Nonlinear spin dynamics in ferromagnets with electron-nuclear coupling

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Nonlinear spin motion in ferromagnets is considered with nonlinearity due to three factors: (i) the sample is prepared in a strongly nonequilibrium state, so that evolution equations cannot be linearized as would be admissible for spin motion not too far from equilibrium, (ii) the system considered consists of interacting electron and nuclear spins coupled with each other via hyperfine forces, and (iii) the sample is inserted into a coil of a resonant electric circuit producing a resonator feedback field. Due to these nonlinearities, coherent motion of spins can develop, resulting in their ultrafast relaxation. A complete analysis of mechanisms triggering such a coherent motion is presented. This type of ultrafast coherent relaxation can be used for studying intrinsic properties of magnetic materials. [S0163-1829(99)02726-5]

I. INTRODUCTION

There are several different types of spin dynamics in condensed matter, which can be distinguished according to whether the sample studied is in equilibrium, weak nonequilibrium, or strong nonequilibrium. Microscopic spin oscillations in equilibrium magnetic materials are related to magnons and are studied by scattering techniques, such as neutron[1] or light[2] scattering. Small deviations from equilibrium, caused by an alternating external field, are characteristic of resonance experiments, like electron-spin resonance[3] or nuclear magnetic resonance.[4] However, when the initial state of a spin system is made strongly nonequilibrium, several types of spin relaxation can occur. If there are no transverse external fields acting on the spins, they relax to an equilibrium state by an exponential law with a longitudinal relaxation time T_1. When the motion of spins is triggered, at the initial time, by a transverse magnetic field, the relaxation is again exponential, but with a transverse relaxation time T_2 which is usually much shorter than T_1.

A rather different relaxation regime from a strongly nonequilibrium initial state arises if the spin system is coupled to a resonator. This can be done by inserting the sample into a coil connected with a resonance electric circuit. Because of the action of the resonator feedback field, the motion of spins can become highly coherent resulting in their ultrafast relaxation during a characteristic time much shorter than T_2.[5] This latter type of coherent-spin relaxation from a strongly nonequilibrium state in the presence of coupling with a resonator is the most difficult to realize experimentally and to describe theoretically. Experimental difficulties have been overcome in a series of observations of this phenomenon for a system of nuclear spins in a paramagnetic matrix.[6–10] A theory of the coherent-spin relaxation could be based on the phenomenological Bloch equations, but solely for the case when the process is triggered by a sufficiently strong coherent pulse thrust on spins at the initial time, so that spin interactions are of no importance and only the resonator field plays a role. However, the most interesting case is when the coherent relaxation develops in a self-organized way from an initially incoherent state, with no external coherent pulses triggering the process. For such a *self-organized coherent relaxation* spin interactions are of crucial importance. Then the Bloch equations become inapplicable and one has to resort to microscopic models.

A microscopic approach for describing coherent processes in spin systems has been recently developed[11,12] and applied to a system of nuclear spins interacting through dipole forces. It was shown that the main role in initiating self-organized coherent relaxation is played by the anisotropic (so-called nonsecular) part of the dipole interactions.

In the present paper we extend the microscopic theory of coherent-spin relaxation[11,12] to a much wider class of materials. We consider a rather general Hamiltonian including both nuclear as well as electron subsystems interacting with each other through hyperfine forces. The electrons can possess a long-range magnetic order as in ferromagnets or ferrimagnets, and magnetocrystalline anisotropy is taken into account. A general investigation of strongly nonequilibrium nonlinear processes in realistic magnetic materials is of interest by itself and can also be useful for many applications. For instance, the self-organized coherent relaxation, being quite different from other types of relaxations, may give additional information on intrinsic properties of magnetic materials. The ultrafast relaxation can be employed for repolarizing solid-state targets used in scattering experiments.[10,13] Coherent effects in spin systems, being similar to their coherent counterparts in optics,[14,15] could be used for analogous purposes but in another frequency region. For example, spin masers[16–18] can be realized. The sensitivity of the characteristic times of coherent relaxation to initial conditions could be used for creating ultrasensitive particle detectors.[19]

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II. ELECTRON-NUCLEAR SPIN HAMILTONIAN

To make our consideration applicable to a wide class of magnetic materials, we take a rather general Hamiltonian

$$\hat{H}=\hat{H}_e+\hat{H}_n+\hat{H}_{\rm int}, \tag{1}$$

describing a realistic situation where the sample contains electrons with a Hamiltonian $\hat{H}_e$ and nuclei with a Hamiltonian $\hat{H}_n$, their interaction being given by $\hat{H}_{\rm int}$. The electron Hamiltonian is

$$\hat{H}_e=-\frac{1}{2}\sum_{i\neq j} J_{ij}\vec{S}_i\cdot\vec{S}_j-\mu_e\sum_i \vec{B}\cdot\vec{S}_i, \tag{2}$$

where J_{ij} is an exchange interaction; the indices $i,j=1,2,\ldots,N_e$ here enumerate electrons; $\vec{S}_i$ is a spin operator; $\mu_e=g_e\mu_B$, with g_e being the electronic gyromagnetic ratio and μ_B being the Bohr magneton; $\vec{B}$ is a magnetic field. The nuclear Hamiltonian has the form[20] commonly accepted in the theory of nuclear magnetic resonance,

$$\hat{H}_n=\frac{1}{2}\sum_{i\neq j}\sum_{\alpha\beta} C_{ij}^{\alpha\beta}I_i^\alpha I_j^\beta-\mu_n\sum_i \vec{B}\cdot\vec{I}_i, \tag{3}$$

in which $i,j=1,2,\ldots,N_n$ enumerate nuclei with dipole interactions

$$C_{ij}^{\alpha\beta}=\frac{\mu_n^2}{r_{ij}^3}(\delta_{\alpha\beta}-3n_{ij}^\alpha n_{ij}^\beta) \tag{4}$$

between each other, where $\mu_n=g_n\mu_N$, g_n being the nuclear gyromagnetic ratio, μ_N, is the nuclear magneton, and $r_{ij}\equiv|\vec{r}_{ij}|$, $\vec{n}_{ij}\equiv\vec{r}_{ij}/r_{ij}$, $\vec{r}_{ij}\equiv\vec{r}_i-\vec{r}_j$; the indices α and β label the components of Cartesian vectors ($\alpha,\beta=x,y,z$); $\vec{I}_i$ is a nuclear-spin operator. The general form of the hyperfine interactions[20,21] between electrons and nuclei is

$$\hat{H}_{\rm int}=A\sum_i \vec{S}_i\cdot\vec{I}_i+\frac{1}{2}\sum_{i\neq j}\sum_{\alpha\beta} A_{ij}^{\alpha\beta}S_i^\alpha I_j^\beta, \tag{5}$$

containing an isotropic contact part with an interaction intensity A and a dipole part with the interactions

$$A_{ij}^{\alpha\beta}=\frac{\mu_e\mu_n}{r_{ij}^3}(\delta_{\alpha\beta}-3n_{ij}^\alpha n_{ij}^\beta). \tag{6}$$

The total magnetic field is the sum

$$\vec{B}=H_0\vec{e}_z+H_1\vec{e}_x, \quad H_1=H_a+H, \tag{7}$$

of an external magnetic field in the z direction and of a transverse field including an effective field of a transverse magnetocrystalline anisotropy[22] and a feedback field H of a resonator. The longitudinal part of the magnetocrystalline anisotropy can be included into the external magnetic field H_0.

