Nonlinear spin relaxation in strongly nonequilibrium magnets

V. I. Yukalov
Institut für Theoretische Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany
and Bogolubov Laboratory of Theoretical Physics, Joint Institute for Nuclear Research, Dubna 141980, Russia
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A general theory is developed for describing the nonlinear relaxation of spin systems from a *strongly nonequilibrium* initial state, when, in addition, the sample is coupled to a resonator. Such processes are characterized by nonlinear stochastic differential equations. This makes these strongly nonequilibrium processes principally different from the spin relaxation close to an equilibrium state, which is represented by linear differential equations. The consideration is based on a realistic microscopic Hamiltonian including the Zeeman terms, dipole interactions, exchange interactions, and a single-site anisotropy. The influence of cross correlations between several spin species is investigated. The critically important function of coupling between the spin system and a resonant electric circuit is emphasized. The role of all main relaxation rates is analyzed. The phenomenon of self-organization of transition coherence in spin motion, from the quantum chaotic stage of incoherent fluctuations, is thoroughly described. Local spin fluctuations are found to be the triggering cause for starting the spin relaxation from an incoherent nonequilibrium state. The basic regimes of collective coherent spin relaxation are studied.

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I. INTRODUCTION

The problem of spin relaxation from a state close to equilibrium has a long history and is well studied, being related to the description of spin motion in the vicinity of different magnetic resonances. This type of spin relaxation is usually characterized by linear differential equations, such as Bloch equations. The theory of spin motion close to equilibrium has been expounded in numerous literature, among which it would be possible to mention several good books.[1–7]

Essentially nonlinear spin motion arises if the system is prepared in a strongly nonequilibrium initial state, e.g., with magnetization opposite to an external magnetic field, and, in addition, is coupled to a resonator. Such nonlinear dynamics are commonly treated by the Bloch equations supplemented by the Kirchhoff equation for a resonator electric circuit.[8–11] However, the phenomenological Bloch equations do not allow for the elucidation of different physical processes involved in the behavior of the system and are not able to describe several, probably the most interesting, self-organized regimes of spin motion, as was demonstrated in Refs. 12–14. Some physical models, based on microscopic spin Hamiltonians, have also been considered, whose survey can be found in recent reviews.[15,16] But in each of these models one standardly studies only some particular substances and considers only a part of spin interactions, mainly secular dipole-dipole interactions, and one takes into account only some of the known attenuation processes. At the same time, it is evident that taking care of only particular model elements can easily lead to wrong physical conclusions, since real physical materials always include several different characteristics competing with each other. The study of nonlinear spin relaxation is of paramount importance not solely owing to its theoretical beauty but also because it can be employed in a variety of applications, such as the measurement of materials parameters, ultrafast repolarization of solid-state targets, creation of sensitive field detectors, usage in quantum computing and others, as is discussed in reviews.[15,16] One of the major possible applications is in achieving the regime of superradiant operation by spin masers.[13,17–19] Punctuated nonlinear dynamics of spin assemblies can also be a new tool for information processing.[20]

The aim of the present paper is to develop a general theory of nonlinear spin relaxation, being based on a realistic microscopic Hamiltonian including, in addition to the Zeeman terms, the main spin interactions, and taking account of the different major mechanisms of spin attenuation. By considering just some limited models, it is easy to come to false conclusions and to predict fictitious physical effects that by no means can exist in real materials. It is only by carefully treating different competing mechanisms that one can derive reliable physical implications.

II. BASIC SPIN HAMILTONIAN

Keeping in mind the applicability of the theory to a wide class of spin systems, we start with a rather general Hamiltonian including the major spin interactions the most often met in magnetic materials.[1–7,21–23] Let us consider a solid sample containing N vector spins $\mathbf{S}_i$ enumerated by the index $i=1,2,\ldots,N$. The spin operators $\mathbf{S}_i$ can represent any particle of spin S, starting from $S=1/2$ to very high spin values. These can be nuclear or electronic spins, as in the standard problems of nuclear or electronic spin resonances.[1–7,15] Magnetic molecules, forming molecular magnets, can possess various spins ranging from $S=1/2$ up to $S=27/2$, as is reviewed in Refs. 16, 19, and 24–26. Bose-Einstein condensates of dilute gases (see reviews[27–30]), being placed in optical lattices can form localized clouds with an effective spin per site of order 10^2 or 10^3. Spin dynamics (mainly linear) is an intensively developing field of research, called spintronics.[31]

The Hamiltonian of a spin system can, generally, be separated into two parts,

$$\hat{H} = \sum_i \hat{H}_i + \frac{1}{2}\sum_{i\neq j} \hat{H}_{ij}, \tag{1}$$

the first term being related to individual spins, while the second representing spin interactions. The single-spin Hamiltonian

$$\hat{H}_i = -\mu_0 \mathbf{B}\cdot\mathbf{S}_i - D(S_i^z)^2 \tag{2}$$

consists of the Zeeman energy and the energy of the single-site magnetic anisotropy. Here $\mu_0 \equiv \hbar\gamma_S$, with γ_S being the gyromagnetic ratio of a particle with spin S. For electronic spins, $\mu_0 < 0$, while for nuclear spins μ_0 can be either positive or negative. The total magnetic field

$$\mathbf{B} = B_0\mathbf{e}_z + (B_1 + H)\mathbf{e}_x \tag{3}$$

contains external longitudinal, B_0, and transverse, B_1, magnetic fields, and also a feedback field H of a resonator, if the sample is coupled to a resonant electric circuit. The anisotropy parameter D is positive for an easy-axis anisotropy and negative in the case of an easy-plane anisotropy.

The interaction Hamiltonian

$$\hat{H}_{ij} = \sum_{\alpha\beta} D_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta - J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j \tag{4}$$

includes dipole and exchange interactions. The dipolar tensor is

$$D_{ij}^{\alpha\beta} = \frac{\mu_0^2}{r_{ij}^3}(\delta_{\alpha\beta} - 3n_{ij}^\alpha n_{ij}^\beta), \tag{5}$$

where $\alpha,\beta = x,y,z$ and

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

This tensor enjoys the properties

$$\sum_\alpha D_{ij}^{\alpha\alpha} = 0, \quad \sum_{j(\neq i)} D_{ij}^{\alpha\beta} = 0, \tag{6}$$

of which the first is exact and the second one is asymptotically exact for a macroscopic sample with a large number of spins $N \gg 1$. A positive exchange integral corresponds to ferromagnetic interactions and negative, to antiferromagnetic interactions.

It is convenient to represent the Hamiltonians through the ladder spin operators $S_i^\pm \equiv S_i^x \pm iS_i^y$. Then the single-spin term (2) writes as

$$\hat{H}_i = -\mu_0 B_0 S_i^z - \frac{1}{2}\mu_0(B_1 + H)(S_i^+ + S_i^-) - D(S_i^z)^2. \tag{7}$$

With the notation

$$a_{ij} \equiv D_{ij}^{zz}, \quad b_{ij} \equiv \frac{1}{4}(D_{ij}^{xx} - D_{ij}^{yy} - 2iD_{ij}^{xy}),$$
$$c_{ij} \equiv \frac{1}{2}(D_{ij}^{xz} - iD_{ij}^{yz}), \tag{8}$$

the interaction Hamiltonian (4) transforms to

$$\hat{H}_{ij} = a_{ij}\left(S_i^z S_j^z - \frac{1}{2}S_i^+ S_j^-\right) + b_{ij}S_i^+ S_j^+ + b_{ij}^* S_i^- S_j^- + 2c_{ij}S_i^+ S_j^z + 2c_{ij}^* S_i^- S_j^z - J_{ij}(S_i^+ S_j^- + S_i^z S_j^z). \tag{9}$$

The interaction parameters $a_{ij} = a_{ji}$, $b_{ij} = b_{ji}$, and $c_{ij} = c_{ji}$ are symmetric and have the property

$$\sum_{j(\neq i)} a_{ij} = \sum_{j(\neq i)} b_{ij} = \sum_{j(\neq i)} c_{ij} = 0, \tag{10}$$

following from Eqs. (6).

The equations of motion for the spin operators are obtained from the Heisenberg equations and the commutation relations

$$[S_i^+, S_j^-] = 2\delta_{ij}S_i^z, \quad [S_i^z, S_j^\pm] = \pm\,\delta_{ij}S_i^\pm.$$

In order to represent the evolution equations in a compact form, it is convenient to introduce the local fields

$$\xi_0 \equiv \frac{1}{\hbar}\sum_{j(\neq i)} [a_{ij}S_j^z + c_{ij}^* S_j^- + c_{ij}S_j^+ + J_{ij}(S_i^z - S_j^z)], \tag{11}$$

$$\xi \equiv \frac{i}{\hbar}\sum_{j(\neq i)}\left[2c_{ij}S_j^z - \frac{1}{2}a_{ij}S_j^- + 2b_{ij}S_j^+ + J_{ij}(S_i^- - S_j^-)\right]$$

and the effective force

$$f \equiv -\frac{i}{\hbar}\mu_0(B_1 + H) + \xi. \tag{12}$$

There is a characteristic frequency, the Zeeman frequency, which we denote as

$$\omega_0 \equiv -\frac{\mu_0}{\hbar}B_0. \tag{13}$$

Then as the equations of motion for the spin operators, we obtain

$$\frac{dS_i^-}{dt} = -i(\omega_0 + \xi_0)S_i^- + fS_i^z + i\frac{D}{\hbar}(S_i^- S_i^z + S_i^z S_i^-), \tag{14}$$

with its Hermitian conjugate, and

$$\frac{dS_i^z}{dt} = -\frac{1}{2}(f^+ S_i^- + S_i^+ f). \tag{15}$$

The following description of spin dynamics will be based on these equations.

