varepsilon_* z_0(1 - e^{-2\gamma_2 t}).\end{aligned}\quad (96)$$

The latter regime is entirely due to local fields, since if ε_* were zero, then $z \approx z_0$ and there would be no relaxation.

In the strong coupling limit $g \gg 1$, from (79) we find

$$\gamma_0 \approx \gamma_2 \left(g \sqrt{m_0^2 - 2\varepsilon_* z_0} + \frac{z_0}{\sqrt{m_0^2 - 2\varepsilon_* z_0}} \right).$$

This, using the inequality $\varepsilon_* \ll 1$, can be reduced to

$$\begin{aligned}\gamma_0 &\approx gm_0 \gamma_2 \left[1 + \frac{z_0}{gm_0^2} - \left(1 - \frac{z_0}{gm_0^2} \right) \frac{\varepsilon_* z_0}{m_0^2} \right], \\ \tau_0 &\approx \frac{T_2}{gm_0} \left[1 - \frac{z_0}{gm_0^2} + \left(1 - \frac{3z_0}{gm_0^2} \right) \frac{\varepsilon_* z_0}{m_0^2} \right].\end{aligned}\quad (97)$$

The delay time (80) takes the form

$$t_0 \approx \frac{\tau_0}{2} \ln \left| \frac{m_0^2(m_0 - z_0)(gm_0 - 1) - (gm_0^2 - z_0)\varepsilon_* z_0}{m_0^2(m_0 + z_0)(gm_0 + 1) - (gm_0^2 - z_0)\varepsilon_* z_0} \right|. \quad (98)$$

For the final polarization (92) at $t \gg t_0$ we obtain

$$z \approx m_0 - \frac{1}{g} \left[1 - \frac{z_0}{m_0} + \left(1 - \frac{z_0}{gm_0^2} \right) g\varepsilon_* z_0 \right]. \quad (99)$$

These formulas for $gv_0 > 1$, depending on the value of gz_0 , correspond either to collective induction (86) or to triggered superradiance (91). When $v_0 = 0$, we come, again depending on the value of gz_0 , to collective relaxation (88), weak superradiance (89), or pure superradiance (90).

Note that if $gz_0 < -1$, then for any v_0 the maximal coherence is reached at $t = t_0 > 0$, when

$$z(t_0) \approx -\frac{1}{g}, \quad v(t_0) \approx m_0. \quad (100)$$

To better emphasize the role of local fields, let us analyze the case when there is no initial coherence, that is,

$$m_0 = |z_0|, \quad v_0 = 0, \quad (101)$$

and $g|z_0| > 1$. Then

$$\begin{aligned}\gamma_0 &\approx g|z_0| \gamma_2 \left[1 + \frac{1}{gz_0} - \left(1 - \frac{1}{gz_0} \right) \frac{\varepsilon_*}{z_0} \right], \\ \tau_0 &\approx \frac{T_2}{g|z_0|} \left[1 - \frac{1}{gz_0} + \left(1 - \frac{3}{gz_0} \right) \frac{\varepsilon_*}{z_0} \right].\end{aligned}\quad (102)$$

The delay time (80) becomes

$$t_0 \approx \frac{\tau_0}{2} \ln \left| \frac{(|z_0| - z_0)(g|z_0| - 1) - (gz_0 - 1)\varepsilon_*}{(|z_0| + z_0)(g|z_0| + 1) - (gz_0 - 1)\varepsilon_*} \right|. \quad (103)$$

The final polarization (92) at $t \gg t_0$ is

$$z \approx |z_0| - \frac{1}{g|z_0|} [|z_0| - z_0 + (gz_0 - 1)\varepsilon_*]. \quad (104)$$

Consider separately the cases of positive and negative initial polarizations. When the latter is positive, i.e.,

$$z_0 = |z_0|, \quad (105)$$

then the delay time (103) and final polarization (104) are

$$\begin{aligned}t_0 &\approx \frac{\tau_0}{2} \ln \left| \frac{(gz_0 - 1)\varepsilon_*}{2z_0(gz_0 + 1) - (gz_0 - 1)\varepsilon_*} \right|, \\ z &\approx z_0 - \left(1 - \frac{1}{gz_0} \right) \varepsilon_* \quad (t \gg t_0).\end{aligned}\quad (106)$$

Simplifying this for asymptotically large $gz_0 \gg 1$, and keeping in mind that $\varepsilon_* \ll 1$, we have

$$\begin{aligned}t_0 &\approx \frac{\tau_0}{2} \ln \left| \frac{\varepsilon_*}{2z_0} \right|, \quad \tau_0 \approx \frac{T_2}{gz_0}, \\ z &\approx z_0 - \varepsilon_* \quad (t \gg t_0).\end{aligned}\quad (107)$$

Formulas (106) and (107) correspond to collective relaxation (88) due to local fields.