In the preceding formulas we have used, for simplicity, the same indices, i and j, to enumerate electrons and nuclei, keeping in mind that for each particular case these indices run over different sets, so that for electrons $i=1,2,\ldots,N_e$ and for nuclei $i=1,2,\ldots,N_n$. The corresponding electron and nuclear densities, $\rho_e\equiv N_e/V$ and $\rho_n\equiv N_n/V$, where V is the volume of a sample, are, in general, different.

The resonator coil is directed along the x axis, so that the current induced in it is caused by the motion of the transverse magnetization

$$M_x=\frac{1}{V}\sum_i(\mu_e\langle S_i^x\rangle+\mu_n\langle I_i^x\rangle), \tag{8}$$

where the angle brackets mean statistical averaging. The resonant electric circuit is characterized by a natural frequency ω, ringing time γ_3, and quality factor Q, given by

$$\omega\equiv\frac{1}{\sqrt{LC}}, \quad \gamma_3\equiv\frac{\omega}{2Q}, \quad Q\equiv\frac{\omega L}{R}, \tag{9}$$

where L, C, and R are inductance, capacity, and resistance, respectively. The resonator feedback field is given[12] by the Kirchhoff equation

$$\frac{dH}{dt}+2\gamma_3 H+\omega^2\int_0^t H(\tau)d\tau=-4\pi\eta\frac{dM_x}{dt}, \tag{10}$$

in which η is a filling factor.

III. COUPLED SYSTEM OF EQUATIONS

Spin dynamics is defined by the Heisenberg equations for the electron spin operators $S_i^\pm$ and S_i^z, and for the nuclear-spin operators $I_i^\pm$ and I_i^z, where $S_i^\pm=S_i^x\pm iS_i^y$, $I_i^\pm=I_i^x\pm iI_i^y$. These equations are coupled to each other by hyperfine interactions between electron and nuclear spins. Besides that, both types of spin motion are coupled with the resonator through the resonator feedback field, defined by Eq. (10), and the average transverse magnetization (8) expressed by means of the average spins. To describe the dynamics of spins coupled with each other as well as with a resonator, we need to derive the time evolution equations for the average electron and nuclear spins

$$x\equiv\frac{1}{N_e}\sum_i\langle S_i^-\rangle, \quad z\equiv\frac{1}{N_e}\sum_i\langle S_i^z\rangle, \tag{11}$$

$$u\equiv\frac{1}{N_n}\sum_i\langle I_i^-\rangle, \quad s\equiv\frac{1}{N_n}\sum_i\langle I_i^z\rangle. \tag{12}$$

The main steps of deriving these equations are the same as in Refs. 11,12 except that now the Hamiltonian (1) is more complicated.

We introduce the Zeeman frequencies

$$\omega_e\equiv\mu_e H_0, \quad \omega_n\equiv\mu_n H_0, \tag{13}$$

and the anisotropy parameters

$$\alpha_e\equiv\mu_e H_a, \quad \alpha_n\equiv\mu_n H_a, \tag{14}$$

where the Planck constant is set $\hbar\equiv1$. We use the notation

$$\xi_0\equiv\frac{1}{2}\sum_{j(\neq i)}(\overline{a}_{ij}\langle I_j^z\rangle+\overline{c}_{ij}\langle I_j^+\rangle+\overline{c}_{ij}^*\langle I_j^-\rangle), \tag{15}$$

$$\xi \equiv \frac{1}{2} \sum_{j(\neq i)} (\overline{e}_{ij} \langle I_j^- \rangle + 2\overline{b}_{ij} \langle I_j^+ \rangle + 2\overline{c}_{ij} \langle I_j^z \rangle), \tag{16}$$

in which

$$\overline{a}_{ij} \equiv A_{ij}^{zz}, \quad \overline{e}_{ij} \equiv \frac{1}{2}(A_{ij}^{xx} + A_{ij}^{yy}),$$

$$\overline{b}_{ij} \equiv \frac{1}{4}(A_{ij}^{xx} - A_{ij}^{yy} - 2iA_{ij}^{xy}), \quad \overline{c}_{ij} \equiv \frac{1}{2}(A_{ij}^{xz} - iA_{ij}^{yz}),$$

$A_{ij}^{\alpha\beta}$ being given in Eq. (6). Also we write

$$\varphi_0 \equiv \sum_{j(\neq i)} [(a_{ij} - e_{ij}) \langle I_j^z \rangle + c_{ij} \langle I_j^+ \rangle + c_{ij}^* \langle I_j^- \rangle + \tfrac{1}{2} (\overline{a}_{ij} \langle S_j^z \rangle + \overline{c}_{ij} \langle S_j^+ \rangle + \overline{c}_{ij}^* \langle S_j^- \rangle)], \tag{17}$$

$$\varphi \equiv \sum_{j(\neq i)} \left[2(b_{ij} \langle I_j^+ \rangle + c_{ij} \langle I_j^z \rangle) + \frac{1}{2}(\overline{e}_{ij} \langle S_j^- \rangle + 2\overline{b}_{ij} \langle S_j^+ \rangle + 2\overline{c}_{ij} \langle S_j^z \rangle) \right], \tag{18}$$

where

$$a_{ij} \equiv C_{ij}^{zz}, \quad e_{ij} \equiv \frac{1}{2}(C_{ij}^{xx} + C_{ij}^{yy}),$$

$$b_{ij} \equiv \frac{1}{4}(C_{ij}^{xx} - C_{ij}^{yy} - 2iC_{ij}^{xy}), \quad c_{ij} \equiv \frac{1}{2}(C_{ij}^{xz} - iC_{ij}^{yz}),$$

$C_{ij}^{\alpha\beta}$ being defined in Eq. (4). Equations (15)–(18) describe local random fields caused by spin fluctuations. In the uniform approximation, all these quantities would be zero, because of the properties of dipole interactions. However, these local fields cannot be neglected, since they play a crucial role at the initial stage of spin relaxation. Therefore, they must be retained and treated as local random variables. For other terms in the evolution equations, having a long-range nature in real space, one may employ the semiclassical approximation. This approach of using for long-range terms the semiclassical approximation complemented by the stochastic quantization of short-range terms has been developed in Refs. 11,12.

Following these steps and taking into account the longitudinal γ_1, Γ_1 and transverse γ_2, Γ_2 attenuations for the electron and nuclei, respectively, we obtain the evolution equations for the electron-spin variables (11) and nuclear-spin variables (12). For the transverse and longitudinal electron spins we have, respectively,

$$\frac{dx}{dt} = i(\omega_e + i\gamma_2 - \xi_0 - As)x - i(\alpha_e + \mu_e H - \xi - Au)z, \tag{19}$$

$$\frac{dz}{dt} = \frac{i}{2}(\alpha_e + \mu_e H - \xi - Au)x^* - \frac{i}{2}(\alpha_e + \mu_e H - \xi^* - Au^*)x - \gamma_1(z - \sigma), \tag{20}$$

where γ_1 and γ_2 are attenuation parameters and σ is the stationary value of z. The variable x is complex, while z is real, so we should add either an equation for x^* or for $|x|$. It is convenient to consider

$$\frac{d|x|^2}{dt} = -2\gamma_2 |x|^2 + i(\alpha_e + \mu_e H - \xi^* - Au^*)zx - i(\alpha_e + \mu_e H - \xi - Au)zx^*. \tag{21}$$

In the case of nuclear spins, we obtain

$$\frac{du}{dt} = i(\omega_n + i\Gamma_2 - \varphi_0 - Az)u - i(\alpha_n + \mu_n H - \varphi - Ax)s, \tag{22}$$

$$\frac{ds}{dt} = \frac{i}{2}(\alpha_n + \mu_n H - \varphi - Ax)u^* - \frac{i}{2}(\alpha_n + \mu_n H - \varphi^* - Ax^*)u - \Gamma_1(s - \mathfrak{s}), \tag{23}$$

where Γ_1 and Γ_2 are the longitudinal and transverse attenuations, respectively, and $\mathfrak{s}$ is the stationary value of s. In addition, we shall need

$$\frac{d|u|^2}{dt} = -2\Gamma_2 |u|^2 + i(\alpha_n + \mu_n H - \varphi^* - Ax^*)su - i(\alpha_n + \mu_n H - \varphi - Ax)su^*. \tag{24}$$

All equations (19)–(24) contain the resonator feedback field H described by Eq. (10). The latter can be transformed[12] to the integral feedback equation

$$H = -4\pi\eta \int_0^t G(t - \tau) dM_x(\tau), \tag{25}$$

expressed through a Stieltjes integral with the Green function

$$G(t) = \left(\cos \omega_3 t - \frac{\gamma_3}{\omega_3} \sin \omega_3 t \right) \exp(-\gamma_3 t)$$

and the differential measure dM_x with M_x defined in Eq. (8). Here the effective frequency is $\omega_3 \equiv \sqrt{\omega^2 - \gamma_3^2}$.