III. TRIGGERING SPIN FLUCTUATIONS

Suppose that the spin system is prepared in a strongly nonequilibrium state, being polarized along the z axis. What then could be the triggering mechanisms initiating spin motion and their relaxation to an equilibrium state? It is evident that imposing transverse magnetic fields would push the spins to move. But assume that there are no transverse magnetic fields at the initial time and no transverse coherence is imposed on the system. What then would initiate the spin motion? Here it is important to stress the role of local spin

waves as of the triggering mechanism for starting the spin relaxation.

The appearance of spin waves is due to the local fields (11). In order to consider spin waves, or more generally, spin fluctuations that arise in a state which is not necessarily equilibrium, it is appropriate to work with the operator equations (14) and (15). Let us define the operator deviation

$$\delta S_i^\alpha \equiv S_i^\alpha - \langle S_i^\alpha \rangle \tag{16}$$

from an average $\langle S_i^\alpha \rangle$, which is not necessarily an equilibrium average, but which can be an average over a nonequilibrium statistical operator, though such that $\langle S_i^\alpha \rangle$ weakly depends on the index i, because of which it can be taken out of the sums in Eqs. (11). Then, owing to Eqs. (10), we have

$$\xi_0 = \frac{1}{\hbar} \sum_{j(\neq i)} [a_{ij} \delta S_j^z + c_{ij}^* \delta S_j^- + c_{ij} \delta S_j^+ + J_{ij}(\delta S_i^z - \delta S_j^z)], \tag{17}$$

$$\xi = \frac{i}{\hbar} \sum_{j(\neq i)} \left[2c_{ij} \delta S_j^z - \frac{1}{2} a_{ij} \delta S_j^- + 2b_{ij} \delta S_j^+ + J_{ij}(\delta S_i^- - \delta S_j^-) \right],$$

which demonstrates that these local fields really correspond to local spin fluctuations.

To emphasize the role of the spin fluctuations, let us set $B_1 = H = 0$, that is, looking at the case when the transverse fields do not initiate the spin motion. And, respectively, let $\langle S_i^\pm \rangle = 0$, but the longitudinal polarization be finite, $\langle S_i^z \rangle \neq 0$. Then $S_i^\pm = \delta S_i^\pm$. The behavior of spin fluctuations is characterized by linearizing Eqs. (14) and (15) with respect to the operator deviations (16). The linearization of the single-site anisotropy term in Eq. (14) has to be done so that we satisfy the known exact relations for $S = 1/2$ and $S \to \infty$, which can be represented[16,32] as

$$S_i^- S_i^z + S_i^z S_i^- = \left(2 - \frac{1}{S} \right) \langle S_i^z \rangle S_i^- . \tag{18}$$

Introduce the single-site anisotropy frequency

$$\omega_D \equiv (2S - 1) \frac{D}{\hbar} \tag{19}$$

and the effective spin frequency

$$\omega_s \equiv \omega_0 - \omega_D \frac{\langle S_i^z \rangle}{S}, \tag{20}$$

where ω_0 is defined in Eq. (13). Then, linearizing Eqs. (14) and (15), we find

$$\frac{d}{dt} S_i^- = -i \omega_s S_i^- + \langle S_i^z \rangle \xi, \quad \frac{d}{dt} \delta S_i^z = 0. \tag{21}$$

The second of these equations, under the initial condition $\delta S_i^z(0) = 0$, gives $\delta S_i^z = 0$.

Now let us employ the Fourier transforms for the interactions

$$a_{ij} = \frac{1}{N} \sum_k a_k e^{i\mathbf{k}\cdot\mathbf{r}_{ij}}, \quad a_k = \sum_{j(\neq i)} a_{ij} e^{-i\mathbf{k}\cdot\mathbf{r}_{ij}},$$

with the analogous transforms for b_{ij} and J_{ij}, and for the spin operators

$$S_j^\pm = \sum_k S_k^\pm e^{\mp i\mathbf{k}\cdot\mathbf{r}_j}, \quad S_k^\pm = \frac{1}{N} \sum_j S_j^\pm e^{\pm i\mathbf{k}\cdot\mathbf{r}_j}.$$

Using the notation

$$\alpha_k \equiv \omega_s + \frac{1}{\hbar} \left(\frac{a_k}{2} + J_k - J_0 \right) \langle S_i^z \rangle, \quad \beta_k \equiv \frac{2}{\hbar} b_k \langle S_i^z \rangle, \tag{22}$$

from the first of Eqs. (14), we obtain

$$\frac{d}{dt} S_k^- = -i \alpha_k S_k^- + i \beta_k S_k^+ . \tag{23}$$

Looking for the solution of the latter equation in the form

$$S_k^- = u_k e^{-i\omega_k t} + v_k^* e^{i\omega_k t},$$

we find the spectrum of spin waves

$$\omega_k = \sqrt{\alpha_k^2 - |\beta_k|^2}. \tag{24}$$

In the long-wave limit, one gets

$$\omega_k \simeq |\omega_s| \left[1 - \langle S_i^z \rangle \sum_{\langle j \rangle} \frac{a_{ij} + 2J_{ij}}{4\hbar\omega_s} (\mathbf{k}\cdot\mathbf{r}_{ij})^2 \right], \tag{25}$$

where $k \to 0$, and the summation is over the nearest neighbors.

In this way, in the spin system there are always transverse fluctuations, which can be named spin waves. The latter, as they have been described, are not necessarily the spin waves in an equilibrium state, as they are usually understood,[33] but are to be considered in a generalized sense. Under spin waves, we mean here just transverse spin fluctuations. It is these transverse fluctuations that are responsible for triggering the initial motion of polarized spins, when there are no external transverse magnetic fields. This is why these transverse spin fluctuations can be called triggering spin waves. Taking into account such quantum spin fluctuations makes it possible to describe the dynamical regimes of spin motion, which do not exist for classical Bloch equations. And it becomes possible to develop a detailed picture of how the transverse spin coherence arises from initially chaotic fluctuations. This self-organized process of coherence emerging from chaos is one of the most interesting and challenging problems of spin dynamics.

IV. SPIN EVOLUTION EQUATIONS

The equations of motion (14) and (15) for spin operators are highly nonlinear. The nonlinearity comes from two sources. One is caused by the spin interactions accumulated in the local fluctuating fields (11). Another kind of nonlinearity enters through the effective force (12) containing feedback fields included in the term H. The treatment of the nonlinear spin dynamics will be done here by means of the

scale separation approach,[11–15,34] which is a generalization of the averaging technique[35] to stochastic differential equations.

Notice, first of all, that there are two different spatial scales. One of them is related to local fields (11) describing random spin fluctuations (17), which is characterized by a spatial length of the order of the mean interparticle distance a_0. At this length scale, chaotic quantum spin fluctuations prevail. Another length scale is the wavelength $\lambda \gg a_0$ corresponding to coherent effects associated with the characteristic spin rotation frequency ω_s. At the latter scale, coherent spin correlations are important. These two different length scales allow us to distinguish two types of operators. One type are the local fluctuating fields (11), that is, the variables ξ_0, ξ, and ξ^+, and another type are the spin operators S_i^-, S_i^+, and S_i^z. The former, responsible for local short-range fluctuations, can be represented by random variables,[2,5,11,16,36] while the latter keep track of long-range coherent effects. Respectively, it is convenient to define two sorts of averaging with respect to the corresponding variables. Then the statistical averaging over spin operators will be denoted by the single angle brackets $\langle \cdots \rangle$, while the averaging over the random local fields will be denoted by the double angle brackets $\langle\langle \cdots \rangle\rangle$. The latter, treating the chaotic local spin fluctuations as white noise, are defined as

$$\langle\langle \xi_0(t) \rangle\rangle = \langle\langle \xi(t) \rangle\rangle = 0, \quad \langle\langle \xi_0(t) \xi_0(t') \rangle\rangle = 2\gamma_3 \delta(t-t'),$$

$$\langle\langle \xi_0(t) \xi(t') \rangle\rangle = \langle\langle \xi(t) \xi(t') \rangle\rangle = 0, \quad \langle\langle \xi^*(t) \xi(t') \rangle\rangle = 2\gamma_3 \delta(t-t'), \tag{26}$$

where γ_3 is the width of inhomogeneous dynamic broadening.

It is worth stressing that the white-noise approximation (26) is not principal and could be generalized to taking into account a colored noise by including finite relaxation times. This, however, would result in much more complicated and cumbersome equations. It is therefore more convenient, following the ideas of the scale separation approach,[11–15] to separate in the temporal behavior of spin correlations two parts, fast and slow. The fast part is connected to the local spin fluctuations described by the spectrum of local spin waves (24). The characteristic frequencies of these fluctuations are defined by the near-neighbor spin coupling as well as by the applied external magnetic field. Here and in what follows, we assume that this external field is sufficiently strong, so that the fluctuation spectrum (24) is characterized by the frequencies of the order of the Zeeman frequency ω_0, which is essentially larger than the frequency terms due to spin interactions. With the time $2\pi/\omega_0$ being the shortest among all other characteristic times, the related fast spin fluctuations can be effectively treated as white noise, as is done in Eq. (26). The influence of spin correlations slowly decaying in time can be appropriately included into the transverse relaxation time T_2 determined by the strength of the spin-spin coupling allowing for dipolar as well as exchange interactions. This effective relaxation time will also be taken into account in the following consideration, together with the effect of line narrowing due to high spin polarization.[6]

Averaging over spin operators, because of their long-range role, one can employ the decoupling

$$\langle S_i^\alpha S_j^\beta \rangle = \langle S_i^\alpha \rangle \langle S_j^\beta \rangle \quad (i \neq j). \tag{27}$$

Though this looks like a mean-field approximation, one should not forget that the restricted averaging, denoted by the single angle brackets $\langle \cdots \rangle$, by definition, involves only the spin degrees of freedom, without touching the stochastic variables ξ_0 and ξ^*. Therefore the quantum fluctuations are not lost in decoupling (27) but are preserved because of the dependence of the spin averages $\langle S_i^\alpha \rangle$ on the random variables ξ_0 and ξ. Then decoupling (27) is termed the stochastic mean-field approximation.[11–16]

A special care is to be taken in considering the single-site term of Eq. (14). When averaging the latter, one has to preserve the exact limiting properties known for $S=1/2$ and $S \to \infty$. The corresponding decoupling, correctly interpolating between the exact limiting behaviors[16,19,32] is

$$\langle S_i^- S_i^z + S_i^z S_i^- \rangle = \left(2 - \frac{1}{S} \right) \langle S_i^- \rangle \langle S_i^z \rangle. \tag{28}$$

Thus, for $S=1/2$, expression (28) becomes zero, as it should be, and for $S \to \infty$, one has $2\langle S_i^- \rangle \langle S_i^z \rangle$, again in agreement with the correct asymptotic behavior.