Pass to the case of the negative initial polarization

$$z_0 = -|z_0|. \quad (108)$$

Then, for the delay time (103) and final polarization (104) we find

$$\begin{aligned}t_0 &\approx \frac{\tau_0}{2} \ln \left| \frac{2|z_0|(g|z_0| - 1) + (g|z_0| + 1)\varepsilon_*}{(g|z_0| + 1)\varepsilon_*} \right|, \\ z &\approx |z_0| - \frac{2}{g} + \left(1 + \frac{1}{g|z_0|} \right) \varepsilon_* \quad (t \gg t_0).\end{aligned}\quad (109)$$

This can describe weak superradiance (89) or pure superradiance (90). Under the inequalities $g|z_0| \gg 1$ and $\varepsilon_* \ll 1$ the latter expressions change to

$$t_0 \approx \frac{\tau_0}{2} \ln \left| \frac{2z_0}{\varepsilon_*} \right|, \quad \tau_0 \approx \frac{T_2}{g|z_0|},$$

$$z \approx |z_0| - \frac{2}{g} + \varepsilon_* \quad (t \ll t_0), \quad (110)$$

which corresponds to pure superradiance (90). The origin of this phenomenon is completely due to local fluctuating fields.

An interesting question is, which part of dipole interaction is mainly responsible for starting the relaxation process in the regime of pure superradiance? Looking at Eqs. (42) and (43), we see that it is the random field φ which initiates the process, while φ_0 only shifts the oscillation frequency. The stochastic field φ represents the local fields (39), which are related to the terms b_{ij} and c_{ij} of the dipole interactions. These terms are called nonsecular dipole interactions contrary to a_{ij} , which is called the secular dipole interaction.² In this way, it is the nonsecular dipole interactions that originate an initial relaxation and, consequently, the pure spin superradiance.

The results obtained make it possible to give one more justification for the term "spin superradiance." For a system of N nuclei an effective number of radiators may be defined as

$$N_{\text{eff}} \equiv \frac{m_0 N}{S},$$

where m_0 is the initial magnetization introduced in (82) and S is nuclear spin. Averaging the power of current

$$P_{\varphi}(t) \equiv Rj^2 = N \frac{\hbar \gamma_3^2}{4\alpha_0} h^2,$$

according to (62), we have

$$P(t) \equiv \langle \langle P_{\varphi}(t) \rangle \rangle = N(\alpha^2 + \beta^2) \frac{\hbar \gamma_2^2}{\alpha_0} v^2.$$

The average current power for a superradiant regime has a maximum at $t = t_0 > 0$, where $v(t_0) = m_0$, in compliance with (100). Therefore,

$$P(t_0) \sim m_0^2 \sim N_{\text{eff}}^2.$$

Also, as is seen from (97), the radiation time

$$\tau_0 \sim m_0^{-1} \sim N_{\text{eff}}^{-1}.$$

The situation when the radiation pulse is proportional to the number of radiators squared, and the radiation time is inversely proportional to this number, is characteristic of superradiance.

Note that the intensity of magnetodipole radiation, $I(t)$, as a function of time, behaves similarly to the current power

$P(t)$ but contains a small factor making $I(t) \ll P(t)$, so that $P(t)$ is much easier to measure.^{17,19}

VI. NUMERICAL ESTIMATES

The aim of the present paper is not to discuss some particular experiments but rather to give the general picture of possible relaxation processes. Nevertheless, the general qualitative picture can be better understood if illustrated by quantitative estimates. For this purpose, let us accept the values of parameters typical of experiments¹¹⁻¹⁵ with proton-rich materials, such as propanadiol $C_3H_8O_2$, butanol C_4H_9OH , and ammonia NH_3 . Employing the method of dynamic nuclear polarization, it is possible to polarize spins to a level of polarization reaching almost 100%. The samples polarized in this way are good examples of metastable nuclear magnets. The lifetime of such metastable materials at low temperature is very long. This time, T_1 , is related to the spin-lattice relaxation time. The order of its magnitude is given by the relation $T_1 \sim (a/\Delta l)^2 T_2$, in which $a \sim 10^{-8}$ cm is the mean distance between spins, $\Delta l \sim 10^{-5} a \sim 10^{-13}$ cm is the coefficient of linear magnetostriction, and T_2 is the spin-spin relaxation time. Whence, $T_1/T_2 \sim 10^{10}$.

The spin-spin relaxation time is characterized by dipole interactions yielding $T_2 \sim \hbar a^3 / \mu^2 \sim 10^{-5}$ s. Consequently, $T_1 \sim 10^5$ s. The relaxation time T_2^* , related to local spin fluctuations, is also due to dipole interactions because of which $T_2^* \sim 10^{-5}$ s.