The system of seven nonlinear equations (19)–(25) determines the dynamics of electron and nuclear spins coupled with each other as well as with a resonator.

IV. SCALE SEPARATION APPROACH

Our aim here is to study the strongly nonequilibrium regimes of spin motion. This problem is different from considering the equilibrium properties of coupled electron and nuclear spins.[23–26] An additional complication, in our case, arises from the coupling of spins with a resonator by means of the feedback equation (25). To solve Eqs. (19)–(25), we employ the scale separation approach[11,12] which is a generalization of the averaging techniques of dynamical theory[27,28] to statistical systems.

To understand what different time scales exist for the system considered, we need to specify what small parameters we have. Since we have a sample coupled with a resonator, some small parameters should appear by concretizing the

corresponding resonance conditions, assuming the ringing width is much smaller than the natural frequency,

$$\frac{\gamma_3}{\omega} \ll 1. \tag{26}$$

The resonator natural frequency can be tuned either to the frequency of electron spin resonance ω_e, so that

$$\left|\frac{\Delta_e}{\omega_e}\right| \ll 1, \quad \Delta_e \equiv \omega - \omega_e, \tag{27}$$

or to the frequency of nuclear magnetic resonance

$$\omega_N \equiv \omega_n - A z_{\rm eff}, \tag{28}$$

in which $z_{\rm eff}$ is the longitudinal electron spin z averaged over the period $2\pi/\omega$, so that

$$\left|\frac{\Delta_N}{\omega_N}\right| \ll 1, \quad \Delta_N \equiv \omega - \omega_N. \tag{29}$$

We assume that the external magnetic field H_0 is sufficiently strong that

$$\left|\frac{\alpha_e}{\omega_e}\right| \ll 1, \quad \left|\frac{\mu_e H_{\rm eff}}{\omega_e}\right| \ll 1, \quad \left|\frac{A}{\omega_e}\right| \ll 1, \tag{30}$$

where $H_{\rm eff}$ is the resonator feedback field averaged over a period $2\pi/\omega$. We also assume

$$\left|\frac{\gamma_1}{\omega_e}\right| \ll 1, \quad \left|\frac{\gamma_2}{\omega_e}\right| \ll 1. \tag{31}$$

Then from Eqs. (19) to (21) it follows that the variables z and $|x|^2$ are to be treated as slow compared to the fast variable x. Similarly, for nuclei we assume

$$\left|\frac{\alpha_n}{\omega_N}\right| \ll 1, \quad \left|\frac{\mu_n H_{\rm eff}}{\omega_N}\right| \ll 1, \quad \left|\frac{A x_{\rm eff}}{\omega_N}\right| \ll 1, \tag{32}$$

where the subscript eff means again that the corresponding quantity is averaged over $2\pi/\omega$, and we keep in mind the usual inequalities

$$\left|\frac{\Gamma_1}{\omega_N}\right| \ll 1, \quad \left|\frac{\Gamma_2}{\omega_N}\right| \ll 1. \tag{33}$$

Then Eqs. (22), (23), and (24) show that the variables s and $|u|^2$ are slow compared to the fast variable u. As the nuclear magnetic moment μ_n is much smaller than that of an electron μ_e, we have the inequalities

$$\left|\frac{\mu_n}{\mu_e}\right| \ll 1, \quad \frac{\Gamma_1}{\gamma_1} \ll 1, \quad \frac{\Gamma_2}{\gamma_2} \ll 1. \tag{34}$$

Then, comparing Eqs. (19) and (22), we see that the variable u is slow compared to the fast variable x. And the comparison of Eqs. (21) and (24) tells us that $|u|^2$ is slow compared to the faster $|x|^2$.

One more condition assumed is related to the local random fields (15)–(18). These local fields define the parameters of inhomogeneous broadening due to the electron-nuclear, $\Gamma_{\rm en}$, and nuclear-nuclear $\Gamma_{\rm nn}$ interactions. These widths are assumed to satisfy

$$\left|\frac{\Gamma_{\rm en}}{\omega_N}\right| \ll 1, \quad \left|\frac{\Gamma_{\rm nn}}{\omega_N}\right| \ll 1. \tag{35}$$

Using conditions (30) and (32), we may simplify the feedback equation (25) to

$$H = -2\,{\rm Re}\left(\beta_e \frac{dx}{dt} + \beta_n \frac{du}{dt}\right), \tag{36}$$

in which the parameters

$$\beta_e \equiv \pi^2 \eta \frac{\mu_e \rho_e}{\omega}, \quad \beta_n \equiv \pi^2 \eta \frac{\mu_n \rho_n}{\omega} \tag{37}$$

characterize the effective coupling of the sample with the resonator. The details are given in the Appendix. Substituting into Eq. (36) the derivatives from Eqs. (19) and (22), we find

$$H = -2\,{\rm Re}\, i\beta_e[(\omega_e - As - \xi_0 + i\gamma_2)x + (Au + \xi)z] - 2\,{\rm Re}\, i\beta_n[(\omega_n - Az - \varphi_0 + i\Gamma_2)u + (Ax + \varphi)s]. \tag{38}$$

The feedback field (38) is to be substituted into Eqs. (19)–(24).

Then we solve Eq. (19) treating there all slow variables as quasi-integrals of motion. The solution reads

$$x = (x_0 - \overline{x})\exp\{(i\overline{\Omega}_e - \overline{\gamma}_2)t\} + \overline{x}, \tag{39}$$

where $x_0 = x(0)$ and

$$\overline{\Omega}_e \equiv \omega_e - As - \xi_0 - \mu_e\beta_e\gamma_2 z,$$

$$\overline{\gamma}_2 \equiv \gamma_2 + \mu_e\beta_e(\omega_e - As - \xi_0)z,$$

$$\overline{x} \equiv \frac{1}{\overline{\Omega}_e}\{(\alpha_e - Au - \xi)z + i\mu_e\beta_e[A(u^* - u) + \xi^* - \xi]z^2\}.$$