Let us average the equations of motion (14) and (15) over the spin degrees of freedom, not touching the fluctuating random fields ξ_0 and ξ. Our aim is to obtain the evolution equations for the following variables: The *transition function*

$$u \equiv \frac{1}{SN} \sum_{i=1}^N \langle S_i^- \rangle, \tag{29}$$

describing the average rotation of transverse spin components; the *coherence intensity*

$$w \equiv \frac{1}{S^2 N(N-1)} \sum_{i \neq j}^N \langle S_i^+ S_j^- \rangle, \tag{30}$$

showing the level of coherence in the spin motion, and the *spin polarization*

$$s \equiv \frac{1}{SN} \sum_{i=1}^N \langle S_i^z \rangle, \tag{31}$$

defining the average polarization per particle.

In order to have the evolution equations representing realistic spin systems, but not just some unreasonable models, an accurate account must be taken of the main relaxation mechanisms. Being based on unrealistic models, omitting important existing attenuation processes, it would be easy to fall into the sin of predicting physical effects that in reality can never occur. We shall consider the following basic relaxation rates.

(1) *Spin-lattice longitudinal attenuation* γ_1, caused by spin-lattice interactions. The corresponding longitudinal relaxation time is $T_1 \equiv 1/\gamma_1$. For different materials, γ_1 can be of different order. At low temperature, when spin-phonon interactions are suppressed, the parameter γ_1 can be rather small. For instance, in polarized nuclear targets[16] at tempera-

ture of 1 K, one has $\gamma_1 \sim 10^{-5}$ s^{-1}. In molecular crystals below the blocking temperature of the order of 1 K, the spin-lattice rate can be between $\gamma_1 \sim 10^{-7}$ and 10^{-5} s^{-1} (see more details in Refs. 16 and 24–26). Being small, this relaxation parameter may not play an essential role at the initial stage of spin motion, however, it always plays a principal role at the late stages of spin relaxation.

(2) *Polarization pump rate* γ_1^*, which is added to γ_1 when the sample is subject to a permanent pump supporting a non-equilibrium level of the longitudinal spin polarization. This rate can be made much larger than γ_1. Thus, by means of dynamic nuclear polarization, the pump rate for nuclear spins in solids can be as large as $\gamma_1^* \sim 0.01$ and 10 s^{-1}.[16] The sum of γ_1 and γ_1^* will be denoted as

$$\Gamma_1 \equiv \gamma_1 + \gamma_1^*. \tag{32}$$

(3) *Spin dephasing rate* γ_2, due to spin-spin interactions. This rate has been calculated by many authors, and the generally accepted value[1–7] writes as

$$\gamma_2 = n_0 \rho \frac{\mu_0^2}{\hbar} \sqrt{S(S+1)}, \tag{33}$$

where $\rho \equiv N/V$ is density and n_0 is a coefficient approximately equal to the number of nearest neighbors. The process of spin dephasing is mainly due to dipolar forces. Exchange interactions slightly narrow the line width (33), yielding[4,6,21] a factor of about 0.8. The coefficient in Eq. (33) also depends on the type of lattice, so that the numerical factor here is approximate. The value of γ_2 is usually larger than that of γ_1. For example, in polarized solid targets[16] $\gamma_2 \sim 10^5$ s^{-1}, in molecular magnets[16,24] it is $\gamma_2 \sim 10^{10}$ s^{-1}. Inverse of γ_2 defines the spin dephasing time $T_2 \equiv 1/\gamma_2$.

(4) *Effective homogeneous broadening* $\gamma_2(s)$ takes into account a correction to the spin dephasing rate γ_2, appearing in the case of strongly polarized spin systems. Such a strong polarization can be achieved in magnetically ordered materials, by applying strong longitudinal magnetic fields, or by dynamic polarization techniques. This effective broadening reads as

$$\gamma_2(s) = \gamma_2(0)(1 - s^2), \quad \gamma_2(0) \equiv \gamma_2, \tag{34}$$

where s is an average spin polarization (31) and γ_2 is given by Eq. (33). The derivation of Eq. (34) is explained in Appendix A. Under weak polarization, when $s^2 \ll 1$, one has $\gamma_2(s) \simeq \gamma_2$.

(5) *Static inhomogeneous broadening* γ_2^* is due to various magnetic defects, crystalline defects, field gradients, and a variety of additional interactions always present in any real materials.[1–7,21,31] Very often the inhomogeneity develops in matter not because of externally incorporated defects, but being due to the internal properties, when a heterogeneous state is more thermodynamically stable than a homogeneous state.[37,38] This, e.g., happens in many colossal-magnetoresistance materials[39–41] and in high-temperature superconductors,[42–46] where there appears mesoscopic phase separation. In general, γ_2^* can be both smaller as well as larger than γ_2. However in the majority of cases, to a very good approximation $\gamma_2^* \sim \gamma_2$. Summarizing the homogeneous and inhomogeneous mechanisms, discussed above, we denote the overall transverse relaxation rate as

$$\Gamma_2 \equiv \gamma_2(1 - s^2) + \gamma_2^*. \tag{35}$$

(6) *Dynamic inhomogeneous broadening* γ_3 is caused by fast dynamic spin fluctuations, or the local spin waves, discussed in Sec. III. It comes into play through the stochastic averaging (26). The value of the broadening, due to local spin waves, is of the order or smaller than γ_2.[14–16,21] As is emphasized in Sec. III, this dynamic broadening is crucially important at the initial stage of spin relaxation, when there are no applied transverse fields.

(7) *Cross relaxation rates* arise when there are several spin species in the system. For example, if there are two types of spins, S and F, then the dynamic broadening for spin S becomes

$$\gamma_3 = \sqrt{\gamma_{SS}^2 + \gamma_{SF}^2}. \tag{36}$$

Cross correlations can influence other relaxation rates, especially if the Zeeman frequencies of the spins S and F are close to each other.[1–7,15,16]

(8) *Spin radiation rate* γ_r arises when there exist the so-called wave packets of strongly correlated spins interacting with each other through the common radiation field. The possibility of the appearance of such an electromagnetic friction was, first, noticed by Ginzburg[47] and later discussed by many authors (see, e.g., Ref. 48). This collective radiation rate is

$$\gamma_r = \frac{2}{3\hbar} \rho \mu_0^2 S (k L_s)^3, \tag{37}$$

where k is the wave vector of the radiating field and L_s is an effective linear size of a spin packet radiating coherently. Rate (37) has earlier been obtained[47,48] in the classical approximation. In Appendix B, we briefly sketch how this rate can be derived in a fully quantum-mechanical picture. It is important to stress that the existence of rate (37) presupposes the occurrence of monochromatic radiation with a well-defined constant spin frequency ω_s and wave vector k, and that the radiation wavelength is much larger than the linear size L_s of a spin packet, so that

$$k L_s \ll 1 \quad \left(k \equiv \frac{\omega_s}{c} \right). \tag{38}$$

If these conditions do not hold, no noticeable relaxation rate arises. And under the validity of these conditions, one has

$$\frac{\gamma_r}{\gamma_2} \approx 0.1 (k L_s)^3 \ll 1. \tag{39}$$

The rate γ_r is so much smaller than γ_2, and usually much smaller than γ_2^*, that it can be safely neglected, being absolutely unable to influence the motion of spins. Actually, Bloembergen[1] has already analysed this problem and come to the conclusion that the interaction of spins through the magnetodipole radiation field is completely negligible. However, one may put the following question. Suppose that the considered sample is ideally homogeneous, so that γ_2^* is very small, and let the initial spin polarization be very high, such

that $s_0^2 \approx 1$. Then the effective transverse rate (35) at the initial time $t=0$ can become rather small. Could then the radiation rate (37) play any noticeable role, at least at the very initial stage of spin motion? We study this problem below.

(9) *Thermal noise attenuation* γ_T emerges when the spin system is coupled to a resonant electric circuit. The resonator Nyquist noise, due to the thermal fluctuations of current in the circuit creates a fluctuational magnetic field, which has to be included in the effective force (12). The magnitude of the thermal field, produced by the Nyquist noise, is well known.[10] It was found[12–16] that the resulting thermal attenuation is

$$\gamma_T = \frac{\eta\rho\mu_0^2\omega}{4\hbar\gamma N}\coth\frac{\omega}{2\omega_T}, \tag{40}$$

where η is a filling factor, ω is the natural frequency of the electric circuit, γ is the resonator ringing width, and $\omega_T \equiv k_B T/\hbar$ is the thermal frequency. Bloembergen and Pound[8] first mentioned that, because of the macroscopic number of spins N entering the denominator of γ_T, the latter is unable to influence any spin motion in a macroscopic sample. This conclusion was confirmed by accurate calculations.[12–16]

(10) *Resonator relaxation rate* arises when the sample is coupled to a resonant electric circuit. Then in the effective force (12) the magnetic field H is the resonator feedback field. The role of this field will be thoroughly studied in what follows.