In principle, there exists another longitudinal relaxation time due to the interaction of spins through the common electromagnetic field formed under the magnetodipole spin radiation. This time, which will be denoted by T_1' , to distinguish it from the spin-lattice relaxation time T_1 , can be estimated as $T_1' \sim (\lambda/a)^2 T_2$, where λ is the radiation wavelength. For the external magnetic field $H_0 \sim 10^4$ G, spins radiate in the radio-frequency region with $\omega_0 \sim 10^8$ s⁻¹, and thus with the wavelength $\lambda \sim 10^2$ cm. This gives $T_1'/T_2 \sim 10^{20}$ or $T_1' \sim 10^{15}$ s. As far as $T_1'/T_1 \sim 10^{10}$, the longitudinal relaxation is practically due to the spin-lattice interactions only. The interaction through the radiation electromagnetic field is so weak, as compared to dipole interactions, that it does not play any role. This drastically distinguishes spin systems from atomic and molecular ones exhibiting superradiance. In the latter systems, the effective interaction through the common radiation field is not only important but serves as the basic mechanism for the appearance of strong collective correlations and coherence.

The resonator ringing time T_3 in the case of quasiresonance, when $\omega \sim \omega_0 \sim 10^8$ s⁻¹, and for the quality factor $Q \sim 10^2$ is $T_3 \sim 10^{-6}$ s. The time of fast oscillations, defined in (49), is $T_0 \sim 10^{-8}$ s; so it is really the shortest among other characteristic times.

The damping parameters corresponding to the characteristic times in (48) are $\gamma_1 \sim 10^{-5}$ s⁻¹, $\gamma_2 \sim 10^5$ s⁻¹, $\gamma_2^* \sim 10^5$ s⁻¹, and $\gamma_3 \sim 10^6$ s⁻¹. In this way, for the small parameters in (46) we have

$$\frac{\gamma_1}{\omega_0} \sim 10^{-13}, \quad \frac{\gamma_2}{\omega_0} \sim 10^{-3}, \quad \frac{\gamma_2^*}{\omega_0} \sim 10^{-3}, \quad \frac{\gamma_3}{\omega} \sim 10^{-2}.$$

The coupling constant (34), owing to the relations $\hbar\gamma_2 \sim \mu^2/a^3$ and $\rho a^3 = 1$, where ρ is the particle density, is $\alpha_0 \sim \pi\eta\gamma_2/\gamma_3 \sim 10^{-1}$. The average coupling functions in (63) are $\alpha \sim \gamma_2/\omega_0 \sim 10^{-3}$ and $\beta \leq \gamma_2/\omega_0 \sim 10^{-3}$. In the case of exact resonance, when $\Delta = 0$, the latter is identically zero, $\beta = 0$. Thus, α and β are also small parameters.

The maximal value of the effective coupling parameter (65) is of the order of π^2 . Therefore it varies in the interval $0 \leq g \leq 10$.

Consider the dampings (66) and (67) caused by the action of the electromotive force corresponding to a resonance mode of the thermal Nyquist noise with the amplitude (71). The typical temperature in experiments¹¹⁻¹⁵ is $T \sim 0.1$ K. As far as $k_B T \sim 10^{-5}$ eV and $\hbar\omega \sim 10^{-7}$ eV, we have $\hbar\omega/k_B T \sim 10^{-2}$; hence the approximation (72) is justified. Using $\hbar\gamma_3 \sim 10^{-9}$ eV, for the forcing-field amplitude we find $f_0 \sim 10^2/\sqrt{N}$. Then, for the damping (66) we get $\gamma_s \propto (10^5/\sqrt{N}) \text{ s}^{-1}$. In the case of passive initial conditions, when $x_0 = y_0 = 0$, the value of (66) is exactly zero, $\gamma_s = 0$. Expression (67) yields $\gamma_f \sim (10^7/N) \text{ s}^{-1}$. For a sample of about 1 cm^3 the number of protons is $N \sim 10^{23}$. Hence, the thermal-noise forcing field has the amplitude $f_0 \sim 10^{-10}$, so for the damping (66) and (67) we get $\gamma_s \leq 10^{-7} \text{ s}^{-1}$ and $\gamma_f \sim 10^{-16} \text{ s}^{-1}$. These quantities are so much less than γ_2 that there is no reason to keep them in the equations. This also concerns γ_1 . Really, the relations in (73) are

$$\frac{\gamma_s}{\gamma_2} \sim 10^{-12}, \quad \frac{\gamma_f}{\gamma_2} \sim 10^{-21}, \quad \frac{\gamma_1}{\gamma_2} \sim 10^{-10}.$$

Therefore, the thermal Nyquist noise of a resonator has no influence on the spin dynamics in a macroscopic sample.

One might ask a question: What should be the size of a sample on which the resonator thermal noise could produce a noticeable effect? This would happen if $\gamma_s \sim \gamma_2$, which gives $N \sim 1$, or when $\gamma_f \sim \gamma_2$, from where $N \sim 100$. For $N > 100$ the Nyquist noise is practically of no importance.