After this, we solve Eq. (22), keeping the slow variables fixed and averaging the fast variables $x(t)$ and $z(t)$ to obtain $x_{\rm eff}$ and $z_{\rm eff}$. Since $x(t)$ is already known, we have

$$x_{\rm eff} = \begin{cases} A_e z/\Omega_e, & \omega \approx \omega_e \\ 0, & \omega \approx \omega_N, \end{cases}$$

where $\Omega_e \equiv \omega_e - As - \xi_0$ and $A_e \equiv \alpha_e - Au - \xi$. The solution of Eq. (22) is

$$u = (u_0 - \overline{u})\exp\{(i\overline{\Omega}_n - \overline{\Gamma}_2)t\} + \overline{u}, \tag{40}$$

where $u_0 \equiv u(0)$ and

$$\overline{\Omega}_n \equiv \omega_n - A(1 + \mu_n\beta_e s)z_{\rm eff} - \varphi_0 - \mu_n\beta_n\Gamma_2 s,$$

$$\overline{\Gamma}_2 \equiv \Gamma_2 + \mu_n\beta_n(\omega_n - Az_{\rm eff} - \varphi_0)s,$$

$$\overline{u} \equiv \frac{1}{\overline{\Omega}_n}[(\alpha_n - \varphi)s + i\mu_n\beta_e(\xi^* - \xi)sz_{\rm eff}].$$

The solutions (39) and (40) are to be substituted into the equations for the slow variables, with the right-hand sides of

the latter equations being averaged over time and over random local fields according to the rule

$$\langle\langle f(t,\psi)\rangle\rangle = \int \left[\frac{\omega}{2\pi} \int_0^{2\pi/\omega} f(t,\psi)dt \right] dm(\psi),$$

with the stochastic measure $m(\psi)$ such that

$$\langle\langle \xi_0 \rangle\rangle = \langle\langle \xi \rangle\rangle = \langle\langle \xi_0 \xi \rangle\rangle = 0,$$

$$\langle\langle \varphi_0 \rangle\rangle = \langle\langle \varphi \rangle\rangle = \langle\langle \varphi_0 \varphi \rangle\rangle = 0,$$

$$\langle\langle \xi_0 \varphi_0 \rangle\rangle = \langle\langle \xi_0 \varphi \rangle\rangle = \langle\langle \xi \varphi_0 \rangle\rangle = \langle\langle \xi \varphi \rangle\rangle = 0,$$

$$\langle\langle \xi_0^2 \rangle\rangle = \langle\langle |\xi|^2 \rangle\rangle = \gamma_*^2 = \Gamma_{\rm en}^2,$$

$$\langle\langle \varphi_0^2 \rangle\rangle = \langle\langle |\varphi|^2 \rangle\rangle = \Gamma_*^2 = \Gamma_{\rm nn}^2 + \Gamma_{\rm en}^2.$$

The constants γ_* and Γ_* are the parameters of inhomogeneous broadening caused by hyperfine electron-nuclear dipole interactions and by nuclear dipole interactions.

We introduce the effective coupling parameters

$$g_e \equiv \pi^2 \eta \frac{\rho_e \mu_e^2 \omega_E}{\gamma_2 \omega} \left(1 + \frac{\rho_n \mu_n A s}{\rho_e \mu_e \omega_E} \right), \tag{41}$$

$$g_n \equiv \pi^2 \eta \frac{\rho_n \mu_n^2 \omega_N}{\Gamma_2 \omega} \left(1 + \frac{\rho_e \mu_e A z_{\rm eff}}{\rho_n \mu_n \omega_N} \right), \tag{42}$$

characterizing the strength of coupling between electron or nuclear spins, respectively, and the resonator. The effective frequencies are

$$\omega_E \equiv \omega_e - As, \quad \omega_N \equiv \omega_n - A z_{\rm eff}, \tag{43}$$

which are the frequencies of the electron-spin resonance and nuclear magnetic resonance.

Also, we define slow variables for electrons,

$$v = |x|^2 - \frac{\alpha_e^2 + A^2 |u|^2 + \gamma_*^2}{\omega_E^2} z^2, \tag{44}$$

and for nuclei

$$w = |u|^2 - \frac{\alpha_n^2 + \Gamma_*^2 + \delta^2}{\omega_N^2} s^2, \tag{45}$$

where

$$\delta \equiv \sqrt{2}\pi^2 \eta \gamma_* \frac{\rho_e \mu_e \mu_n}{\omega_N} z_{\rm eff}. \tag{46}$$

Accomplishing all these steps, we obtain from Eqs. (20)–(24)

$$\frac{dz}{dt} = \gamma_2 g_e v - \gamma_1 (z - \sigma), \tag{47}$$

$$\frac{dv}{dt} = -2\gamma_2 (1 + g_e z) v, \tag{48}$$

$$\frac{ds}{dt} = \Gamma_2 g_n w - \Gamma_1 (s - \zeta), \tag{49}$$

$$\frac{dw}{dt} = -2\Gamma_2 (1 + g_n s) w. \tag{50}$$

Equations (47)–(50) define the averaged motion of slow variables. We require their solutions, since all observable quantities can be expressed through them.

V. NUCLEAR-SPIN DYNAMICS

Equations (47) to (50) show that the electron-spin dynamics is qualitatively similar to that of nuclear spins, but there are three main points distinguishing electron from nuclear relaxation. First, electron-spin processes are usually much faster than nuclear processes,[3] which is related to the fact that $\gamma_2 \gg \Gamma_2$. Second, as the electronic magnetic moment is three orders of magnitude larger than the nuclear magneton, electron-spin motion is much less influenced by the presence of nuclei than the motion of nuclear spins by the existence of electrons. Third, the stronger influence of electrons on the motion of nuclear spins is caused by a long-range magnetic order that more readily occurs in electronic systems than in nuclear ones. Therefore, nuclear-spin dynamics is a little more complicated but at the same time richer than the dynamics of electron spins.

Suppose that the electron spins either were not perturbed at the initial time or, if perturbed, that fast electron processes have already been relaxed to their stationary state. Let us study the dynamics of nuclear spins that were initially prepared in a strongly nonequilibrium state. We denote the initial conditions for the nuclear-spin variables as

$$s(0) = s_0, \quad w(0) = w_0, \quad u(0) = u_0. \tag{51}$$

If relaxation of nuclear spins lasts for times of order $T_1 \equiv \Gamma_1^{-1}$, this would mean that coherent processes do not develop. Such a case would be of no interest for us, since our aim here is to investigate the fast coherent relaxation. Hence, we shall consider times such that $t \ll \Gamma_1^{-1}$. In this case, we may omit in Eq. (49) the term containing Γ_1. Then we can solve Eqs. (49) and (50) analytically obtaining for the longitudinal nuclear spin

$$s = \frac{\gamma_0}{g\Gamma_2} \tanh\left(\frac{t - t_0}{\tau_0} \right) - \frac{1}{g}, \tag{52}$$

where, for the sake of simplicity, we write $g \equiv g_n$, and

$$w = \left(\frac{\gamma_0}{g\Gamma_2} \right)^2 \operatorname{sech}^2\left(\frac{t - t_0}{\tau_0} \right). \tag{53}$$

Here $\gamma_0 \equiv \tau_0^{-1}$ is the relaxation width given by the equation

$$\gamma_0 = \Gamma_2 [(1 + g s_0)^2 + g^2 w_0]^{1/2} \tag{54}$$

with τ_0 being the relaxation time, and t_0 being the delay time,

$$t_0 = \frac{\tau_0}{2} \ln \left| \frac{\gamma_0 - \Gamma_2 (1 + g s_0)}{\gamma_0 + \Gamma_2 (1 + g s_0)} \right|. \tag{55}$$

According to Eq. (45), the modulus squared of the transverse nuclear spin is

$$|u|^2=\left(\frac{\gamma_0}{g\Gamma_2}\right)^2 \operatorname{sech}^2\left(\frac{t-t_0}{\tau_0}\right)+\frac{\alpha_n^2+\Gamma_*^2+\delta^2}{\omega_N^2}s^2. \quad (56)$$

