Relaxation Regimes of Spin Maser

V.I. Yukalov

International Centre of Condensed Matter Physics
University of Brasilia, CP 04513
Brasilia, DF 70919–970, Brazil
and
Bogolubov Laboratory of Theoretical Physics
Joint Institute for Nuclear Research
Dubna 141980, Russia

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Abstract

Spin relaxation in a microscopic model of spin maser is studied theoretically. Seven qualitatively different regimes are found: free induction, collective induction, free relaxation, collective relaxation, weak superradiance, pure superradiance, and triggered superradiance. The initiation of relaxation can be originated either by an imposed initial coherence or by local spin fluctuations due to non-secular dipole interactions. The Nyquist noise of resonator does not influence processes in macroscopic samples. The relaxation regimes not initiated by an imposed coherence cannot be described by the standard Bloch equations.
1 Introduction

Several experiments have been recently accomplished studying the peculiarities of spin relaxation in nonequilibrium systems of nuclear spins coupled with a resonator. Such systems include \( \text{Al} \) nuclear spins in ruby \((\text{Al}_2\text{O}_3)\) [1], and proton spins in propanediol \((\text{C}_3\text{H}_8\text{O}_2)\) [2–5], butanol \((\text{C}_4\text{H}_9\text{OH})\), and ammonia \((\text{NH}_3)\) [6]. Radiation processes in these systems occur at radiofrequencies, that is, in the maser frequency region. Similar processes, also occurring at radiofrequencies, exist in ensembles of electron spins (see discussion and references in [7,8]). The main attention in studying these processes has been payed to the possibility of coherent effects in spin relaxation, the effects that resemble the well known optical superradiance [9–11].

For describing spin relaxation, one usually invokes the Bloch equations supplemented by the Kirchhoff equation for resonator. In addition, to solve this complicated system of equations, one resorts to adiabatic approximation, when the resonator feedback field is proportional to transverse magnetization. The latter approximation is, of course, unphysical for transient phenomena and could be used only at the final stage of relaxation. Moreover, the Bloch equations as such, even being solved in a more realistic approximation [7], have the following generic limitations: (1) The role of dipole spin interactions is reduced to the inclusion of the spin–spin relaxation time \( T_2 \); (2) Coherent effects can be induced either by external magnetic field acting directly on spins or by imposing coherent initial conditions for transverse magnetization; (3) No self–organized coherence, that is called pure superradiance, can appear from a noncoherent state. These deficiencies do not permit to reach good agreement with experiment.

To give a realistic picture of spin dynamics in nuclear magnets, one has to deal with a microscopic model. This kind of model has been treated by means of computer simulations [12–14]. However, great number of parameters in the model makes it practically impossible to analyse different relaxation regimes by varying these parameters turn by turn. Here we present an analytical solution of the problem obtained with the help of a method developed in Refs. [15–17].

2 Relaxation Regimes

The microscopic model for a system of nuclear spins is given [18] by the Hamiltonian

\[
\hat{H} = \frac{1}{2} \sum_{i \neq j}^N H_{ij} - \mu \sum_{i=1}^N \vec{B} \vec{S}_i
\]

(1)
with dipole spin interactions

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

where \( \mu \) is a nuclear magneton and

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

The total magnetic field

\[ \vec{B} = \vec{H}_0 + \vec{H} \quad (3) \]

consists of a constant external field \( \vec{H}_0 \) and an alternating field \( H \) of a resonator coil,

\[ \vec{H}_0 = H \hat{e}_z, \quad \vec{H} = H \hat{e}_x. \quad (4) \]

The equations of motion, to be considered in what follows, will involve the average transverse spin component

\[ u \equiv \frac{1}{N} \sum_{i=1}^{N} \langle S_i^x - iS_i^y \rangle \quad (5) \]

and the average longitudinal component

\[ s \equiv \frac{1}{N} \sum_{i=1}^{N} \langle S_i^z \rangle, \quad (6) \]

in which \( \langle \ldots \rangle \) implies statistical averaging. Since (5) is complex, we will need one more equation either for \( u^* \) or for

\[ v \equiv |u|. \quad (7) \]

