Nonlinear Dynamics of Nuclear–Electronic Spin Processes in Ferromagnets

V.I. Yukalov\textsuperscript{1,2}, M.G. Cottam\textsuperscript{1}, and M.R. Singh\textsuperscript{1}

\textsuperscript{1}Center for Chemical Physics
University of Western Ontario, London, Ontario N6A 3K7, Canada

\textsuperscript{2}Bogolubov Laboratory of Theoretical Physics
Joint Institute for Nuclear Research, Dubna 141980, Russia

Spin dynamics is considered in ferromagnets consisting of electron and nuclear subsystems interacting with each other through hyperfine forces. In addition, the ferromagnetic sample is coupled with a resonance electric circuit. Under these conditions, spin relaxation from a strongly nonequilibrium initial state displays several peculiarities absent for the standard set-up in studying spin relaxation. The main feature of the nonlinear spin dynamics considered in this communication is the appearance of ultrafast coherent relaxation, with characteristic relaxation times several orders shorter than the transverse relaxation time $T_2$. This type of coherent spin relaxation can be used for extracting additional information on the intrinsic properties of ferromagnetic materials and also can be employed for different technical applications.
I. INTRODUCTION

Spin systems can exhibit rather nontrivial dynamics when the magnetic sample is prepared in a strongly nonequilibrium state and, in addition, is coupled with a resonance electric circuit [1]. Due to the resonator feedback field, a coherent motion of spins can develop resulting in their ultrafast relaxation. However, the feedback field can organize coherent relaxation only when some initial mechanism triggers the process. A simple case could be the application of an external pulse at the initial time. If this pulse is sufficiently strong, the spin dynamics could be described by the Bloch equations. A more difficult, but interesting, situation is when no external pulse starts the process, but the latter develops in a self-organized way due to local spin fluctuations caused by their interactions. In such a case, the Bloch equations are not appropriate [1] and one has to resort to microscopic models.

A theory of coherent spin relaxation in a system of nuclear spins inside a paramagnetic matrix has been developed [2,3], being based on a microscopic Hamiltonian with dipole interactions between nuclei. In the present paper we generalize this theory to include ferromagnetic materials. As far as ferrimagnets are often described as ferromagnets [2,3], being based on a microscopic Hamiltonian with dipole interactions between nuclei. In the present paper we insert into a coil connected with a resonance electric circuit. The general Hamiltonian describing a wide class of magnetic materials can be taken in the form

\[ \hat{H} = \hat{H}_c + \hat{H}_n + \hat{H}_{\text{int}} , \]

in which

\[ \hat{H}_c = -\frac{1}{2} \sum_{ij \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \mu_e \sum_i \mathbf{B} \cdot \mathbf{S}_i \]

is the Hamiltonian of electron spins,

\[ \hat{H}_n = \frac{1}{2} \sum_{ij \neq j} C_{ij}^{\alpha \beta} I_i^\alpha I_j^\beta - \mu_n \sum_i \mathbf{B} \cdot \mathbf{I}_i \]

is the nuclear spin Hamiltonian, and

\[ \hat{H}_{\text{int}} = A \sum_i \mathbf{S}_i \cdot \mathbf{I}_i + \frac{1}{2} \sum_{ij \neq j} A_{ij}^{\alpha \beta} S_i^\alpha I_j^\beta \]

is the term corresponding to hyperfine interactions. Here \( \mu_e = g_e \mu_B \), with \( g_e \) being the electron gyromagnetic ratio and \( \mu_B \), the Bohr magneton; the nuclear dipole interactions \( C_{ij}^{\alpha \beta} = \mu_n^2 \left( \delta_{\alpha \beta} - 3 n_i^{\alpha} n_j^{\beta} \right) / r_{ij}^3 \) contain \( \mu_n = g_n \mu_N \), with \( g_n \) being the nuclear gyromagnetic ratio and \( \mu_N \), nuclear magneton, and \( r_{ij} = |\mathbf{r}_{ij}| \), \( n_i = \mathbf{r}_{ij} / r_{ij} \), \( n_j = \mathbf{r}_{ij} / r_{ij} \); the hyperfine interactions consist of a contact part with a constant \( A \) and of a dipole-dipole part with \( A_{ij}^{\alpha \beta} = \mu_e \mu_n \left( \delta_{\alpha \beta} - 3 n_i^{\alpha} n_j^{\beta} \right) / r_{ij}^3 \); the indices \( i \) and \( j \) enumerate electrons or nuclei according to the context, and \( \alpha, \beta = x, y, z \); \( \mathbf{S}_i \) is an electron spin operator while \( \mathbf{I}_i \) is a nuclear spin operator. The total magnetic field \( \mathbf{B} \) is the vector sum of an external magnetic field in the \( z \) direction and of a transverse field \( H_1 = H_n + H \) in the \( x \) direction, consisting of an effective field \( H_n \) of a transverse magnetocrystalline anisotropy and of a resonator feedback field \( H \). The latter satisfies the Kirchhoff equation