If the coupling parameter g, is small, $g\ll 1$, then Eqs. (49) and (50) show that the relaxation of nuclear spins follows the standard exponential law with the relaxation times $T_1\equiv\Gamma_1^{-1}$ and $T_2\equiv\Gamma_2^{-1}$. This trivial regime is not interesting for us, so we concentrate attention on the case of strong coupling, when $g\gg 1$. Then the relaxation width (54) can be written as

$$\gamma_0=\Gamma_2\left(g\sqrt{s_0^2+w_0}+\frac{s_0}{\sqrt{s_0^2+w_0}}\right). \quad (57)$$

For the delay time (55), we find

$$t_0=\frac{\tau_0}{2}\ln\left|\frac{g(s_0^2+w_0)+s_0-(1+gs_0)\sqrt{s_0^2+w_0}}{g(s_0^2+w_0)+s_0+(1+gs_0)\sqrt{s_0^2+w_0}}\right|. \quad (58)$$

For the relaxation time, after using again the inequality $g\gg 1$, we have

$$\tau_0=\frac{T_2}{g\sqrt{s_0^2+w_0}}. \quad (59)$$

A large value of the coupling constant g means, according to its definition in Eq. (42), that nuclear spins are strongly correlated with each other by means of an effective interaction through the resonator feedback field. As a result of this correlation they move coherently, which leads to the nonzero value of $w\approx|u|^2$. Recall that, by definition (12), $u=0$ for incoherent spins. Coherent motion of the spins results in their ultrafast relaxation, which follows from Eq. (59) yielding $\tau_0\ll T_2$ when $g\gg 1$. This is why the characteristic time (59) can be called the *coherent relaxation time*. Notice also that the coupling parameter (42) is proportional to the number of nuclei N_n, and so $\tau_0\sim 1/g\sim 1/N_n$. Such a dependence of the relaxation time, $\tau_0\sim 1/N_n$, on the number of radiators is typical for coherent processes that in optics are called superradiance.[14,15]

The relaxation characteristics, as is seen, essentially depend on the initial conditions (51). If $u_0\neq 0$ at the initial time, this implies that an initial coherence is imposed on the spins, which can be done by means of a short external pulse of a transverse field. When $u_0\sim 1$, then we have $w_0\approx|u_0|^2$, since the second term in Eq. (45) is small. In this case we get the regime of triggered relaxation.[12]

A much more interesting question concerns how the relaxation of nuclear spins starts when no initial coherence is thrust upon the spins. This problem is also more important than consideration of the case when relaxation is triggered by external fields. For, when the relaxation is initiated not by external forces but by internal interactions, the relaxation characteristics, such as the delay time and the relaxation time, significantly depend on the parameters of these internal interactions. Therefore, the self-organized relaxation reflects (and provides information about) the intrinsic properties of matter.

Let us consider the *self-organized relaxation* in the system of nuclear spins strongly coupled with a resonator. That is, we analyze the case corresponding to the conditions:

$$u_0=0, \quad s_0\neq 0, \quad g|s_0|\gg 1. \quad (60)$$

If $u_0=0$, then, according to relation (45), we have $|w_0|\ll s_0^2$. This permits us to simplify the relaxation width (57) getting

$$\gamma_0=\frac{\Gamma_2}{2s_0^2}[g|s_0|(2s_0^2+w_0)+\varepsilon(2s_0^2-w_0)], \quad (61)$$

where $\varepsilon\equiv$ sgn s_0. The delay time (58) becomes

$$t_0=\frac{\tau_0}{2}\ln\left|\frac{2(1-\varepsilon)s_0^2+w_0}{2(1+\varepsilon)s_0^2+w_0}\right|, \quad (62)$$

and for the relaxation time (59), we get

$$\tau_0=\frac{2s_0^2-w_0}{2g|s_0|^3\Gamma_2}\simeq\frac{T_2}{g|s_0|}. \quad (63)$$

The delay time (62) strongly depends on the sign of the initial polarization of nuclear spins, $\varepsilon\equiv$ sgn s_0. When this initial polarization is directed along the external magnetic field H_0, i.e., along the z axis, then

$$t_0=\frac{\tau_0}{2}\ln\left|\frac{w_0}{4s_0^2}\right| \quad (\varepsilon=1), \quad (64)$$

and if the initial polarization is directed opposite to the external field, then

$$t_0=\frac{\tau_0}{2}\ln\left|\frac{4s_0^2}{w_0}\right| \quad (\varepsilon=-1). \quad (65)$$

In the case when the initial polarization is along the z axis, Eq. (64) shows that $t_0<0$, since $|w_0|\ll s_0^2$. Then the function (53) quickly decreases starting from $t=0$. This function $w(t)$, being proportional to $|u|^2$, describes the degree of coherence in the motion of nuclear spins. In turn, the function $|u|^2$ is proportional to the power flow in the resonator circuit and, thus, is a directly measurable quantity.[11,12] Hence, when $w(t)$ quickly decreases starting from the initial time $t=0$ and $|w_0|\ll s_0^2$, this means that no noticeable coherence develops in the system.

In contrast, if the initial polarization of nuclear spins is directed opposite to the external magnetic field, so that the system is prepared in a strongly nonequilibrium state, then, from Eq. (65), there is a positive solution for the delay time $t_0>0$. In such a case, the function (53) increases from its initial value w_0, reaching a maximum at $t=t_0$, when

$$w(t_0)=s_0^2, \quad s(t_0)=-\frac{1}{g}. \quad (66)$$

This means that a self-organized coherent pulse develops with a maximum at $t=t_0$, which explains why t_0 is called the delay time.

Combining both the cases, Eqs. (64) and (65), into one and substituting the expressions for τ_0 and w_0, we obtain

$$t_0=\frac{T_2}{2gs_0}\ln\left|\frac{\alpha_n^2+\Gamma_{\rm en}^2+\Gamma_{\rm nn}^2+\delta^2}{4\omega_N^2}\right|. \tag{67}$$

This is the central formula for analyzing which internal microscopic mechanisms are responsible for the self-organized development of coherent relaxation. Each of these internal mechanisms is related to the corresponding parameter entering formula (67). Among such internal causes that may trigger self-organized coherence, we have the transverse magnetocrystalline anisotropy, α_n; the dipole part of the hyperfine interactions, characterized by the parameter $\Gamma_{\rm en}$; the dipole interactions between nuclear spins $\Gamma_{\rm nn}$; and the parameter δ defined in Eq. (46), which is due to the simultaneous existence of the hyperfine interactions, of coupling with a resonator, and of the magnetization of electron spins.

The relaxation time (54), for the case of self-organized relaxation, when $u_0=0$, reads

$$\tau_0=T_2\left[(1+gs_0)^2+g^2s_0^2\frac{\alpha_n^2+\Gamma_{\rm en}^2+\Gamma_{\rm nn}^2+\delta^2}{\omega_N^2}\right]^{-1/2}. \tag{68}$$

This demonstrates that the value of the relaxation time (68) depends mainly on the strength g of coupling with a resonator. In this way, the delay time (67) and the relaxation time (68) are related to different characteristics of the system considered.