Summarizing all said above, for the spin averages (29) to (31), we obtain the evolution equations

$$\frac{du}{dt} = -i(\omega_s + \xi_0 - i\Gamma_2)u + fs, \tag{41}$$

$$\frac{dw}{dt} = -2\Gamma_2 w + (u^* f + f^* u)s, \tag{42}$$

$$\frac{ds}{dt} = -\frac{1}{2}(u^* f + f^* u) - \Gamma_1(s-\zeta), \tag{43}$$

supplemented by the initial conditions

$$u(0) = u_0, \quad w(0) = w_0, \quad s(0) = s_0.$$

In these equations, ζ is a stationary spin polarization, the characteristic spin frequency is

$$\omega_s = \omega_0 - \omega_D s, \tag{44}$$

with ω_0 given by Eq. (13) and ω_D by Eq. (19). The total longitudinal rate Γ_1 is defined in Eq. (32) and the total transverse rate Γ_2, in Eq. (35). The effective force is

$$f = -\frac{i}{\hbar}\mu_0(B_1 + H) + \xi + \gamma_r u, \tag{45}$$

where the last term is the friction force due to the interaction through magnetodipole radiation, and γ_r is the magnetodipole radiation rate (37). Equations (41) to (43) are stochastic differential equations, since they contain the random variables ξ_0 and ξ, whose stochastic averages are given in Eqs. (26). The external transverse field B_1 and the resonator feedback field H need yet to be specified.

V. RESONATOR FEEDBACK FIELD

The resonator feedback field H is created by the electric current of the coil surrounding the spin sample. We assume that the coil axis is along the axis x. The electric circuit is characterized by resistance R, inductance L, and capacity C. The spin sample is inserted into a coil of n turns, length l, cross-section area A_c, and volume $V_c = A_c l$. The electric current in the circuit is described by the Kirchhoff equation

$$L\frac{dj}{dt} + Rj + \frac{1}{C}\int_0^t j(t')dt' = E_f - \frac{d\Phi}{dt}, \tag{46}$$

in which E_f is an electromotive force, if any, and the magnetic flux

$$\Phi = \frac{4\pi}{c} nA_c \eta m_x, \tag{47}$$

where $\eta \approx V/V_c$ is a filling factor, is formed by the x component of the magnetization density

$$m_x \equiv \frac{\mu_0}{V}\sum_i \langle S_i^x \rangle. \tag{48}$$

The electric current, circulating over the coil, creates a magnetic field

$$H = \frac{4\pi n}{cl} j. \tag{49}$$

The circuit natural frequency is

$$\omega \equiv \frac{1}{\sqrt{LC}} \quad \left(L \equiv 4\pi \frac{n^2 A_c}{c^2 l} \right) \tag{50}$$

and the circuit damping is

$$\gamma \equiv \frac{1}{\tau} = \frac{R}{2L} = \frac{\omega}{2Q}, \tag{51}$$

where τ is called the circuit ringing time and $Q \equiv \omega L/R$ is the quality factor. Also, let us define the reduced electromotive force

$$e_f \equiv \frac{cE_f}{nA_c\gamma}. \tag{52}$$

Then the Kirchhoff equation (46) can be transformed to the equation

$$\frac{dH}{dt} + 2\gamma H + \omega^2 \int_0^t H(t')dt' = \gamma e_f - 4\pi\eta\frac{dm_x}{dt} \tag{53}$$

for the feedback magnetic field created by the coil.

The feedback equation (53) can be represented in another equivalent form that proved to be very convenient for defining the feedback field.[12–15] For this purpose, we involve the method of Laplace transforms and introduce the transfer function

$$G(t) = \left(\cos \omega' t - \frac{\gamma}{\omega'} \sin \omega' t \right) e^{-\gamma t}, \tag{54}$$

where

$$\omega' \equiv \sqrt{\omega^2 - \gamma^2}.$$

Thus we transform the feedback-field equation (53) to the integral representation

$$H = \int_0^t G(t-t') [\gamma e_f(t') - 4\pi\eta \dot{m}_x(t')] dt', \tag{55}$$

in which

$$\dot{m}_x(t) \equiv \frac{1}{2} \rho \mu_0 S \frac{d}{dt} (u^* + u). \tag{56}$$

Let the resonant part of the reduced electromotive force (52) be

$$e_f(t) = h_2 \cos \omega t. \tag{57}$$

And let us introduce the notation

$$\nu_2 \equiv \frac{\mu_0 h_2}{2\hbar}. \tag{58}$$

As usual, we assume that all attenuation parameters are much smaller than the characteristic spin frequency ω_s. Then Eq. (55) can be solved by an iteration procedure, which in first order gives

$$\frac{\mu_0 H}{\hbar} = i(\alpha u - \alpha^* u^*) + 2\beta \cos \omega t. \tag{59}$$

Here the coupling function

$$\alpha = \gamma_0 \omega_s \left[\frac{1 - \exp\{-i(\omega - \omega_s)t - \gamma t\}}{\gamma + i(\omega - \omega_s)} + \frac{1 - \exp\{-i(\omega + \omega_s)t - \gamma t\}}{\gamma - i(\omega + \omega_s)} \right] \tag{60}$$

describes the coupling of spins with the resonator and the function

$$\beta = \frac{\nu_2}{2} (1 - e^{-\gamma t}) \tag{61}$$

characterizes the action of the resonator electromotive force on spins. In Eq. (60) the notation for the natural spin width

$$\gamma_0 \equiv \frac{\pi}{\hbar} \eta \rho \mu_0^2 S \tag{62}$$

is employed.

The spin-resonator coupling can be characterized by the dimensionless coupling parameter

$$g \equiv \frac{\gamma \gamma_0 \omega_s}{\gamma_2 (\gamma^2 + \Delta^2)}, \tag{63}$$

in which $\Delta \equiv \omega - |\omega_s|$ is the detuning. As is evident from Eq. (60), an efficient spin-resonator coupling is possible only when the detuning from the resonance is small, such that

$$\frac{|\Delta|}{\omega} \ll 1 \quad (\Delta \equiv \omega - |\omega_s|). \tag{64}$$

When the resonance is sufficiently sharp, so that $|\Delta| < \gamma$, then the coupling function (60) reduces to

$$\alpha = g \gamma_2 (1 - e^{-\gamma t}). \tag{65}$$

Thus the resonator feedback field H is defined by Eq. (59), in which α is given by Eq. (65) and β, by Eq. (61).

VI. AVERAGED EVOLUTION EQUATIONS

The resonator field, defined in Eq. (59), has to be substituted in the effective force (45) entering the evolution equations (41) to (43). In Eq. (45), we also need to specify the external magnetic field B_1. In general, the latter may contain a constant part and an alternating term. So, let us take this transverse field in the form

$$B_1 = h_0 + h_1 \cos \omega t. \tag{66}$$

In what follows, we shall use the notation

$$\nu_0 \equiv \frac{\mu_0 h_0}{\hbar}, \quad \nu_1 \equiv \frac{\mu_0 h_1}{2\hbar}. \tag{67}$$

Equations (41) to (43) are stochastic differential equations, containing the random variables ξ_0 and ξ describing local spin fluctuations. In order to derive the evolution equations in terms of ordinary differential equations, we have to accomplish the averaging over random fluctuations. This can be done by following the scale separation approach,[11–16] the usage of the stochastic averages (26), and by invoking the known techniques of treating stochastic variables.[49]

Keeping in mind that the attenuation parameters are substantially smaller than the characteristic spin frequency ω_s, we notice from Eqs. (41) to (43) that the function u can be classified as fast, being compared with the temporal behavior of the functions w and s. The latter play the role of temporal quasi-invariants with respect to u.

First, we substitute into Eqs. (41) to (43) the effective force (45), the resonator field (59), and the transverse magnetic field (66). This results in the equations

$$\frac{du}{dt} = -i(\omega_s + \xi_0)u - (\Gamma_2 - \alpha s - \gamma_r s)u + f_1 s - \alpha s u^*, \tag{68}$$

$$\frac{dw}{dt} = -2(\Gamma_2 - \alpha s - \gamma_r s)w + (u^* f_1 + f_1^* u)s - \alpha s(u^2 + (u^*)^2), \tag{69}$$

$$\frac{ds}{dt} = -(\alpha + \gamma_r)w - \frac{1}{2}(u^* f_1 + f_1^* u) - \Gamma_1(s - \zeta) + \frac{1}{2}\alpha(u^2 + (u^*)^2), \tag{70}$$

in which

$$f_1 \equiv -i\nu_0 - 2i(\nu_1+\beta)\cos\omega t + \xi. \tag{71}$$

Then we solve Eq. (68) for the fast variable u, keeping the quasi-invariants fixed, which yields

$$u = u_0 \exp\left\{ -(i\omega_s + \Gamma_2 - \alpha s - \gamma_r s)t - i\int_0^t \xi_0(t')dt' \right\} + s\int_0^t f_1(t')\exp\left\{ -(i\omega_s + \Gamma_2 - \alpha s - \gamma_r s)(t-t') - i\int_{t'}^t \xi_0(t'')dt'' \right\} dt'. \tag{72}$$

Solution (72) must be substituted in Eqs. (69) and (70) for the slow functions w and s. After this, the latter equations have to be averaged over time and over the stochastic variables ξ_0 and ξ, again keeping the quasi-invariants fixed. To slightly simplify the resulting equations, one can take the initial condition for the transition function u in the real form, such that $u_0^* = u_0$, which is not principal but just makes the equations less cumbersome.