For the resonator magnetic field we introduce the dimensionless notation

\[ h \equiv \frac{\mu H}{\hbar \gamma_3} \left( \gamma_3 \equiv \frac{\omega}{2Q} \right), \quad (8) \]

in which \( \gamma_3 \) is the ringing width; \( \omega \), the resonator natural frequency; and \( Q \) is the resonator quality factor. The resonator is a coil of \( n \) turns of a cross-section area \( A_0 \). For the electromotive force \( E_f \) it is convenient to pass to the dimensionless quantity

\[ f \equiv \frac{c\mu E_f}{nA_0 \hbar \gamma_3^2} = f_0 \cos \omega t. \quad (9) \]
For the functional variables (5)–(8), we can derive [16,17] the system of equations

\[
\frac{du}{dt} = i(\omega_0 - \xi_0 + i\gamma_2)u - i(\gamma_3 h + \xi)s, \quad (10)
\]

\[
\frac{ds}{dt} = \frac{i}{2} (\gamma_3 h + \xi)(u^* - \frac{i}{2}(\gamma_3 h + \xi^*) u - \gamma_1(s - s_\infty)), \quad (11)
\]

\[
\frac{dv}{dt} = -2\gamma_2 v^2 - i(\gamma_3 h + \xi) su^* + i(\gamma_3 h + \xi^*) su, \quad (12)
\]

\[
\frac{dh}{dt} + 2\gamma_3 h + \omega^2 \int_0^t h(\tau)d\tau = -2\alpha_0 \frac{d}{dt}(u^* + u) + \gamma_3 f, \quad (13)
\]

in which \(\gamma_1\) is a spin–lattice width; \(s_\infty\), a stationary magnetization per spin, and

\[
\omega_0 \equiv \frac{\mu H_0}{\hbar}, \quad \gamma_2 \equiv \frac{\rho \mu^2}{\hbar}, \quad \alpha_0 \equiv \pi \eta \frac{\rho \mu^2}{\hbar \gamma_3} \equiv \pi \eta \frac{\gamma_2}{\gamma_3},
\]

where \(\rho\) is the density of spins and \(\eta\), a filling factor. The set \(\bar{\xi} \equiv \{\xi_0, \xi, \xi^*\}\)

of stochastic variables models local spin fluctuations, \(\xi_0\) being responsible for secular dipole interactions while \(\xi\) and \(\xi^*\) for nonsecular dipole interactions. The distribution \(p(\bar{\xi})\) of these stochastic variables is assumed to be Gaussian with a width \(\gamma_2^*\) corresponding to a nonhomogeneous width.

Equations (10)–(13) are completed by the initial conditions

\[
u(0) = u_0, \quad s(0) = z_0, \quad v(0) = v_0, \quad h(0) = 0.\quad (14)
\]

To solve the system of nonlinear integro–differential equations (10)-(13), a method has been developed [15–17] combining the ideas of the multifrequency averaging method, guiding center approach, and of generalized asymptotic expansion. Here we shall delineate the main steps of the method only in brief.

### 2.1 Classification of functional variables

Take into account that there are the following small parameters

\[
\frac{\gamma_1}{\gamma_2} \ll 1, \quad \frac{\gamma_2}{\omega_0} \ll 1, \quad \frac{\gamma_2^*}{\omega_0} \ll 1, \quad \frac{\gamma_3}{\omega} \ll 1. \quad (15)
\]

Also, the quasi–resonance case will be considered, when the detuning is small:

\[
\frac{|\Delta|}{\omega_0} \ll 1 \quad (\Delta \equiv \omega - \omega_0). \quad (16)
\]
As compared to the characteristic times

\[ T_1 \equiv \frac{1}{\gamma_1}, \quad T_2 \equiv \frac{1}{\gamma_2}, \quad T_2^* \equiv \frac{1}{\gamma_2^*}, \quad T_3 \equiv \frac{1}{\gamma_3}, \]

the spin oscillation time

\[ T_0 \equiv \frac{2\pi}{\omega_0} \ll \min\{T_1, T_2, T_2^*, T_3\} \quad (17) \]

is small.