In order to decide what kind of interactions, direct dipole interactions between nuclei or hyperfine interactions, influences more the values of the characteristic times, we should compare the corresponding widths $\Gamma_{\rm nn}\sim\rho_n\mu_n^2$ and $\Gamma_{\rm en}\sim\rho\mu_e\mu_n$, where $\rho\approx\min\{\rho_e,\rho_n\}$. There are two limiting cases. The first is when $\rho_n\leqslant\rho_e$, and then

$$\frac{\Gamma_{\rm nn}}{\Gamma_{\rm en}}\sim\frac{\mu_n}{\mu_e}\sim10^{-3}\quad(\rho_n\leqslant\rho_e). \tag{69}$$

Hence, when the density of nuclei is lower or comparable with that of electrons, nuclear dipole interactions are negligibly small compared to the hyperfine interactions between nuclei and electrons. Another case is when the density of nuclei is much higher than that of electrons; then

$$\frac{\Gamma_{\rm nn}}{\Gamma_{\rm en}}\sim\frac{\rho_n\mu_n}{\rho_e\mu_e}\sim\frac{\rho_n}{\rho_e}10^{-3}\quad(\rho_n\geqslant\rho_e). \tag{70}$$

Thus, the nuclear dipole interactions become stronger than the hyperfine interactions only when the density of nuclei surpasses by three orders of magnitude the density of electrons.

In equilibrium theory, the influence of hyperfine interactions is often modeled by the effective Suhl-Nakamura forces directly acting between nuclear spins.[21] These forces are responsible for the appearance of nuclear-spin waves corresponding to well-defined excitations, even at those temperatures where the nuclear spins are completely disordered. The underlying cause of the formation of the nuclear-spin waves is the existence of magnetic long-range order in the electronic subsystem, which defines both the long-range interaction radius of the Suhl-Nakamura force and its existence as such. The effective Suhl-Nakamura force describes an indirect interaction of nuclear spins through magnetically ordered electrons.[21]

For strongly nonequilibrium processes, such as those considered in this paper, the role of the magnetic order of the electrons is essentially different. This order does strongly influence several important characteristics. For instance, in addition to the usual shift of the nuclear magnetic resonance frequency $\omega_N=\omega_n-Az_{\rm eff}$, it leads to the appearance of parameter (46) playing for nuclei the role of an additional inhomogeneous width. Nevertheless, even if this order is absent, so that $z_{\rm eff}\rightarrow0$, coherent nuclear-spin relaxation can exist. When $z_{\rm eff}\rightarrow0$, the Suhl-Nakamura force is not well defined, but the hyperfine interactions do not stop existing. These interactions define the width $\Gamma_{\rm en}$, which is not zero even if the electronic magnetic order is absent. Thus, the presence of hyperfine interactions is already important, even when there is no long-range magnetic order, when the Suhl-Nakamura force and nuclear-spin waves are not well defined.

However, it is worth emphasizing that the appearance of electronic magnetic order can strongly change the values of the characteristic parameters. Thus, one of the most important parameters is the effective coupling (42) describing the coupling of nuclear spins with a resonator. The value of this parameter essentially depends on whether $z_{\rm eff}$ is zero or not. The appearance of magnetic order in the electronic system can change the value of the parameter (42) by several orders of magnitude which, in turn, drastically changes the values of the delay time t_0 and the relaxation time τ_0.

VI. CHARACTERISTIC NUMERICAL VALUES

To understand better the role of different factors in the coherent relaxation of nuclear spins and the magnitudes of the related characteristic parameters, let us now make numerical estimates. We take the values of parameters that are typical of many ferromagnetic materials in which one usually studies nuclear magnetic resonance and nuclear spin echo.[21,29,30] These can be pure materials, such as Co, or various ferromagnetic alloys and compounds.[29,30] Since ferrimagnets are often treated by effective ferromagnetic models, ferrimagnetic materials, such as $MnFe_2O_3$, are also included here.[31]

For the characteristic magnetic fields and the corresponding frequencies we have the following values. The contact hyperfine field $H_A\equiv A/\mu_n\sim10^5$ G, the related frequency $\omega_A\equiv A/\hbar\sim10^9$ s^{-1}. The hyperfine field is smaller than the electron exchange field $H_J\equiv J/\mu_e\sim10^6$ G, the corresponding frequency being $\omega_J\equiv\mu_eH_J/\hbar\sim10^{13}$ s^{-1}. However, both these fields are important for nonequilibrium processes in the nuclear-spin system, although the role of these fields is different. The hyperfine field acts directly on the nuclear spins, and the exchange field influences nuclear-spin relaxation indirectly, through the formation of magnetic order in the electronic subsystem. If we take an external magnetic field $H_0\sim10^4$ G, then the Zeeman frequencies (13) are $\omega_e\equiv\mu_eH_0/\hbar\sim10^{11}$ s^{-1} and $\omega_n\equiv\mu_nH_0/\hbar\sim10^8$ s^{-1}. The magnetocrystalline anisotropy field $H_a\leqslant10^3$ G, depending on the particular structure of matter. The anisotropy parameters (14) are $\alpha_e\equiv\mu_eH_a/\hbar\leqslant10^{10}$ s^{-1} and $\alpha_n\equiv\mu_nH_a/\hbar\leqslant10^7$ s^{-1}, respectively. The longitudinal widths γ_1 and Γ_1

can vary within rather wide intervals, but usually $\gamma_1 \ll \gamma_2$ and $\Gamma_1 \ll \Gamma_2$. For the transverse widths we may take the estimates $\gamma_2 \sim \rho_e \mu_e^2/\hbar$ and $\Gamma_2 \sim \rho_n \mu_n^2/\hbar$. This, with $\mu_e \sim 10^{-20}$ erg/G, $\mu_n \sim 10^{-23}$ erg/G, and $\rho_e \sim \rho_n \sim 10^{23}$ cm^{-3}, gives $\gamma_2 \sim 10^{10}$ s^{-1} and $\Gamma_2 \sim 10^4$ s^{-1}. The value for γ_2 is to be treated as the upper limit, since the density of electrons is usually less than 10^{23} cm^{-3}, being, for instance, 10^{22} cm^{-3} for typical ordinary metals.[32] In the case when the considered electrons are related to impurity ions inside an insulator, as in Refs. 6–10 then their density can be $\rho_e \sim 10^{20}$ cm^{-3}, resulting in $\gamma_2 \sim 10^7$ s^{-1}. The estimated value of Γ_2 is in agreement with experimental measurements.[30] For the resonator ringing width, we may take a typical experimental value of $\gamma_3 \sim 10^6$ s^{-1}. Then $\gamma_3/\omega_e \sim 10^{-5}$ and $\gamma_3/\omega_n \sim 10^{-2}$, so that inequality (26) is satisfied.

Since $\gamma_1 \ll \gamma_2$ and $\gamma_2/\omega_e \leqslant 10^{-1}$, condition (31) is valid. The nuclear magnetic resonance frequency (28) is $\omega_N \sim \omega_n \sim 10^8$ s^{-1} if $z_{\rm eff}=0$, that is if the magnetic order is absent, and if $z_{\rm eff} \neq 0$, then $\omega_N \sim 10^9$ s^{-1}. Hence, $\Gamma_2/\omega_N \sim \Gamma_2/\omega_n \sim 10^{-4}$, when $z_{\rm eff}=0$, and $\Gamma_2/\omega_N \sim 10^{-5}$ for a ferromagnetic material with $z_{\rm eff} \neq 0$. This, together with $\Gamma_1 \ll \Gamma_2$, shows that condition (33) holds true.