The method of solving the equations, used in the present paper, makes it possible to take into account the retardation effects, related to the appearance of factors like $(1 - e^{-\gamma_3 t})$. These effects are important for the correct description of relaxation processes. For example, the threshold of initial polarization for superradiance, weak or triggered, as follows from (89) and (91), is $z_0 \sim -1/g$. As a percentage, for spin 1/2 and $g \sim 20$, this means that the superradiance threshold is -10% . Respectively, the threshold of pure superradiance, given in (90), is -20% . These values are in agreement with experiments,¹¹⁻¹⁵ while, if we would neglect the retardation replacing the factor $(1 - e^{-\gamma_3 t})$ by 1, then for the superradiance threshold we would get $-\gamma_2/\alpha_0\omega_0 = -\pi\gamma_3/g\omega_0 \sim 10^{-3}$. As a percentage, this makes -0.1% , which is unrealistically small.

In the regime of pure spin superradiance, the characteristic times τ_0 and t_0 can be estimated from (110). Since $\tau_0 \sim T_2/g|z_0|$, taking $g|z_0| \sim 10$, we find the radiation time $\tau_0 \sim 10^{-6}$ s. The local-field parameter, defined in (69), is $\varepsilon_* \sim 10^{-6}$. Whence, for the delay time we obtain $t_0 \sim (3-5)\tau_0$, that is, $t_0 \sim 10^{-6} - 10^{-5}$ s. The reversed final polarization, according to (110), can reach 90%. Note that the problem of the fast polarization reversal of proton solid-

state targets is of great practical importance for the study of scattering in high- and intermediate-energy physics.¹⁵ The phenomenon of spin superradiance can be used to achieve the desired fast repolarization.

VII. DISCUSSION

A method has been presented for treating the nonlinear systems of differential or integro-differential equations with stochastic terms. Such terms simulate local random fields of interacting statistical systems.

The use of the method has made it possible to give a detailed analysis of nonlinear spin relaxation in nuclear magnets coupled with resonance electric circuits. The consideration is based on a microscopic Hamiltonian with realistic dipole spin interactions.

Seven qualitatively different regimes of relaxation are found. Three of them, free induction, collective induction, and triggered superradiance, are initiated by primary coherence imposed upon spins, with local fields playing no essential role. These cases can be reasonably treated by the Bloch equations. Four other regimes, free relaxation, collective relaxation, weak superradiance, and pure superradiance, are triggered by local fields. The cases when no primary coherence is imposed cannot be treated by the standard Bloch equations.

Dealing with superradiant relaxation in nuclear magnets, one may naturally remember superradiance occurring in atomic systems. The latter has been studied in detail both experimentally and theoretically. There exists a number of books and surveys on atomic superradiance. We cite here only one of the recent reviews¹⁸ where further references can be found. Although the relaxation process in an atomic system with photon transitions between two selected levels shares a few formal similarities with the spin relaxation, there are much more principal differences, both in physics and mathematics, between the superradiant phenomena in spin and atomic systems. It is instructive to emphasize the corresponding analogies and differences.

The main formal similarities are as follows: An atomic system is often put into a resonator cavity discriminating one or several resonance modes. Pure spin superradiance is analogous to the superfluorescence of atomic systems, both being the processes of coherent self-organization. In the collection of induced atomic dipoles, superfluorescence evolves from a state of complete inversion of atoms, and is due to effective dipole-dipole interactions induced by the exchange of photons between atoms. These induced dipole-dipole interactions of atoms are similar in their form, to real dipole interactions of spins, since both of them can be expressed through Pauli matrices. Using the pseudospin representation for two-level systems, the atomic relaxation can be described by an effective Bloch equation. The relaxation regimes in spin systems, which can also be described by the Bloch equations, should be similar to their atomic counterparts. This concerns, for instance, the triggered spin superradiance and triggered atomic superradiance, or the free nuclear induction and free atomic induction.

However, many of these similarities are only formal and even can be misleading, since there are several principal differences between atomic and spin systems.

(i) The relaxation of induced atomic dipoles is physically different from that of polarized nuclear spins.

The process of relaxation of an inverted atomic system is accompanied by radiation into free space. This electric dipole radiation can be of quite high intensity.

The relaxation in a spin system coupled with a resonator, as has been pointed out by Friedberg and Hartmann,¹⁶ is a radiationless process involving merely nonradiative transfer of energy from the sample to the coil, in which the energy is dissipated.

(ii) The role of a low-quality cavity for atoms and of a resonant circuit for spins is drastically different.

For atoms a low-quality cavity is used whose aim is to define a sufficiently narrow band of preferential modes. In the following process the cavity does not play an active role in developing coherence, but superfluorescence evolves due to induced dipole-dipole interactions.

In the case of nuclear spins there is no need to discriminate modes, as far as, for spin one-half, the sole mode is already defined by the Zeeman splitting of the spin energy in an external magnetic field. In other words, one could say that a cavity for atoms does the same, that is, defines working modes, as an external magnetic field for spins. But a high-quality resonator for spins plays another and crucial role during the whole process of spin relaxation, the resonator feedback field being the driving force for developing the collective spin evolution.