\[ \frac{dH}{dt} + 2 \gamma_3 H + \omega^2 \int_0^t H(\tau) d\tau = -4 \pi \eta \frac{dM_r}{dt} , \]

in which \( \eta \) is a filling factor; \( \omega \), resonator natural frequency; \( \gamma_3 \equiv \omega / 2Q \) is the resonator ringing width; \( Q \) is a quality factor; and \( M_r = \frac{1}{2} \sum_i (\mu_e < S_i^x > + \mu_n < I_i^x >) \) is the transverse magnetization, where the angle brackets mean the statistical averaging.
Employing the Heisenberg equations of motion, we derive the time evolution equations for the following averages, related to the electron and nuclear spins,

\[ x \equiv \frac{1}{N_e} \sum_i < S_i^- > , \quad z \equiv \frac{1}{N_e} \sum_i < S_i^z > , \]
\[ u \equiv \frac{1}{N_n} \sum_i < I_i^- > , \quad s \equiv \frac{1}{N_n} \sum_i < I_i^z > , \]

(6)

where \( N_e \) and \( N_n \) are the number of electrons and nuclei, respectively, and \( S_i^- \) and \( I_i^- \) are the ladder operators. As the transverse variables \( x \) and \( u \) are complex, we need also the equations of motion for either \( x^2 \) and \( |x|^2 \) or \( |u|^2 \) and \( |u|^2 \). In this way, we obtain seven evolution equations, three for the electron variables \( x, z, \) and \( |u|^2 \), three for the nuclear variables \( u, s, \) and \( |u|^2 \), and the feedback–field equation (5). Although this is a rather complicated system of nonlinear equations, it can be treated by using the scale separation approach [2,3]. Details of this approach have been thoroughly described in Refs. [3–5].

Note first that if one invokes the standard semiclassical decoupling of spin correlators, assuming the translational invariance of the average spins, then some of the terms in the evolution equations become zero because of the properties of the dipolar interactions. The translation invariance of averages is equivalent to neglecting inhomogeneous local spin fluctuations. However, the latter are crucially important for the correct description of spin dynamics [1–3]. The inhomogeneous spin fluctuations can be retained by treating them as random local fields. Thus we come to the stochastic semiclassical approximation [3–5]. Then, using the method of the Laplace transforms, we may express the feedback field from Eq. (5) through the derivatives of spin variables and employ this relation in the evolution equations for fast variables while treating slow variables as quasi–integrals of motion. We keep in mind the following usual inequalities:

\[ |\gamma_1/\omega_E| \ll 1 , \quad |\gamma_2/\omega_E| \ll 1 , \quad |\Gamma_1/\omega_N| \ll 1 , \quad |\Gamma_2/\omega_N| \ll 1 , \]

(7)

in which \( \gamma_1 \) and \( \Gamma_1 \) are the transverse attenuations for the electron and nuclear spins, \( \gamma_2 \) and \( \Gamma_2 \) are the longitudinal attenuations for electron and nuclear spins, respectively, and

\[ \omega_E \equiv (\mu_e H_0 - As)/\hbar , \quad \omega_N \equiv (\mu_n H_0 - Am)/\hbar \]

(8)

are the electron spin resonance frequency and the nuclear magnetic resonance frequency, respectively; \( m \) being an average magnetization in the electron system. Another reasonable assumption is that the external magnetic field \( H_0 \) is stronger than the magnetocrystalline anisotropy field \( H_a \) and that, similarly to (7), the inhomogeneous broadening, caused by local spin fluctuations, is smaller than the corresponding frequencies, so that

\[ |\alpha_e/\omega_E| \ll 1 , \quad |\alpha_n/\omega_N| \ll 1 , \quad |\gamma_e/\omega_E| \ll 1 , \quad |\Gamma_e/\omega_N| \ll 1 , \quad |\gamma_e/\omega_E| \ll 1 , \]

(9)

where \( \alpha_e \equiv \mu_e H_a/\hbar \) and \( \alpha_n \equiv \mu_n H_a/\hbar \) are the anisotropy parameters and \( \gamma_e \) and \( \Gamma_e \) are the inhomogeneous widths for electrons and nuclei, respectively. Also, because the nuclear magneton is three orders smaller than the Bohr magneton, we have

\[ |\mu_n/\mu_e| \ll 1 , \quad \Gamma_1/\gamma_1 \ll 1 , \quad \Gamma_2/\gamma_2 \ll 1 . \]

(10)

Finally, we consider the case of a high quality resonator, having a large quality factor, and we assume that the resonator natural frequency is tuned close to the frequency of nuclear magnetic resonance, so that \( |\Delta_N/\omega_N| \ll 1 \) and \( \gamma_3/\omega \ll 1 \), where \( \Delta_N = \omega - \omega_N \).