To present the resulting equations in a compact form, we introduce the *effective attenuation*

$$\Gamma_3 \equiv \gamma_3 + \frac{\nu_0^2\Gamma}{\omega_s^2+\Gamma^2} - \frac{\nu_0(\nu_1+\beta)\Gamma}{\omega_s^2+\Gamma^2}e^{-\Gamma t} + \frac{(\nu_1+\beta)^2\Gamma}{\Delta^2+\Gamma^2}(1-e^{-\Gamma t}), \tag{73}$$

in which

$$\Gamma \equiv \Gamma_2 + \gamma_3 - (\alpha+\gamma_r)s. \tag{74}$$

And finally, after the described averaging, we obtain the evolution equations

$$\frac{dw}{dt} = -2(\Gamma_2 - \alpha s - \gamma_r s)w + 2\Gamma_3 s^2, \tag{75}$$

$$\frac{ds}{dt} = -(\alpha+\gamma_r)w - \Gamma_3 s - \Gamma_1(s-\zeta). \tag{76}$$

These equations are very general. They include various attenuation processes, described in Sec. IV, and take into account transverse constant and alternating fields (66), as well as the resonator electromotive force (57) entering through function (61). The resonator feedback field is responsible for the appearance of the coupling function (65). Notice that the radiation relaxation rate γ_r, defined in Eq. (37), enters everywhere together with the spin-resonator coupling α. However their values are drastically different. Since

$$\frac{\gamma_r}{\alpha} \sim 0.1\frac{\gamma}{\omega_s}(kL_s)^3 \ll 1,$$

the value of γ_r is so incomparably smaller than $\alpha \sim g\gamma_2$, that it is evident, in the presence of a resonator, the rate γ_r must be forgotten.

Moreover, even when there is no resonator, so that $\alpha=\beta=0$, the radiation rate γ_r plays no role, since it is much smaller than γ_2, γ_2^*, and γ_3. One might think that γ_r could play a role in the following unrealistic case. Let us imagine an absolutely ideal lattice with no inhomogeneous broadening, that is, let us set $\gamma_2^*=0$, which is certainly a purely imaginary situation. Then, according to Eq. (35), one has $\Gamma_2=\gamma_2(1-s^2)$. Assume that the spin system is completely polarized, with $s_0=1$. Hence, at the initial time, $\Gamma_2=0$. Could then the spin motion be started by the term with γ_r? The answer is evident: As far as the largest terms in both Eqs. (75) and (76) are those containing Γ_3, the terms with γ_r are always negligible, even if $\Gamma_2=0$. Even more, functions (30) and (31), by their definition, satisfy the inequality

$$w + s^2 \leqslant 1. \tag{77}$$

Therefore, if one sets $s_0=1$, then $w_0=0$, and the term $\gamma_r w$ simply disappears from the equations. Vice versa, if one sets a noticeable $w_0 \sim 1$, then $s^2 \ll 1$, and $\Gamma_2 \approx \gamma_2 \gg \gamma_r$. In this way, the radiation rate γ_r never plays any role in the spin motion, which is in agreement with the estimates by Bloembergen.[1]

Note that the situation in spin systems is principally different from that happening in atomic systems. In the latter, both the linewidth $\gamma_2 = 2|\mathbf{d}|^2k^3/3$ as well as the collective radiation rate $\gamma_r = (2/3)|\mathbf{d}|^2k^3N_c$, where N_c is the number of correlated atoms, forming a wave packet, are caused by the same physical process, by the interaction of atoms with their radiation field. Hence $\gamma_r/\gamma_2 = N_c \gg 1$, which results in the coherentization of the dipole transitions. This is possible even if $kL \gg 1$, but the number of atoms in a partial wave packet is $N_c \gg 1$, since $\gamma_r/\gamma_2 = N_c \gg 1$. Contrary to this, in spin systems the linewidth γ_2, given in Eq. (33), is due to direct dipole-dipole interactions, while the radiation rate (37) is a result of the spin interactions with their radiation field. This is why in the latter case, one always has $\gamma_r \ll \gamma_2$, and the radiation rate γ_r plays no part in the motion of spins.

We may also notice that in the effective attenuation (73) the terms due to the presence of a constant transverse field are less important than the terms caused by the local spin fluctuations and by the alternating transverse fields. Therefore, omitting the terms corresponding to a permanent transverse magnetic field, we have

$$\Gamma_3 = \gamma_3 + \frac{(\nu_1+\beta)^2\Gamma}{\Gamma^2+\Delta^2}(1-e^{-\Gamma t}). \tag{78}$$

Finally, we obtain the evolution equations

$$\frac{dw}{dt} = -2(\Gamma_2 - \alpha s)w + 2\Gamma_3 s^2, \tag{79}$$

$$\frac{ds}{dt} = -\alpha w - \Gamma_3 s - \Gamma_1(s-\zeta), \tag{80}$$

describing the averaged motion of spins.

VII. COHERENCE EMERGING FROM CHAOS

One of the most intriguing questions is how the spin motion could become coherent if initially it was not. This is a particular case of the general physical problem of how coherence emerges from chaos.

Being interested in a self-organized process of arising coherence, let us consider the case, when there are no external transverse fields pushing spins, that is $\nu_1=\beta=0$. Then Eq. (78) yields $\Gamma_3=\gamma_3$. Assume also that there is no pumping, so that $\gamma_1^*=0$, hence $\Gamma_1=\gamma_1$. Under these conditions, the initial spin motion, for the time t such that

$$\gamma_1 t \ll 1, \quad \gamma_2 t \ll 1, \quad \gamma_3 t \ll 1, \tag{81}$$

follows from Eqs. (79) and (80) in the form

$$w \simeq w_0 + 2[\gamma_3 s_0^2 - \gamma_2(1 - s_0^2 + \kappa)w_0]t,$$

$$s \simeq s_0 - [(\gamma_1 + \gamma_3)s_0 - \gamma_1\zeta]t, \tag{82}$$

where the inhomogeneity coefficient is introduced,

$$\kappa \equiv \gamma_2^*/\gamma_2. \tag{83}$$

If at the initial time no transverse polarization is imposed on the system, and the initial coherence function is zero, $w_0=0$, nevertheless the coherent spin motion starts developing according to the law

$$w \simeq 2\gamma_3 s_0^2 t \quad (w_0 = 0), \tag{84}$$

provided there is an initial longitudinal polarization $s_0 \neq 0$. The initiation of the emerging coherent motion is caused by local spin fluctuations creating the effective rate γ_3. Recall that in the Bloch equations coherent motion never appears if it is not imposed by the initial conditions. Contrary to this, Eqs. (79) and (80) take into account the local spin fluctuations triggering the motion of spins. The second of Eqs. (82), keeping in mind that usually $\gamma_1 \ll \gamma_3 \sim \gamma_2$, can be simplified to

$$s \simeq s_0(1 - \gamma_3 t). \tag{85}$$

At the initial stage of spin motion, their coherence is yet incipient, and the motion is mainly governed by quantum chaotic spin fluctuations. The coherentization of the transverse motion goes through the resonator feedback field and the growing coupling function (65). The quantitative change in the spin motion happens when the coupling function (65) becomes so large that the term $(\Gamma_2 - \alpha s)$ in Eq. (79) goes negative, which means that an efficient generation of coherence has started in the system. This is analogous to the beginning of maser generation.[15–19] The moment of time, when the regime of mainly chaotic quantum fluctuations transforms into the regime of predominantly coherent spin motion, can be called the *chaos time*. This time t_c is defined by the equality $\alpha s=\Gamma_2$, that is by the equation

$$\alpha s = \gamma_2(1 - s^2) + \gamma_2^* \quad (t = t_c). \tag{86}$$

From here, the estimate for the chaos time is

$$t_c = \tau \ln \frac{g s_0}{g s_0 - 1 + s_0^2 - \kappa}, \tag{87}$$

where τ is the resonator ringing time defined in Eq. (51). The regime of chaotic spin fluctuations lasts till the chaos time (87), after which the coherent stage of spin motion comes into play. As is clear from the above equations, the transformation from the chaotic to coherent regime goes as a gradual crossover. Notice that the quantity $1-s_0^2+\kappa$ is positive since $s_0^2 \leqslant 1$. Then, in order that the chaos time (87) be positive and finite, the inequality

$$g s_0 > 1 - s_0^2 + \kappa > 0 \tag{88}$$

must hold. For a strong spin-resonator coupling, when $g s_0 \gg 1$, the chaos time (87) reduces to

$$t_c \simeq \frac{\tau}{g s_0}(1 - s_0^2 + \kappa). \tag{89}$$

As is seen, there exists a well-defined stage of chaotic spin fluctuations, with a finite chaos time $t_c>0$, after which the coherent regime develops, if $g s_0>0$. The coupling parameter g is defined in Eq. (63), from which it follows that one should have $\omega_s s_0>0$. Assuming that the initial spin polarization is positive, $s_0>0$, one gets the requirement that $\omega_s>0$. The latter, by definition (44), is equivalent to the condition $\omega_0>\omega_D s$. Moreover, the coupling function (65) is obtained under the resonance condition (64), which implies that ω_s has to be close to the resonator natural frequency ω. There are two ways of preserving the resonance condition (64). First, one can impose a sufficiently strong external magnetic field B_0, such that the frequency ω_0, given by Eq. (13), would be much larger than ω_D, defined in Eq. (19). This becomes trivial for $S=1/2$, when $\omega_D=0$. If $\omega_0 \gg \omega_D$, then it is easy to realize the resonance condition (64), with $\omega_s \approx \omega$ and slightly varying in time detuning $\Delta=\omega-\omega_s$.

The second way of keeping the resonance condition (64) is by means of the chirping effect.[16,19] This requires to vary in time the external magnetic field B_0 so that to maintain the equality

$$\frac{\mu_0 B_0}{\hbar} + (\omega + \omega_D s) = \Delta, \tag{90}$$

with a fixed detuning.