(iii) The effective interactions between induced atomic dipoles and the real interactions between spins have nothing common with each other neither in their physical origin or in their role in collective processes.

The interaction between induced atomic dipoles describes an effective interaction of atoms through the common radiation field. It is just this interaction which is solely responsible for the formation of such a collective coherent state as superfluorescence.

The dipole interaction between spins is a real interaction existing without any radiation field. The actual form of this interaction is quite different from that of the effective atomic interaction. Related to this is the principal difference of their roles. As is known from the theory of such phenomena as nuclear magnetic resonance or nuclear induction, the dipole spin interactions can never be responsible for organizing a collective spin evolution. Vice versa, the dipole spin interactions are responsible for dephasing the motion of spins destroying their coherence. At the same time, the effective interactions of spins through the common radiation field is too weak for playing any noticeable role.

(iv) The origin of the evolving coherence is absolutely different for atomic and for spin systems.

In an inverted atomic system, the process starts with incoherent spontaneous radiation of atoms. This creates an effective interaction between atoms through the common radiation field. The arising interaction collectivizes the atoms leading to their coherent radiation.

In a polarized spin system, the relaxation begins owing to random spin fluctuations caused by spin-spin interactions. Then the resonator feedback field appears organizing the coherence of spin motion, while the spin interactions at this stage start playing a destructive role. Thus, the effective interaction of atoms through the common radiation field is

analogous, not to the real spin-spin interaction, but to the effective coupling between spins through the resonator feedback field. The formal analogy between the effective interaction of induced atomic dipoles and the real interaction of spins, based on the fact that both these interactions are expressed through the Pauli matrices, is misleading.

(v) The mathematical description of atomic and spin relaxation requires, in general, different methods.

The radiation in atomic systems is usually treated by the slow-varying-amplitude approximation which is somewhat similar to the averaging method although the latter is more regular and mathematically grounded.

For the system of nuclear spins, the complication comes from the necessity of taking into account local random fields without which it is impossible to describe several important regimes, e.g., such as pure spin superradiance. Also, there is an additional equation, the Kirchhoff equation, characterizing the coupling between the spin sample and a resonant electric circuit. This coupling, for a correct description of spin relaxation, cannot be reduced to a simple proportionality of the sample magnetization to the resonator field. The method developed in the present paper seems to be the most adequate for considering the spin relaxation in nonequilibrium magnets with resonators.

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APPENDIX A: EVOLUTION EQUATIONS

The evolution equations (40) and (41) are derived in the following way.

Expressing the spin operators $S_i^x = \frac{1}{2}(S_i^- + S_i^+)$ and $S_i^y = i/2(S_i^- - S_i^+)$ through the ladder operators $S_i^- = S_i^x - iS_i^y$ and $S_i^+ = S_i^x + iS_i^y$, we may cast the dipole interaction energy (27) into the form

$$H_{ij} = a_{ij} \left(S_i^z S_j^z - \frac{1}{2} S_i^+ S_j^- \right) + b_{ij} S_i^+ S_j^+ + b_{ij}^* S_i^- S_j^- \\ + 2(c_{ij} S_i^+ + c_{ij}^* S_i^-) S_j^z,$$

in which the interaction coefficients are

$$a_{ij} \equiv \frac{\mu^2}{r_{ij}^3} (1 - 3 \cos^2 \vartheta_{ij}),$$

$$b_{ij} \equiv -\frac{3\mu^2}{4r_{ij}^3} \sin^2 \vartheta_{ij} \exp(-i2\varphi_{ij}),$$

$$c_{ij} \equiv -\frac{3\mu^2}{4r_{ij}^3} \sin(2\vartheta_{ij}) \exp(-i\varphi_{ij}),$$

where ϑ_{ij} and φ_{ij} are the spherical angles of \vec{n}_{ij} . These interaction coefficients enjoy the symmetry property

$$a_{ij} = a_{ji}, \quad b_{ij} = b_{ji}, \quad c_{ij} = c_{ji}.$$

With the commutation relations $[S_i^z, S_j^\pm] = \pm \delta_{ij} S_i^\pm$ and $[S_i^+, S_j^-] = 2\delta_{ij} S_i^z$ we have the Heisenberg equations of motion

$$\begin{aligned} i\hbar \frac{d}{dt} S_i^- &= \sum_{j(\neq i)}^N \left\{ a_{ij} \left(S_i^- S_j^z + \frac{1}{2} S_i^z S_j^- \right) - 2b_{ij} S_i^z S_j^+ \right. \\ &\quad \left. + c_{ij} (S_i^- S_j^+ - 2S_i^z S_j^z) + c_{ij}^* S_i^- S_j^- \right\} - \mu H_0 S_i^- \\ &\quad + \mu H S_i^z, \end{aligned}$$

$$\begin{aligned} i\hbar \frac{d}{dt} S_i^z &= \sum_{j(\neq i)}^N \left\{ \frac{a_{ij}}{4} (S_i^- S_j^+ - S_i^+ S_j^-) + b_{ij} S_i^+ S_j^+ - b_{ij}^* S_i^- S_j^- \right. \\ &\quad \left. + (c_{ij} S_i^+ - c_{ij}^* S_i^-) S_j^z \right\} + \frac{\mu}{2} H (S_i^- - S_i^+) \\ &\quad - i\hbar \gamma_1 (S_i^z - \zeta_i), \end{aligned}$$

which are supplemented by a term taking into account spin-lattice interactions leading to the longitudinal damping γ_1 , and ζ_i being a stationary value of the spin z component.