If all small parameters in (10)-(13) tend to zero, then

\[ \frac{du}{dt} \nrightarrow 0, \quad \frac{ds}{dt} \rightarrow 0, \]

\[ \frac{dv^2}{dt} \rightarrow 0, \quad \frac{dh}{dt} \nrightarrow 0. \]

This shows that the variables \( u \) and \( h \) can be classified as fast and \( s \) and \( v^2 \), as slow.

### 2.2 Solution for fast variables

The slow variables have the meaning of quasi–integrals of motion. Solve the equations (10) and (13) for the fast variables under fixed slow variables

\[ s = z, \quad |u|^2 = v^2, \]

treated as parameters in the corresponding solution

\[ u = u(z, v, t), \quad h = h(z, v, t). \]

### 2.3 Temporal and stochastic averaging

The right–hand sides of the equations (11) and (12) are to be averaged over the small oscillation time (17) and over stochastic local fields \( \xi \). This is to be done after substituting into these right–hand sides the solutions for the fast variables found at the previous step. The resulting equations

\[ \frac{dz}{dt} = \int \left( \frac{1}{T_0} \int_0^{T_0} \frac{ds}{dt} dt \right) p(\xi) d\xi, \]

\[ \frac{dv^2}{dt} = \int \left( \frac{1}{T_0} \int_0^{T_0} \frac{d|u|^2}{dt} dt \right) p(\xi) d\xi \]

yield the solution

\[ z = z(t), \quad v = v(t). \]
2.4 Definition of guiding centers

The solutions $z(t)$ and $v(t)$ play the role of guiding centers for the slow variables. This role for the fast variables is played by

$$
\bar{u} = u(z(t), v(t), t),
\bar{h} = h(z(t), v(t), t).
$$

2.5 Generalized asymptotic expansion

Corrections to the guiding–center solutions can be obtained by expanding the functions

$$
\begin{align*}
  u &= \bar{u} + \ldots, \\
  h &= \bar{h} + \ldots, \\
  s &= z + \ldots, \\
  |u|^2 &= v^2 + \ldots
\end{align*}
$$

about the guiding centers and substituting these generalized asymptotic expansions into the equations (10)-(13). In the latter, one has to separate out corrections of different orders which leads to a system of linear equations.

This program has been accurately realized [16,17] yielding the following results. Considering the role of the Nyquist noise of resonator [19], it was found [20] that the relaxation parameter caused by the Nyquist noise is inversely proportional to the number of spins in the sample, $\gamma_N \sim N^{-1}$. The value of this parameter is negligibly small for macroscopic samples with $N \sim 10^{23}$.

The solutions for the slow variables read

$$
|u(t)|^2 = \left( \frac{\gamma_0}{g\gamma_2} \right) \text{sech}^2 \left( \frac{t - t_0}{\tau_0} \right) + 2 \left( \frac{\gamma_2^2}{\omega_0} \right) s(t),
$$

$$
\begin{align*}
  s(t) &= \frac{\gamma_0}{g\gamma_2} \tanh \left( \frac{t - t_0}{\tau_0} \right) - \frac{1}{g}.
\end{align*}
$$

Here

$$
\begin{align*}
  g &\equiv \alpha_0 \left( \frac{\gamma_3}{\gamma_2} \right) \frac{\pi (\gamma_2 - \gamma_3)^2}{(\gamma_2 - \gamma_3)^2 + \Delta^2}.
\end{align*}
$$

is the effective parameter of coupling between the spin system and the resonator. Approximately,

$$
\begin{align*}
  g &\approx \frac{\pi^2 \eta (\gamma_2 - \gamma_3)^2}{(\gamma_2 - \gamma_3)^2 + \Delta^2}.
\end{align*}
$$

Note that $g = 0$ if $\gamma_2 = \gamma_3$ and $\Delta \neq 0$. The effective radiation width $\gamma_0$, related to the radiation time $\tau_0$ as $\gamma_0 \tau_0 = 1$, is given by the equation

$$
\begin{align*}
  \gamma_0^2 &= \Gamma_0^2 + (g\gamma_2)^2 \left[ \nu_0^2 - 2 \left( \frac{\gamma_2^2}{\omega_0} \right) z_0 \right],
\end{align*}
$$