VIII. COHERENT SPIN RELAXATION

After the chaos time (87), the motion of spins becomes more and more coherent, being collectivized by the resonator feedback field, with the coupling function α reaching the value $g\gamma_2$. At the transient stage, when $t>t_c$ but $t \ll T_1$, we may neglect the term with γ_1 in Eq. (80). Assuming that there is no pumping, that is $\gamma_1^*=0$, one has $\Gamma_1=\gamma_1$. Let us continue studying the case of the self-organized coherent spin motion, when there are no transverse external fields, so that $\nu_1=\beta=0$, hence $\Gamma_3=\gamma_3$. When the coherence is well developed, then the main term in Eq. (79) is the first one, while the term with γ_3 can be neglected. Under these conditions, and using expression (35) for the rate Γ_2, Eqs. (79) and (80) reduce to the form

$$\frac{dw}{dt} = -2\gamma_2(1 - s^2 + \kappa - g s)w, \tag{91}$$

$$\frac{ds}{dt} = -g\gamma_2 w. \tag{92}$$

The solution of these equations is explained in Appendix C and it yields

$$w = \left(\frac{\gamma_p}{g\gamma_2}\right)^2 \mathrm{sech}^2\left(\frac{t-t_0}{\tau_p}\right),$$

$$s = -\frac{\gamma_p}{g\gamma_2}\tanh\left(\frac{t-t_0}{\tau_p}\right) + \frac{1+\kappa}{g}. \tag{93}$$

Here

$$\tau_p \equiv 1/\gamma_p \tag{94}$$

is the pulse time showing the duration of the coherent relaxation occurring as a fast pulse. The delay time

$$t_0 = t_c + \frac{\tau_p}{2}\ln\left|\frac{\gamma_p+\gamma_g}{\gamma_p-\gamma_g}\right| \tag{95}$$

defines the time of the maximal coherence. The pulse width is given by the relation

$$\gamma_p^2 = \frac{1}{2}\gamma_g^2\left[1+\sqrt{1+4\left(\frac{g\gamma_2}{\gamma_g}\right)^2 w_c}\right], \tag{96}$$

in which

$$\gamma_g \equiv \gamma_2(gs_c - 1 - \kappa). \tag{97}$$

The boundary values w_c and s_c are

$$w_c = w_0 + 2[\gamma_3 s_0^2 - \gamma_2(1 - s_0^2 + \kappa)w_0]t_c, \quad s_c = s_0(1-\gamma_3 t_c), \tag{98}$$

with the chaos time t_c given in Eq. (87). Since we are interested in the self-organized collective process, when there is no large transverse polarization imposed on the system at the initial time, we may set $w_0 \ll s_0^2$. Then Eq. (96) simplifies to

$$\gamma_p^2 = \gamma_g^2 + (g\gamma_2)^2 w_c. \tag{99}$$

The pulse time (94) reads as

$$\tau_p = \frac{T_2}{\sqrt{(gs_c - 1 - \kappa)^2 + g^2 w_c}}. \tag{100}$$

It is easy to notice that if the spin-resonator coupling is weak, $g \ll 1$, then $\gamma_p \sim \gamma_g \sim \gamma_2$ and $\tau_p \sim T_2$. In that case, no self-organized coherence can arise in the system.

Collective coherent effects appear in the spin motion only if the pulse time τ_p is smaller than the dephasing time T_2. The inequality $\tau_p < T_2$, according to Eq. (100), requires that

$$(gs_c - 1 - \kappa)^2 + g^2 w_c > 1. \tag{101}$$

Three different regimes can satisfy Eq. (101).

The regime of *collective induction* happens when

$$gs_0 < 1 + \kappa, \quad g^2 w_0 > 1. \tag{102}$$

Then, as is clear from Eq. (97), one has $\gamma_g < 0$, because of which $t_0 < t_c$. This means that there is no noticeable maximum in the coherence function w, since, by definition, the delay time (95) should occur after the chaotic stage, so that $t_0 > t_c$. But the latter implies that $\gamma_g > 0$.

The *triggered coherent relaxation* corresponds to

$$gs_0 > 1 + \kappa, \quad 0 < g^2 w_0 < 1. \tag{103}$$

And the purely *self-organized coherent relaxation* takes place when

$$gs_0 > 2 + \kappa, \quad w_0 = 0. \tag{104}$$

In this classification, we keep in mind the inequality $\gamma_3 t_c \ll 1$, owing to which $w_c \approx w_0$ and $s_c \approx s_0$. The initial coherence is assumed to be weak, so that $w_0 \ll 1$.

For $w_0 \ll s_0^2$, the delay time (95) can be represented as

$$t_0 = t_c + \frac{\tau_p}{2}\ln\frac{4(gs_c - 1 - \kappa)^2}{g^2 w_c}. \tag{105}$$

In the case of the purely self-organized coherent relaxation, for sufficiently large coupling and initial polarization, such that $gs_0 \gg 1$, the delay time (105) reduces to

$$t_0 = t_c + \frac{\tau_p}{2}\ln\left|\frac{2}{\gamma_3 t_c}\right|, \tag{106}$$

where $\tau_p = T_2/gs_0$. From these formulas, one sees that if $\gamma_3 \to 0$, then $t_0 \to \infty$, and no coherent relaxation is possible. This emphasizes the crucial role of the local spin fluctuations, whose existence results in the relaxation rate γ_3.

At the delay time (95), solutions (93) are given by the expressions

$$w(t_0) = w_c + \left(s_c - \frac{1+\kappa}{g}\right)^2, \quad s(t_0) = \frac{1+\kappa}{g}. \tag{107}$$

And for $t \gg t_0$, they exponentially decay to the values

$$w \simeq 4w(t_0)\exp(-2\gamma_p t),$$

$$s \simeq -s_c + \frac{2}{g}(1+\kappa) + 2\left(s_c - \frac{1+\kappa}{g}\right)\exp(-2\gamma_p t). \tag{108}$$

At very large times $t \sim T_1$, the transient equations (91) and (92) are no longer valid. Then one has to return to the full equations (79) and (80). With increasing time, the solutions tend to the stationary points defined by the zeros of the right-hand sides of these equations. Among the relaxation regimes to the stationary solutions, one is especially interesting, going through a long series of coherent pulses. This pulsing coherent relaxation takes place under a permanent external pumping described by a large pumping rate $\gamma_1^* \gg \gamma_1$. Then $\Gamma_1 = \gamma_1^*$. If also the coupling parameter is sufficiently large, such that $g\zeta \gg 1$ and

$$\frac{\gamma_3}{g\zeta\gamma_1^*} \ll 1,$$

then the fixed point of Eqs. (79) and (80) is given by the expressions

$$w^* = \frac{\gamma_1^*}{g(\gamma_2+\gamma_2^*)}\left(1-\frac{\gamma_3}{g\zeta\gamma_1^*}\right), \quad s^* = \frac{1}{g}\left(1-\frac{\gamma_3}{g\zeta\gamma_1^*}\right), \tag{109}$$

corresponding to a stable focus. The relaxation to the stationary solutions (109) realizes through a series of sharp coherent pulses, similar to the form of Eqs. (93), with the temporal interval between the pulses asymptotically defined by the separation time

$$T_{sep} = \frac{2\pi}{\sqrt{2g\zeta\gamma_1^*(\gamma_2+\gamma_2^*)}}. \tag{110}$$

The number of the separate coherent pulses can be estimated as $N_{sep}=1/\gamma_1^* T_{sep}$, which gives

$$N_{sep} = \sqrt{\frac{g\zeta(\gamma_2+\gamma_2^*)}{2\pi^2\gamma_1^*}}.$$

Such a highly nontrivial relaxation regime occurs only under a strong pumping and a sufficiently strong coupling with a resonator.

IX. INFLUENCE OF CROSS CORRELATIONS

When in the sample, in addition to the studied spins, there are spins of other nature, the presence of the latter can certainly influence the dynamics of the former. Let us consider the case of two types of coexisting spins, S and F. The total Hamiltonian is the sum

$$\hat{H} = \hat{H}_S + \hat{H}_F + \hat{H}_{SF} \tag{111}$$

of the Hamiltonians for S spins, F spins, and their interactions. The Hamiltonian $\hat{H}_S$ of S spins is the same as in Eqs. (1) to (4). Let us accept for the Hamiltonian $\hat{H}_F$ of F spins a similar general form

$$\hat{H}_F = \sum_i \hat{H}_{iF} + \frac{1}{2}\sum_{i\neq j}\hat{H}_{ijF}. \tag{112}$$

The single-spin terms are

$$\hat{H}_{iF} = -\mu_{0F}\mathbf{B}\cdot\mathbf{F}_i - D_F(F_i^z)^2, \tag{113}$$

with the total magnetic field (3). And the interaction terms are given by

$$\hat{H}_{ijF} = \sum_{\alpha\beta} D_{ijF}^{\alpha\beta} F_i^\alpha F_j^\beta - J_{ijF}\mathbf{F}_i\cdot\mathbf{F}_j, \tag{114}$$

with the dipolar tensor

$$D_{ijF}^{\alpha\beta} = \frac{\mu_{0F}^2}{r_{ij}^3}(\delta_{\alpha\beta} - 3n_{ij}^\alpha n_{ij}^\beta).$$

Assume that the interactions between the S and F spins are represented by the Hamiltonian

$$\hat{H}_{SF} = \sum_i A\mathbf{S}_i\cdot\mathbf{F}_i + \sum_{i\neq j}\sum_{\alpha\beta} A_{ij}^{\alpha\beta} S_i^\alpha F_j^\beta, \tag{115}$$

containing the part of the single-site interactions of intensity A and the part of the dipole interactions, with the dipolar tensor

$$A_{ij}^{\alpha\beta} = \frac{\mu_0\mu_{0F}}{r_{ij}^3}(\delta_{\alpha\beta} - 3n_{ij}^\alpha n_{ij}^\beta).$$

In particular, these could be hyperfine interactions between nuclear and electron spins.[15,50]

We employ notation (8) for the interaction parameters of S spins and an equivalent notation for the interaction parameters a_{ijF}, b_{ijF}, and c_{ijF} of F spins. Similarly, we define the interaction parameters

$$\overline{a_{ij}} \equiv A_{ij}^{zz}, \quad \overline{b_{ij}} \equiv \frac{1}{4}(A_{ij}^{xx} - A_{ij}^{yy} - 2iA_{ij}^{xy}), \quad \overline{c_{ij}} \equiv \frac{1}{2}(A_{ij}^{xz} - iA_{ij}^{yz}) \tag{116}$$

for the spin cross interactions.