Of course, the evolution equations could be written shorter, invoking some tensor notation. However, we need here their explicit form in order to separate different parts of dipole spin interactions. As analysis shows, these different interaction terms play principally different physical role.

Averaging the above equations of motion, in the framework of the corrected mean-field approximation, yields

$$\begin{aligned} i \frac{du_i}{dt} &= -(\omega_0 + i\gamma_2) u_i + \gamma_3 h s_i + \frac{1}{\hbar} \sum_{j(\neq i)}^N \left\{ \frac{a_{ij}}{2} (s_i u_j + 2u_i s_j) - 2b_{ij} s_i u_j^* + c_{ij} (u_i u_j^* - 2s_i s_j) + c_{ij}^* u_i u_j \right\}, \\ i \frac{ds_i}{dt} &= \frac{1}{2} \gamma_3 h (u_i - u_i^*) + \frac{1}{\hbar} \sum_{j(\neq i)}^N \{ a_{ij} (u_i u_j^* - u_i^* u_j) + b_{ij} u_i^* u_j^* - b_{ij}^* u_i u_j + (c_{ij} u_i^* - c_{ij}^* u_i) s_j \} - i\gamma_1 (s_i - \zeta_i). \end{aligned}$$

From here, for the arithmetic averages (37), with the local fields (38) and (39), we obtain (40) and (41).

APPENDIX B: FAST VARIABLES

The formal solution of the last equation in (51) can be written as the sum $h = h_s + h_f$ in which the first term is a feedback field induced in the resonator by moving spins and the second term is a resonator field formed by driving forces. The resonator feedback field may be presented either as the convolution

$$h_s = -4\alpha_0 \int_0^t \frac{d}{d\tau} x(t-\tau) W(\tau) d\tau$$

or as the Stieltjes integral

$$h_s = -4\alpha_0 \int_0^t W(t-\tau) dx(\tau),$$

and the resonator forcing field is given by the convolution

$$h_f = \gamma_3 \int_0^t W(t-\tau) f(\tau) d\tau,$$

where the transfer function is

$$W(t) = \left(\cos \omega_3 t - \frac{\gamma_3}{\omega_3} \sin \omega_3 t \right) e^{-\gamma_3 t},$$

with $\omega_3 \equiv \sqrt{\omega^2 - \gamma_3^2}$.

The action of the resonator field h on the spin system involves, as follows from (51), the small parameter γ_3 . Neglecting this parameter reduces the first two equations in (51) to the system

$$\frac{dx}{dt} \cong -\gamma_2 x + \omega_\varphi y - \varphi_2 z,$$

$$\frac{dy}{dt} \cong -\omega_\varphi x - \gamma_2 y + \varphi_1 z.$$

The solution to the latter is

$$x \cong (a_0 \cos \omega_\varphi t + b_0 \sin \omega_\varphi t) e^{-\gamma_2 t} + \frac{\varphi_1}{\omega_\varphi} z,$$

$$y \cong (b_0 \cos \omega_\varphi t - a_0 \sin \omega_\varphi t) e^{-\gamma_2 t} + \frac{\varphi_2}{\omega_\varphi} z,$$

where

$$a_0 = x_0 - \frac{\varphi_1}{\omega_\varphi} z, \quad b_0 = y_0 - \frac{\varphi_2}{\omega_\varphi} z.$$

Employing this gives the feedback field

$$h_s = -\frac{2}{\gamma_3} \left[\alpha_1 \frac{dx}{dt} + \alpha_2 \omega_\varphi \left(x - \frac{\varphi_1}{\omega_\varphi} z \right) \right],$$

in which

$$\alpha_1 = \frac{\alpha_0 \gamma_3 (\gamma_2 - \gamma_3)}{(\gamma_2 - \gamma_3)^2 + (\Delta + \varphi_0)^2} [e^{(\gamma_2 - \gamma_3)t} - 1],$$

$$\alpha_2 = \frac{\alpha_0 \gamma_3 (\Delta + \varphi_0)}{(\gamma_2 - \gamma_3)^2 + (\Delta + \varphi_0)^2} [e^{(\gamma_2 - \gamma_3)t} - 1].$$