With these inequalities, we can classify all variables into fast and slow with respect to each other. Following the general scheme [3–5], we solve the equations for fast variables while treating slow variables as quasi–integrals of motion. Then, the found solutions for fast variables are substituted into the equations for slow variables and the right–hand sides of the latter equations are averaged over the periods of fast oscillations and over the random local fields. Introducing also the change of variables

\[ w = |u|^2 - \frac{\alpha_n^2 + \Gamma_2^2 + \delta^2}{\omega_N^2} s^2 , \quad \delta \equiv \frac{\sqrt{2}\pi^2 \eta_\gamma \rho_\gamma \mu_\gamma \mu_n m}{\omega_N} \]

(11)

we come to the equations describing the slow nuclear spin variables

\[ \frac{ds}{dt} = \Gamma_2 gw - \Gamma_1 (s - \zeta) , \quad \frac{dw}{dt} = -2\Gamma_2 (1 + gs) w , \]

(12)
where

\[ g \equiv \pi^2 \eta \rho_n \mu_n^2 \omega_N \left( 1 + \frac{\rho_c \mu_c A_m}{\rho_n \mu_n \omega_N} \right) \]  

(13)

is the parameter of effective coupling of nuclear spins with the resonator.

For the relaxation times \( T_1 \equiv \Gamma_1^{-1} \) and \( T_2 \equiv \Gamma_2^{-1} \) one usually has the relation \( T_2 \ll T_1 \). Therefore, for the times \( t \ll T_1 \), equations (12) can be solved analytically giving

\[ s = \frac{T_2}{g \tau_0} \tanh \left( \frac{t - t_0}{\tau_0} - \frac{1}{g} \right), \quad w = \left( \frac{T_2}{g \tau_0} \right)^2 \text{sech}^2 \left( \frac{t - t_0}{\tau_0} \right), \]  

(14)

where \( \tau_0 \) is the collective relaxation time and \( t_0 \) is the delay time, respectively,

\[ \tau_0 = \frac{T_2}{\sqrt{(1 + gs_0)^2 + g^2w_0^2}}, \quad t_0 = \frac{\tau_0}{2} \ln \left| \frac{T_2 - \tau_0 (1 + gs_0)}{T_2 + \tau_0 (1 + gs_0)} \right|, \]  

(15)

with \( s_0 = s(0) \) and \( w_0 = w(0) \) defined by initial conditions.

III. CONCLUSION

We analysed the obtained solutions for the parameters typical of such ferromagnetic materials as EuO, EuS, EuSe, Li$_2$Fe$_3$O$_4$, Mn$_2$Sb$_{1-x}$, NiMnSb, NiMnSi, Co$_2$MnSi, and Co in the fcc and hcp phases [6,7]. Since ferrimagnets can often be modeled as ferromagnets with an effective magnetization [8], our analysis is applicable as well to ferrimagnetic materials, such as MnFe$_2$O$_4$. For these materials, taking as initial conditions \( s_0 = -I \), \( w_0 = 0 \), where \( I \) is a nuclear spin, we obtain the relaxation time \( \tau_0 \equiv T_2/gI \), where \( g \equiv \pi^2 \mu_e / 2 \mu_n \), and the delay time \( t_0 = \tau_0 \ln(2\omega_N/10^7) \). This gives the coupling parameter \( g \sim 10^4 \), which, with \( T_2 \sim 10^{-7} \) s, yields the relaxation time \( \tau_0 \sim 10^{-8} \) s. Because \( \omega_N \sim 10^9 \text{ s}^{-1} \), we have the delay time \( t_0 \sim 5 \times 10^{-8} \) s.

The described regime corresponds to the ultrafast coherent spin relaxation, when during the time \( t_0 + \tau_0 \) the initial strongly nonequilibrium spin polarization \( s_0 = -I \) changes to its equilibrium value \( s(t) \cong I \) at \( t \gg t_0 \). The time \( t_0 + \tau_0 \) can be four orders less than the standard spin–spin relaxation time \( T_2 \). This ultrafast coherent spin relaxation can serve as an additional technique for studying the spin-spin correlations in ferromagnetic materials, complementing other known techniques, such as neutron diffraction, light scattering, and nuclear magnetic resonance. The ultrafast relaxation mechanism can also find application in the important problem of fast repolarization of solid–state targets used in scattering experiments [9,10], as well as for fast switching devices in electronics and computing.

ACKNOWLEDGEMENT

Financial support from the University of Western Ontario and NSERC of Canada is appreciated.

[1] N. Bloembergen and R.V. Pound, Phys. Rev. 95, 8 (1954).
[2] V.I. Yukalov, Phys. Rev. Lett. 75, 3000 (1995).
[3] V.I. Yukalov, Phys. Rev. B 53, 9232 (1996).
[4] V.I. Yukalov, Laser Phys. 3, 870 (1993).
[5] V.I. Yukalov, Laser Phys. 5, 970 (1995).
[6] S.H. Charap and E.L. Boyd, Phys. Rev. A 133, 811 (1964).
[7] I.G. Kiliptari and V.I. Tsifrinovich, Phys. Rev. B 57, 11554 (1998).
[8] A.J. Heeger and T.W. Houston, Phys. Rev. A 135, 661 (1964).
[9] L. Reichertz et. al., Nucl. Instrum. Methods Phys. Res. A 340, 278 (1994).
[10] V.I. Yukalov, Nucl. Instrum. Methods Phys. Res. A 370, 345 (1996).