For electrons, $\alpha_e/\omega_e \leqslant 10^{-1}$ and $\omega_A/\omega_e \sim 10^{-2}$, while for nuclei, $\alpha_n/\omega_N \leqslant 10^{-2}$ and $x_{\rm eff}=0$. The resonator feedback field (38) is $H_{\rm eff} \sim \beta_e \omega_A$. For electrons, with a resonator natural frequency close to the electron spin resonance frequency, $\omega \sim \omega_e$, we have $\mu_e H_{\rm eff}/\omega_e \sim 10^{-3}$, and for nuclei, when $\omega \sim \omega_N$, we find $\mu_n H_{\rm eff}/\omega_N \sim 10^{-2}$. Thus, all inequalities in Eqs. (30) and (32) are valid. Since $|\mu_n/\mu_e| \sim 10^{-3}$ and Γ_1/γ_1 and Γ_2/γ_2 are of the order of $\rho_n \mu_n^2/\rho_e \mu_e^2 \sim (\rho_n/\rho_e) 10^{-6}$, the inequalities in Eq. (34) hold true if ρ_n and ρ_e are not drastically different. Conditions (35) are also satisfied, since $\Gamma_{\rm en}/\omega_N \sim \mu_n \gamma_2/\mu_e \omega_N \sim 10^{-2}$ and $\Gamma_{\rm nn}/\omega_N \sim \Gamma_2/\omega_N \sim 10^{-5}$.

Among the parameters defining the characteristic times of the coherent nuclear spin relaxation, we have the anisotropy parameter $\alpha_n \leqslant 10^7$ s^{-1}, the inhomogeneous broadening due to hyperfine dipole interactions, $\Gamma_{\rm en} \sim \rho \mu_e \mu_n$ with $\rho = \min\{\rho_e, \rho_n\}$, the inhomogeneous broadening due to nuclear dipole interactions, $\Gamma_{\rm nn} \sim \rho_n \mu_n^2 \sim 10^4$ s^{-1}, and the parameter δ is given by Eq. (46), from which $\delta^2 \sim 10^{-2} \Gamma_{\rm en}^2 z_{\rm eff}^2$. The width $\Gamma_{\rm en}$, according to Eqs. (69) and (70), is always larger than $\Gamma_{\rm nn}$, provided that the density of electrons ρ_e is not three orders smaller than the density of nuclei ρ_n, which follows from the relation $\Gamma_{\rm en} \sim 10^3 (\rho_e/\rho_n) \Gamma_{\rm nn}$. For example, if $\rho_e \sim \rho_n$, then $\Gamma_{\rm en} \sim 10^3 \Gamma_{\rm nn}$. If we take $\rho_e \sim 2 \times 10^{20}$ cm^{-3} and $\rho_n \sim (5 \times 10^{22} - 10^{23})$ cm^{-3}, as in the experiments (Refs. 6–10), then $\Gamma_{\rm en} \sim (1-10) \Gamma_{\rm nn}$. In this way, the hyperfine width $\Gamma_{\rm en}$ is usually larger than $\Gamma_{\rm nn}$ and always larger than δ, although $\Gamma_{\rm en}$ may be comparable with α_n, if $\rho_e \ll \rho_n$. When $\rho_e \sim \rho_n$, the largest parameters among those considered above are α_n and $\Gamma_{\rm en} \sim 10^7$ s^{-1}. In such a case, other parameters entering additively with these can be omitted. For instance, the delay time (67) may be written as

$$t_0 = \frac{T_2}{2 g s_0} \ln \left| \frac{\alpha_n^2 + \Gamma_{\rm en}^2}{4 \omega_N^2} \right| .$$

An important parameter entering the expressions for the characteristic times and influencing the behavior of solutions is the coupling parameter g, which is drastically different for the case when there is magnetic order in the electron system compared to the case when the magnetization is absent. For the latter case, when $z_{\rm eff}=0$, we have $g \sim 10$. When a ferromagnetic material is considered, so that $z_{\rm eff} \sim 1$, then the second term in Eq. (42) can become much larger than the first one. Thus, for $\rho_e \sim \rho_n$, we have

$$\frac{\rho_e \mu_e A z_{\rm eff}}{\rho_n \mu_n \omega_N} \sim \frac{\mu_e}{\mu_n} \sim 10^3 .$$

Therefore, the coupling parameter g can be increased by three orders by the presence of electron magnetization, reaching the value $g \sim 10^4$. The system of magnetized electrons acts as an additional resonator strongly strengthening the coupling between the resonance electric circuit and nuclear spins.

To evaluate the characteristic values of the delay time (67) and relaxation time (68), we made calculations for several ferromagnetic materials with typical parameters taken from Ref. 30. In our formulas we take the filling factor $\eta = 1$, consider the purely resonance case, when $\omega = |\omega_N|$, also take $z_{\rm eff} = \frac{1}{2}$, $\rho_e = \rho_n$ and assume that the transverse anisotropy parameter is small compared to $\Gamma_{\rm en}$. We analyze the case of purely self-organized coherent relaxation when at the initial time nuclear spins are polarized against the external magnetic field, so that $s_0 = -I$, where I is an absolute value of a nuclear spin, and there is no initial coherence imposed upon the system, so that $u_0 = 0$. The high initial polarization of nuclear spins can be achieved by the dynamic nuclear polarization technique. The transverse relaxation time $T_2 = \Gamma_2^{-1}$ can be measured by several methods, e.g., the two-pulse echo technique or the single-pulse echo technique,[30] of which the former is likely to be more reliable. Our results are presented in Table I.

VII. DISCUSSION

Nonlinear spin dynamics is considered for ferromagnets consisting of electron and nuclear subsystems coupled through hyperfine forces. The sample is prepared in a strongly nonequilibrium initial state. In addition, the ferromagnetic sample is considered inserted into a coil of a resonant electric circuit. All this makes the spin dynamics highly nonlinear. The evolution of the system is described by seven nonlinear equations, six of which are differential equations for electron and nuclear spins and one equation is an integrodifferential equation for the feedback field of the resonator. These are solved by using the scale separation approach.[11,12] It is shown that due to the resonator feedback field an ultrafast coherent relaxation of spins can occur. The system of magnetized electrons serves as an additional resonator for the nuclear spins, significantly enhancing the effective coupling of the nuclear spins with the resonator circuit. Such an enhancement can reach three orders of magnitude, as compared to the coupling in a paramagnetic material.

The ultrafast coherent relaxation of nuclear spins may be either triggered by an initial pulse or can be self-organized. The latter case is the more interesting, since then all relaxation characteristics, such as the delay time and relaxation

TABLE I. The characteristic parameters related to the self-organized coherent nuclear-spin relaxation in several ferromagnetic materials.

Sample	Nucleus	I	ω_N (10^9 Hz)	T_2 (10^{-4} s)	τ_0 (10^{-8} s)	t_0 (10^{-8} s)
$Li_{0.5}Fe_{2.5}O_4$	^{57}Fe	1/2	0.47	40	88.2	400
$Mn_{0.51}Sb_{0.49}O_4$	^{123}Sb	7/2	1.31	1.70	0.54	2.98
	^{55}Mn	5/2	1.61	0.60	0.26	1.53
Ni-Mn-Sb	^{55}Mn	5/2	1.88	0.95	0.42	2.49
Ni-Mn-Si	^{55}Mn	5/2	2.01	0.60	0.26	1.59
Co_2MnSi	^{59}Co	7/2	0.91	0.38	0.12	0.62
	^{55}Mn	5/2	1.59	0.80	0.35	2.03
Co (fcc)	^{59}Co	7/2	1.37	0.30	0.09	0.53
Co (hcp)	^{59}Co	7/2	1.38	0.65	0.21	1.15

time, depend strongly on the values of the internal parameters. The most important such parameters, starting the process of self-organized coherent relaxation and, therefore, defining the main relaxation characteristics, are the transverse magnetocrystalline anisotropy and the dipole hyperfine interactions. If the density of electrons is more than three orders of magnitude lower than the density of nuclei, then the direct nuclear dipole interactions also become important. An interesting extension of the present approach could be the inclusion of external alternating magnetic fields, as has been done for nuclear magnets.[33–35]