If in the expression for h_s we put $\alpha_1=0, \alpha_2=\text{const}$, we return to the static-coupling approximation, while if we put $\alpha_1=\text{const}, \alpha_2=0$, then we get the dynamic-coupling approximation.⁹ However, in general, $\alpha_1=\alpha_1(t)$ and $\alpha_2=\alpha_2(t)$ are nonzero functions of time. The temporal dependence of the coupling functions portrays the retardation due to a gradual switching on of the coupling between the spins and resonator. Really, as is seen, at the initial moment the coupling is absent,

$$\alpha_1(0) = \alpha_2(0) = 0.$$

Using the first of the equations in (51) for h_s yields

$$h_s = \frac{2}{\gamma_3} [(\alpha_1 \gamma_2 - \alpha_2 \omega_\varphi)x - \alpha_1 \omega_\varphi y + (\alpha_1 \varphi_2 + \alpha_2 \varphi_1)z].$$

Substituting this back into (51) reduces the system of three integro-differential equations to the system of two ordinary differential equations

$$\frac{dx}{dt} = -\gamma_2 x + \omega_\varphi y - \varphi_2 z,$$

$$\begin{aligned} \frac{dy}{dt} = & -(\omega_\varphi - 2\alpha_1 \gamma_2 z + 2\alpha_2 \omega_\varphi z)x - (\gamma_2 + 2\alpha_1 \omega_\varphi z)y \\ & + (\varphi_1 + 2\alpha_1 \varphi_2 z + 2\alpha_2 \varphi_1 z + \gamma_3 h_f)z \end{aligned}$$

for the fast variables.

There is no problem in solving the latter system, which gives

$$x = (a_1 \cos \Omega_\varphi t + b_1 \sin \Omega_\varphi t) e^{-\Gamma_\varphi t} + x_\varphi + x_f,$$

$$y = (a_2 \cos \Omega_\varphi t + b_2 \sin \Omega_\varphi t) e^{-\Gamma_\varphi t} + y_\varphi + y_f,$$

where the first parts describe the spin oscillations with the effective frequency

$$\Omega_\varphi = \omega_\varphi (1 - \alpha_1^2 z^2 + 2\alpha_2 z)^{1/2},$$

effective attenuation

$$\Gamma_\varphi = \gamma_2 + \alpha_1 z \omega_\varphi,$$

and coefficients

$$a_1 = x_0 - x_\varphi, \quad a_2 = y_0 - y_\varphi,$$

$$\begin{aligned} b_1 = & \frac{\omega_\varphi}{\Omega_\varphi} (y_0 + \alpha_1 z x_0) - \frac{\omega_\varphi z}{\Omega_\varphi^2 + \Gamma_\varphi^2} \left[(1 + 2\alpha_2 z) \frac{\Gamma_\varphi}{\Omega_\varphi} \varphi_1 \right. \\ & \left. + \left(\frac{\Omega_\varphi}{\omega_\varphi} + \alpha_1 z \frac{\Gamma_\varphi}{\Omega_\varphi} \right) \varphi_2 \right], \end{aligned}$$

$$\begin{aligned} b_2 = & -\frac{\omega_\varphi}{\Omega_\varphi} [(1 + 2\alpha_2 z)x_0 + \alpha_1 z y_0] \\ & + \frac{\omega_\varphi z}{\Omega_\varphi^2 + \Gamma_\varphi^2} \left\{ (1 + 2\alpha_2 z) \left(\frac{\Omega_\varphi}{\omega_\varphi} + \alpha_1 z \frac{\Gamma_\varphi}{\Omega_\varphi} \right) \varphi_1 \right. \\ & \left. + \left[2\alpha_1 z \left(\frac{\Omega_\varphi}{\omega_\varphi} + \alpha_1 z \frac{\Gamma_\varphi}{\Omega_\varphi} \right) - (1 + 2\alpha_2 z) \frac{\Gamma_\varphi}{\Omega_\varphi} \right] \varphi_2 \right\}; \end{aligned}$$

the terms

$$\begin{aligned} x_\varphi = & \frac{\omega_\varphi z}{\Omega_\varphi^2 + \Gamma_\varphi^2} \left[(1 + 2\alpha_2 z) \varphi_1 - \left(\frac{\Gamma_\varphi}{\omega_\varphi} - \alpha_1 z \right) \varphi_2 \right], \\ y_\varphi = & \frac{\omega_\varphi z}{\Omega_\varphi^2 + \Gamma_\varphi^2} \left\{ (1 + 2\alpha_2 z) \left(\frac{\Gamma_\varphi}{\omega_\varphi} - \alpha_1 z \right) \varphi_1 \right. \\ & \left. + \left[1 + 2\alpha_2 z + 2\alpha_1 z \left(\frac{\Gamma_\varphi}{\omega_\varphi} - \alpha_1 z \right) \right] \varphi_2 \right\} \quad (\text{B1}) \end{aligned}$$

are originated by the local random fields, and the last terms

$$\begin{aligned} x_f = & \gamma_3 \int_0^t G_1(t - \tau) h_f(\tau) d\tau, \\ y_f = & \gamma_3 \int_0^t G_2(t - \tau) h_f(\tau) d\tau \quad (\text{B2}) \end{aligned}$$