The local fields (11), acting on S spins, are generalized to the form

$$\xi_0 \equiv \frac{1}{\hbar}\sum_{j(\neq i)}[a_{ij}S_j^z + c_{ij}^* S_j^- + c_{ij}S_j^+ + J_{ij}(S_i^z - S_j^z) + \overline{a}_{ij}F_j^z + \overline{c}_{ij}^* F_j^- + \overline{c}_{ij}F_j^+], \tag{117}$$

$$\xi \equiv \frac{i}{\hbar}\sum_{j(\neq i)}\left[2c_{ij}S_j^z - \frac{1}{2}a_{ij}S_j^- + 2b_{ij}S_j^+ + J_{ij}(S_i^- - S_j^-) + 2\overline{c}_{ij}F_j^z - \frac{1}{2}\overline{a}_{ij}F_j^- + 2\overline{b}_{ij}F_j^+\right].$$

Analogous local fields act on F spins,

$$\xi_{0F} \equiv \frac{1}{\hbar}\sum_{j(\neq i)}[a_{ijF}F_j^z + c_{ijF}^* F_j^- + c_{ijF}F_j^+ + J_{ijF}(F_i^z - F_j^z) + \overline{a}_{ij}S_j^z + \overline{c}_{ij}^* S_j^- + \overline{c}_{ij}S_j^+],$$

$$\xi_F \equiv \frac{i}{\hbar}\sum_{j(\neq i)}\left[2c_{ijF}F_j^z - \frac{1}{2}a_{ijF}F_j^- + 2b_{ijF}F_j^+ + J_{ijF}(F_i^- - F_j^-) + 2\overline{c}_{ijF}S_j^z - \frac{1}{2}\overline{a}_{ij}S_j^- + 2\overline{b}_{ij}S_j^+\right]. \tag{118}$$

Instead of one effective force (12), we have now two forces

$$f \equiv -\frac{i}{\hbar}\mu_0(B_1+H) + \frac{i}{\hbar}A_i F_i^- + \xi,$$

$$f_F \equiv -\frac{i}{\hbar}\mu_{0F}(B_1+H) + \frac{i}{\hbar}A_i S_i^- + \xi_F. \tag{119}$$

In addition to frequency (13), let us introduce the effective frequencies

$$\omega_{0F} \equiv -\frac{\mu_{0F}}{\hbar}B_0, \quad \varepsilon \equiv \frac{A}{\hbar}. \tag{120}$$

The Heisenberg equations of motion for the system with Hamiltonian (111) yield the equations for S spins

$$\frac{dS_i^-}{dt} = -i(\omega_0 + \varepsilon F_i^z + \xi_0)S_i^- + S_I^z f + \frac{i}{\hbar}D(S_i^- S_i^z + S_i^z S_i^-),$$

$$\frac{dS_i^z}{dt} = -\frac{1}{2}(f^+ S_i^- + S_i^+ f), \tag{121}$$

and the equations for F spins

$$\frac{dF_i^-}{dt} = -i(\omega_{0F} + \varepsilon S_i^z + \xi_{0F})F_i^- + F_I^z f_F + \frac{i}{\hbar}D_F(F_i^- F_i^z + F_i^z F_i^-),$$

$$\frac{dF_i^z}{dt} = -\frac{1}{2}(f_F^+ F_i^- + F_i^+ f_F). \tag{122}$$

Again we assume that the sample is inserted into the coil of a resonant electric circuit. The feedback field acting on the sample is given by Eq. (53) or (55), where now the magnetic-moment density is

$$m_x = \frac{\mu_0}{V}\sum_{i=1}^{N}\langle S_i^x\rangle + \frac{\mu_{0F}}{V}\sum_{j=1}^{N_F}\langle F_j^x\rangle, \tag{123}$$

with N_F being the number of F spins.

Averaging Eqs. (121) and (122), we derive the evolution equations for functions (29), (30), and (31), corresponding to S spins, as well as the equations for the functions

$$u_F \equiv \frac{1}{FN_F}\sum_{i=1}^{N_F}\langle F_i^-\rangle, \tag{124}$$

$$w_F \equiv \frac{1}{F^2 N_F(N_F-1)}\sum_{i\neq j}^{N_F}\langle F_i^+ F_j^-\rangle, \tag{125}$$

$$s_F \equiv \frac{1}{FN_F}\sum_{i=1}^{N_F}\langle F_i^z\rangle, \tag{126}$$

describing F spins. In this notation, the transverse magnetic-moment density (123) is

$$m_x = \frac{1}{2}\rho\mu_0 S(u^* + u) + \frac{1}{2}\rho_F\mu_{0F}F(u_F^* + u_F),$$

where ρ_F is the density of F spins.

The analysis of the evolution equations for the combined system of S and F spins is the same as has been given above for one type of spins S, with the difference that all expressions become much more cumbersome. Again it is possible to show that in the triggering of spin motion an important role is played by the coupled S-F spin fluctuations, which yield the dynamic relaxation rates γ_3 and γ_{3F} defined by the relations

$$\gamma_3^2 = \gamma_{SS}^2 + \gamma_{SF}^2, \quad \gamma_{3F}^2 = \gamma_{FF}^2 + \gamma_{FS}^2, \tag{127}$$

where

$$\gamma_{SS} \approx \rho\frac{\mu_0^2}{\hbar}\sqrt{S(S+1)}, \quad \gamma_{SF} \approx \sqrt{\rho\rho_F}\frac{\mu_0\mu_{0F}}{\hbar}F,$$

$$\gamma_{FF} \approx \rho_F\frac{\mu_{0F}^2}{\hbar}\sqrt{F(F+1)}, \quad \gamma_{FS} \approx \sqrt{\rho\rho_F}\frac{\mu_{0F}\mu_0}{\hbar}S.$$

The effective frequencies of S and F spins, respectively, are

$$\omega_S = \omega_0 - \omega_D s + \varepsilon s_F S, \quad \omega_F = \omega_{0F} - \omega_{DF}s_F + \varepsilon s F, \tag{128}$$

where ω_D is given by Eq. (19) and

$$\omega_{DF} \equiv (2F-1)\frac{D_F}{\hbar}. \tag{129}$$

We shall not overload this paper by a detailed exposition of various cross correlations resulting from the complicated system of the coupled evolution equations for S and F spins. Let us only emphasize the existence of a rather nontrivial nonlinear effect of mutual spin interactions through the resonator feedback field. Calculating the latter from the integral representation (55), with the transverse magnetic density (123), and substituting this into the evolution equations results in an effective mutual influence of spins through the feedback field. If the resonator is tuned to the characteristic frequency ω_S of S spins, then for the latter, we derive the evolution equations similar to Eqs. (79) and (80), but with the effective spin-resonator coupling

$$g = \frac{\gamma\gamma_0\omega_S}{\gamma_2(\gamma^2+\Delta^2)}\left(1 + \frac{\rho_F\mu_{0F}\varepsilon s_F F}{\rho\mu_0\omega_F}\right), \tag{130}$$

instead of Eq. (63), and with γ_3 given by Eq. (127). Depending on the spin characteristics, coupling (130) can substantially surpass the value of Eq. (63). This is because the subsystem of F spins, coupled to a resonator, becomes itself a kind of an additional resonator for S spins.

X. CONCLUSION

A general theory is developed for describing nonlinear spin relaxation, which occurs when the spin system is prepared in a strongly nonequilibrium state and when the sample is coupled to a resonator electric circuit. A strongly nonequilibrium initial state can be realized by placing a polarized sample into an external magnetic field, whose direction is opposite to the sample magnetization. Nonlinearity in spin relaxation comes from direct spin-spin interactions and from their effective interactions through the resonator feedback field. Direct spin interactions are responsible for the appearance of local spin fluctuations, playing a crucial role at the starting stage of relaxation. The resonator feedback field collectivizes the spin motion, leading to coherent collective relaxation. The developed theory is based on a realistic Hamiltonian containing the main spin interactions. The role of various relaxation rates is thoroughly analyzed.