By studying the peculiarities of the coherent spin relaxation, it is possible to extract information on the intrinsic properties of magnetic materials. This especially concerns the regime of self-organized coherent relaxation, whose characteristics are very sensitive to the values of the parameters of the material studied. By observing a coherent pulse in the power flow, one can measure, with a very high precision, the delay time t_0. The latter can be accurately measured because it exactly corresponds to the maximum of the function $w(t)$, which is proportional to the power flow in the resonant circuit.[12] If the delay time t_0 is measured experimentally, then, inverting Eq. (67), one may find the sum of α_n^2 and $\Gamma_*^2 \equiv \Gamma_{\rm en}^2 + \Gamma_{\rm nn}^2$ as

$$\alpha_n^2 + \Gamma_*^2 = 4\omega_N^2 \exp\left(2gs_0 \frac{t_0}{T_2} \right),$$

where the inequality $\delta \ll \Gamma_{\rm en}$ is taken into account. This relation, when α_n is known from other experiments, makes it possible to define the inhomogeneous broadening Γ_*. As is mentioned in Sec. VI, one usually has $\alpha_n \leqslant \Gamma_*$. The transverse-anisotropy parameter α_n depends on the orientation of the sample with respect to the external magnetic field. It is possible to choose an orientation such that $\alpha_n \ll \Gamma_*$. Then we obtain a simple formula giving the inhomogeneous broadening

$$\Gamma_* = 2\omega_N \exp\left(gs_0 \frac{t_0}{T_2} \right)$$

in terms of the known values of the nuclear magnetic resonance frequency ω_N, the coupling parameter $g = g_n$ in Eq. (42), the initial nuclear polarization s_0, the transverse relaxation time T_2, and the measured delay time t_0. It follows from the analysis of Sec. V that $t_0 > 0$ requires $s_0 < 0$. Thus the value $s_0 t_0$ in the formula for Γ_* is negative, making $\Gamma_* \ll \omega_N$. The exponential dependence of Γ_* on the delay time t_0 makes the value of Γ_* very sensitive to t_0.

Another possibility for exploiting the effect of coherent-spin relaxation is its sensitivity to initial conditions, in particular, to the initial amplitude of the transverse spin, $|u_0|$. The latter, in order to influence the delay time (58) and relaxation time (59), should be such that

$$|u_0|^2 > \frac{\alpha_n^2 + \Gamma_*^2}{\omega_N^2} s_0^2 \leqslant 10^{-4}.$$

This is always small, and can be made arbitrarily smaller by reducing s_0. Hence, we conclude that even quite weak external pulses, resulting in nonzero $|u_0|$, can trigger the process of coherent relaxation. For example, from Eqs. (59) and (45) we get

$$|u_0|^2 = \left(\frac{T_2}{g\tau_0} \right)^2 + \left(\frac{\alpha_n^2 + \Gamma_*^2}{\omega_N^2} - 1 \right) s_0^2.$$

This allows us, by measuring the coherent relaxation time τ_0, to find the initial amplitude $|u_0|$. The sensitivity of coherent-spin relaxation to initial conditions could be employed for creating ultrasensitive detectors of weak external pulses. In turn, this can be used to construct sensitive particle detectors.[19]

In conclusion, the main results obtained in this paper can be summarized in the following:

(i) A theory of nonlinear spin dynamics is developed for the systems of electron and nuclear spins coupled with each other through hyperfine forces and also coupled to a resonator electric circuit. This essentially generalizes the previous consideration of nonequilibrium nuclear magnets[11–13] to a much wider class of materials, having long-range magnetic order.

(ii) The very complicated set of nonlinear differential equations is solved by invoking the scale separation approach.[11,12] It is important that, because of the existence of small parameters resulting in different time scales, the motion of electron and nuclear spins can be effectively separated, as is seen in Eqs. (47)–(50).

(iii) The effect of a strong *coupling-parameter enhancement*, due to the presence of magnetic order in the electronic subsystem, is described. The effective nuclear coupling parameter can be enhanced by three orders of magnitude, which makes relaxation really ultrafast, with the relaxation time (63) becoming smaller than T_2 by four orders of magnitude.

(iv) The nature of all main intrinsic mechanisms triggering the self-organized coherent relaxation, that occurs in the absence of external pulses, is elucidated. These mechanisms, defining the delay time (67), are the electron-nuclear interactions through hyperfine dipole forces, nuclear dipole interactions, and magnetocrystalline anisotropy fields.

(v) Two types of applications are discussed. One type concerns the investigation of the internal properties of the materials by measuring the delay time (58) and coherent relaxation time (59). Another type utilizes the sensitivity of these characteristic times to the initial conditions, giving the possibility of employing coherent-spin relaxation for the ultrasensitive detection of weak external pulses.

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APPENDIX: FEEDBACK FIELD

According to the scale separation approach,[11,12] we first consider Eq. (19) for the fastest variable x, treating there the slow variables s and z as quasi-integrals of motion. Because of the second inequality in Eq. (32), we can, in a first approximation, omit the term containing H, where upon the solution of Eq. (19) is

$$x \simeq \left(x_0 - \frac{A_e z}{\Omega_e + i\gamma_2} \right) \exp\{ i(\Omega_e + i\gamma_2)t \} + \frac{A_e z}{\Omega_e + i\gamma_2}$$

with $\Omega_e \equiv \omega_e - As - \xi_0$ and $A_e \equiv \alpha_e - Au - \xi$. In Eq. (22) we keep, as a quasi-integral of motion, the slow variable s. Because of the second inequality in Eq. (32), we again, to the first approximation, may omit the term with H. Averaging, in the right-hand side of Eq. (22), the electron variables over the period $2\pi/\omega$, we get an approximate equation

$$\frac{du}{dt} \simeq i(\Omega_n + i\Gamma_2)u - iA_n s,$$

where $\Omega_n \equiv \omega_n - Az_{\rm eff} - \varphi_0$ and $A_n \equiv \alpha_n - Ax_{\rm eff} - \varphi$. We find

$$u \simeq \left(u_0 - \frac{A_n s}{\Omega_n + i\Gamma_2} \right) \exp\{ i(\Omega_n + i\Gamma_2)t \} + \frac{A_n s}{\Omega_n + i\Gamma_2}.$$

Substituting the approximate expressions into Eq. (25), we come to the form

$$H = -2\,{\rm Re}\left[\beta_e(t)\frac{dx}{dt} + \beta_n(t)\frac{du}{dt} \right],$$

in which

$$\beta_e(t) = \pi\eta\frac{\mu_e\rho_e}{\delta_e}[\exp(\delta_e t) - 1],$$

$$\beta_n(t) = \pi\eta\frac{\mu_n\rho_n}{\delta_n}[\exp(\delta_n t) - 1],$$

$$\delta_e = i(\omega - \omega_e + As + \xi_0) + \gamma_2 - \gamma_3,$$

$$\delta_n = i(\omega - \omega_n + Az + \varphi_0) + \Gamma_2 - \gamma_3.$$

The functions $\beta_e(t)$ and $\beta_n(t)$ do not vary much during the period $2\pi/\omega$, so we may replace them by their averages over this time,

$$\beta_e \equiv \frac{\omega}{2\pi}\int_0^{2\pi/\omega}\beta_e(t)dt = \pi^2\eta\frac{\mu_e\rho_e}{\omega}\left(1 + \frac{2\pi\delta_e}{3\omega} \right),$$

$$\beta_n \equiv \frac{\omega}{2\pi}\int_0^{2\pi/\omega}\beta_n(t)dt = \pi^2\eta\frac{\mu_n\rho_n}{\omega}\left(1 + \frac{2\pi\delta_n}{3\omega} \right).$$

Omitting here the small terms δ_e/ω and δ_n/ω, we obtain Eq. (36).

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