are due to the resonator forcing field, the Green functions being

$$G_1(t) = z \frac{\omega_\varphi}{\Omega_\varphi} \sin \Omega_\varphi t e^{-\Gamma_\varphi t},$$

$$G_2(t) = z \cos \Omega_\varphi t e^{-\Gamma_\varphi t} - \alpha_1 z G_1(t).$$

In this way, the fast variable u , defined by Eq. (42), takes the form of (54) where

$$c_1 = \frac{1}{2} (a_1 - b_2) - \frac{i}{2} (b_1 + a_2),$$

$$c_2 = \frac{1}{2} (a_1 + b_2) + \frac{i}{2} (b_1 - a_2).$$

To find an explicit expression for u_f , induced by an electromotive force E_f , entering into the right-hand side of the Kirchhoff equation (30), we need to concretize the form of E_f . Accepting for the latter the standard expression $E_f = E_0 \cos \omega t$ for the driving force (32) we have (53). Then the convolution for h_f gives

$$h_f = \frac{f_0}{2} \left(\cos \omega t - \frac{\gamma_3}{\omega} \sin \omega t \right) (1 - e^{-\gamma_3 t}).$$

Substituting the resonator forcing field h_f into x_f and y_f , we get

$$x_f = (f_1 e^{i\omega t} + f_1^* e^{-i\omega t}) (1 - e^{-\gamma_3 t}),$$

$$y_f = (f_2 e^{i\omega t} + f_2^* e^{-i\omega t}) (1 - e^{-\gamma_3 t}),$$

where the coefficients are

$$f_1 = -\frac{f_0 \omega_\varphi \gamma_3 z}{8\Omega_\varphi(\Delta_\varphi^2 + \Gamma_\varphi^2)}(\Delta_\varphi + i\Gamma_\varphi),$$

$$f_2 = f_1 \left(i \frac{\Omega_\varphi}{\omega_\varphi} - \alpha_1 z \right),$$

and the effective detuning is $\Delta_\varphi \equiv \omega - \Omega_\varphi$. Therefore $u_f = x_f - iy_f$ transforms to (57) with the coefficients

$$d_1 = f_1 \left(1 + \frac{\Omega_\varphi}{\omega_\varphi} + i\alpha_1 z \right),$$

$$d_2 = f_1^* \left(1 - \frac{\Omega_\varphi}{\omega_\varphi} + i\alpha_1 z \right).$$

APPENDIX C: SLOW VARIABLES

Here we present the basic formulas for the averages defined in (62) and used in Sec. IV when deriving the equations for slow variables.

For the stochastic fields, with the Gaussian distribution in mind, we have

$$\langle\langle \varphi_0 \rangle\rangle = \langle\langle \varphi_1 \rangle\rangle = \langle\langle \varphi_2 \rangle\rangle = 0,$$

$$\langle\langle \varphi_0^2 \rangle\rangle = \langle\langle \varphi_1^2 \rangle\rangle = \langle\langle \varphi_2^2 \rangle\rangle = \gamma_*^2.$$

Note that, instead of defining a particular distribution, we could postulate the above properties of random fields.

In the following expressions the averaging (62) is accompanied by expansions in powers of small parameters (46):

$$\langle\langle e^{-\Gamma_\varphi t} \rangle\rangle \approx 1 - \frac{\pi\Gamma}{\omega_0},$$

$$\langle\langle e^{i(\Omega_\varphi + i\Gamma_\varphi)t} \rangle\rangle \approx \frac{\Omega - \omega_0 + i\Gamma}{\omega_0},$$

$$\langle\langle e^{i(\Delta - \gamma_2)t} \rangle\rangle \approx 1 + \frac{\pi}{\omega_0}(i\Delta - \gamma_2),$$

$$\langle\langle e^{i\omega t}(1 - e^{-\gamma_3 t}) \rangle\rangle \approx -i \frac{\gamma_3}{\omega_0},$$

$$\langle\langle e^{i\Delta t}(1 - e^{-\gamma_3 t}) \rangle\rangle \approx \pi \frac{\gamma_3}{\omega_0},$$

$$\langle\langle e^{i(\omega + \Omega_\varphi + i\Gamma_\varphi)t}(1 - e^{-\gamma_3 t}) \rangle\rangle \approx -i \frac{\gamma_3}{2\omega_0},$$

$$\langle\langle (1 - e^{-\gamma_3 t})^2 \rangle\rangle \approx \frac{4\pi^2 \gamma_3^2}{3\omega_0^2},$$

$$\langle\langle e^{2i\omega t}(1 - e^{-\gamma_3 t})^2 \rangle\rangle \approx (1 - 2\pi i) \frac{\gamma_3^2}{2\omega_0^2},$$

where Ω_φ and Γ_φ are defined in Appendix B, and Ω and Γ are given in (64).

We emphasize the importance of the factor $(1 - e^{-\gamma_3 t})$ responsible for the retardation effects.

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