The aim of the present paper has been to develop a general theory providing an accurate and realistic description of nonlinear spin relaxation. This theory can be employed for a large class of polarized spin materials. Applications to particular substances require a special consideration and separate publications. There exists a large variety of materials that can be treated by the developed theory. Just to give an example, we may mention the class of molecular magnets.[16,19,24–26] For instance, the molecular crystal V_{15} is made of molecules of spin $1/2$, so has no magnetic anisotropy. Its nonlinear spin relaxation can be realized in a rather weak external field $B_0 \geqslant 1$ G. The molecules Mn_{12} and Fe_8 possess the spin $S=10$. They form crystals with density $\rho \sim 10^{21}$ cm^{-3}. The anisotropy frequency is $\omega_D \sim 10^{12}$ s^{-1}. At low temperatures below about 1 K, the molecules can be well polarized, with the spin-lattice relaxation parameters $\gamma_1 \sim 10^{-5}-10^{-7}$ s^{-1}. The line width is caused by rather strong dipole interactions, with $\gamma_2 \sim 10^{10}$ s^{-1}. The condition $\omega_0 > \omega_D$ can be reached for $B_0 > 10^5$ G. In the molecular magnet, formed by the molecules Mn_6, whose spin is $S=12$, the magnetic anisotropy is much weaker, with $\omega_D \sim 10^{10}$ s^{-1}, being of the same order as $\gamma_2 \sim 10^{10}$ s^{-1}. Therefore the required magnetic field is not high, $B_0 > 10^3$ G. Coupling a molecular crystal to a resonant circuit with the natural width $\gamma \equiv \omega/2Q$, where Q is the resonator quality factor, one can attain the values of the coupling parameter as large as $g \sim Q \sim 10^4$. With such a strong coupling, the influence of the resonator feedback field outperforms other relaxation mechanisms, producing fast coherent relaxation, with relaxation times $\tau_p \sim 10^{-13}$ s. Such a fast reorientation of the magnetic moment can result in the emission of radiation pulses of high intensity.

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APPENDIX A: EFFECTIVE HOMOGENEOUS BROADENING

The homogeneous broadening, existing in spin systems, arises from spin-spin interactions and is usually expressed through the moments M_n, which may depend on the level of the longitudinal polarization s, provided the latter is sufficiently large. The moments have been calculated in a number of works 1–7,21. The most general and exact formula, relating the effective broadening with the moments, can be found in Abragam and Goldman,[6] which for the Gaussian line shape is

$$\gamma_2(s) = \sqrt{\frac{\pi M_2^3(s)}{2[M_4(s) - M_2^2(s)]}}.$$

The Lorentzian line shape yields to practically the same expression, with a slightly different coefficient. The broadening $\gamma_2(s)$ for the Lorentzian line is $\sqrt{\pi}$ of the Gaussian broadening. The dependence of the moments on the polarization has been accurately calculated,[6] yielding

$$M_2(s) = M_2(0)(1 - s^2),$$

$$M_4(s) = 2.18 M_2^2(0)(1 - s^2)(1 - 0.42 s^2).$$

Substituting this into $\gamma_2(s)$, and taking into account that $s^2 \leqslant 1$, we obtain Eq. (34).

APPENDIX B: SPIN RADIATION RATE

To get a fully quantum-mechanical microscopic picture of spin interactions with electromagnetic field they radiate, one has to add to the spin Hamiltonian (1) the field Hamiltonian

$$\hat{H}_f = \frac{1}{8\pi} \int (\mathbf{E}^2 + \mathbf{H}^2) d\mathbf{r},$$

where $\mathbf{E}=\mathbf{E}(\mathbf{r},t)$ is electric field and $\mathbf{H}=\mathbf{H}(\mathbf{r},t)$ is magnetic field, and the operator energy of spin-field interactions

$$\hat{H}_{sf} = -\mu_0 \sum_{i=1}^{N} \mathbf{S}_i \cdot \mathbf{H}_i,$$

where $\mathbf{H}_i=\mathbf{H}(\mathbf{r}_i,t)$. From the Heisenberg equations of motion for the field variables, one finds the vector potential

$$\mathbf{A}(\mathbf{r},t) = \frac{1}{c} \int \mathbf{j}\left(\mathbf{r}', t - \frac{|\mathbf{r}-\mathbf{r}'|}{c}\right) \frac{d\mathbf{r}}{|\mathbf{r}-\mathbf{r}'|},$$

in which the current density is

$$\mathbf{j} = -c\mu_0 \sum_{i=1}^{N} \mathbf{S}_i \times \vec{\nabla}\, \delta(\mathbf{r} - \mathbf{r}_i).$$

The vector potential $\mathbf{A}_i \equiv \mathbf{A}(\mathbf{r}_i,t)$ can be represented as

$$\mathbf{A}_i = \mathbf{A}_i^- + \mathbf{A}_i^+ + \mathbf{A}_i',$$

where

$$\mathbf{A}_i^- = -\sum_j \left(1 + \frac{1}{c}\frac{\partial}{\partial t}\right) \frac{\mathbf{r}_{ij}}{r_{ij}^3} \times \vec{\mu}^* S_j^-\left(t - \frac{r_{ij}}{c}\right),$$

$$\mathbf{A}_i' = -\sum_j \frac{\mathbf{r}_{ij}}{r_{ij}} \times \vec{\mu}_0 S_j^z\left(t - \frac{r_{ij}}{c}\right),$$

with the notation

$$\vec{\mu} \equiv \frac{\mu_0}{2}(\mathbf{e}_x - i\mathbf{e}_y), \quad \vec{\mu}_0 \equiv \mu_0 \mathbf{e}_z.$$

From here, we get the magnetic field $\mathbf{H}_i \equiv \mathbf{H}(\mathbf{r}_i,t)$ acting on an ith spin as $\mathbf{H}_i = \vec{\nabla}_i \times \mathbf{A}_i$, which gives the field

$$\mathbf{H}_i = \mathbf{H}_i^- + \mathbf{H}_i^+ + \mathbf{H}_i',$$

in which

$$\mathbf{H}_i^- = -\sum_j \left[\frac{\vec{\mu}^* - (\vec{\mu}^* \cdot \mathbf{n}_{ij})\mathbf{n}_{ij}}{c^2 r_{ij}} \frac{\partial^2}{\partial t^2} + \frac{\vec{\mu}^* - 3(\vec{\mu}^* \cdot \mathbf{n}_{ij})\mathbf{n}_{ij}}{r_{ij}^3} \times \left(1 + \frac{r_{ij}}{c}\frac{\partial}{\partial t}\right)\right] S_j^-\left(t - \frac{r_{ij}}{c}\right),$$

$$\mathbf{H}_i' = -\sum_j \frac{\vec{\mu}_0 - 3(\vec{\mu}_0\cdot\mathbf{n}_{ij})\mathbf{n}_{ij}}{r_{ij}^3} S_j^z\left(t-\frac{r_{ij}}{c}\right).$$

If the spins on different sites move independently of each other, so that the single-spin terms in the above sums chaotically oscillate, then the average magnetic field acting on each spin from the radiation of other spins is zero. Noticeable action of other spins can arise only if there exist the groups of spins, the so-called spin packets, which are strongly correlated, moving together. A substantial mutual interaction between spins, caused by their electromagnetic radiation, can appear only when this radiation is monochromatic, with a well-defined spin frequency ω_s, the related wavelength $\lambda = 2\pi c/\omega_s$, and wave vector $k=\omega_s/c$. This radiation can collectivize spins in a spin packet of size L_s, provided that

$$kL_s \ll 1.$$

When the radiation wavelength λ is much larger than the system length L, then $L_s=L$. This, however, is not compulsory, and the size of a spin packet can be much shorter than L, but it should be much larger than the mean interspin distance. Thus inequality (38) is a necessary condition for the appearance of collective effects.

Under condition (38), the above magnetic fields can be simplified, averaging them over spherical angles. The resulting expressions have to be added to the magnetic field in the effective force (12), which acquires one more term, being the friction force

$$f' = (\gamma_r - i\delta\omega)u,$$

in which the collective radiation rate and frequency shift are

$$\gamma_r \equiv \gamma_0 \sum_j^{N_s} \frac{\sin(kr_{ij})}{kr_{ij}}\Theta(ct-r_{ij}),$$

$$\delta\omega \equiv \gamma_0 \sum_j^{N_s} \frac{\cos(kr_{ij})}{kr_{ij}}\Theta(ct-r_{ij}),$$

where

$$\gamma_0 \equiv \frac{2}{3\hbar}\mu_0^2 S k^3$$

is the single-spin natural width, $\Theta(\cdot)$ is a unit-step function, and $N_s=\rho L_s^3$ is the number of spins in a spin packet. These formulas can be further simplified to

$$\gamma_r = \gamma_0 N_s = \frac{2}{3\hbar}\mu_0^2 S k^3 N_s$$

and

$$\delta\omega = \frac{3\gamma_r}{2kL_s} = \frac{1}{\hbar}\rho\mu_0^2 S (kL_s)^2.$$

The frequency shift is very small, even as compared to γ_2, since

$$\frac{\delta\omega}{\gamma_2} \cong 0.1(kL_s)^2 \ll 1.$$

Of course, such a small shift can be omitted, being negligible as compared to γ_2 and the more so as compared to ω_s. And for the radiation rate γ_r, substituting there $N_s=\rho L_s^3$, we obtain Eq. (37).

APPENDIX C: TRANSIENT STAGE OF RELAXATION

After the chaotic stage of spin fluctuations, the transient stage comes into play, characterized by Eqs. (91) and (92). The latter, by introducing the function

$$y \equiv \gamma_2(1 - s^2 + \kappa - gs)$$

and keeping in mind a sufficiently large coupling parameter $g \gg s$, rearrange to

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

Differentiating the second of these equations, we have

$$\frac{d^2y}{dt^2} + 2y\frac{dy}{dt} = 0,$$

which yields

$$\frac{dy}{dt} + y^2 = \gamma_p^2,$$

with γ_p being an integration parameter. This Riccati equation possesses the solution

$$y = \gamma_p \tanh\left(\frac{t-t_0}{\tau_p}\right),$$

in which $\gamma_p\tau_p \equiv 1$ and t_0 is another integration constant. Inverting the dependence of y on s for $s^2 \leqslant 1$, we get

$$s = -\frac{y}{g\gamma_2} + \frac{1+\kappa}{g}.$$

This gives the second of Eqs. (93), while the first of solutions (93) follows from Eq. (92). The integration constants γ_p and t_0 are defined by the initial conditions, which for the transient stage are $w_c=w(t_c)$ and $s_c=s(t_c)$.

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