7
in which \( v_0 = |u_0|, \ z_0 = s(0) \), and
\[
\Gamma_0 = \gamma_2(1 + g z_0).
\]
The delay time is
\[
t_0 = \frac{\tau_0}{2} \ln \left| \frac{\gamma_0 - \Gamma_0}{\gamma_0 + \Gamma_0} \right|.
\] (22)

Qualitatively different regimes of relaxation can be classified according to the values of two parameters, \( g z_0 \) ans \( g v_0 \).

1. **Free induction:**
\[
g|z_0| < 1, \quad 0 < g v_0 < 1, \\
t_0 < 0, \quad \tau_0 \approx T_2.
\] (23)
This is the usual case of free nuclear induction, with the maximal coherence imposed at \( t = 0 \).

2. **Collective induction:**
\[
g z_0 > -1, \quad g v_0 > 1, \\
t_0 < 0, \quad \tau_0 < T_2.
\] (24)
The coupling with the resonator is sufficiently strong to develop collective effects shortening the relaxation time \( \tau_0 \).

3. **Free relaxation:**
\[
g|z_0| < 1, \quad v_0 = 0, \\
t_0 < 0, \quad \tau_0 \approx T_2.
\] (25)
This corresponds to an incoherent relaxation due to the local fields.

4. **Collective relaxation:**
\[
g z_0 > 1, \quad v_0 = 0, \\
t_0 < 0, \quad \tau_0 < T_2.
\] (26)
Collective effects shorten the relaxation time, as compared to the previous case.

5. **Weak superradiance:**
\[
-2 < g z_0 < -1, \quad v_0 = 0,
\]
Here the delay time is positive; the process is weakly coherent.

6. Pure superradiance:

\begin{equation}
\begin{aligned}
gz_0 < -2, & \quad v_0 = 0. \\
t_0 > 0, & \quad \tau_0 < T_2.
\end{aligned}
\end{equation}

The coherence arises as a purely self–organized process started by local random fields and developed owing to the resonator feedback field.

7. Triggered superradiance:

\begin{equation}
\begin{aligned}
gz_0 < -1, & \quad gv_0 > 1, \\
t_0 > 0, & \quad \tau_0 < T_2.
\end{aligned}
\end{equation}

This is a collective but not purely self–organized process, since the relaxation is triggered by an imposed initial coherence.

The term superradiance is used by analogy with the optical superradiance [9–11], although, as is discussed in Refs. [17,20], there are many principal differences between the atomic and spin superradiance.

The spin polarization, starting at \( s_0 = s(0) \), tends to

\begin{equation}
s(t) \simeq \frac{\gamma_0}{g} (T_2 - \tau_0), \quad t \gg t_0,
\end{equation}

which shows that polarization reversal from negative \( z_0 \) to a positive \( s(t) \) occurs when \( \tau_0 < T_2 \).

To illustrate the numerical values of the parameters occurring in the report, let us write down some characteristic quantities typical of experiments with proton spin systems [2–6], such as propanediol ( \( C_3H_8O_2 \) ), butanol ( \( C_4H_9OH \) ), and ammonia ( \( NH_3 \) ). The spin of a proton is \( S = \frac{1}{2} \). The density of spins is \( \rho \sim 10^{22} cm^{-3} \). The experiments are usually accomplished at low temperature \( T \sim 0.1K \) in a magnetic field \( H_0 \sim 10^4 G \). The Zeeman frequency is \( \omega_0 \sim 10^8 Hz \), and the corresponding wavelength is \( \lambda \sim 10^2 cm \). The characteristic times are

\begin{equation}
\begin{aligned}
T_1 \sim 10^5 sec, & \quad T_2 \sim 10^{-5} sec, & \quad T_3 \sim 10^{-6} sec - 10^{-5} sec.
\end{aligned}
\end{equation}

The parameters in (15) are really small:

\begin{equation}
\begin{aligned}
\frac{\gamma_1}{\gamma_2} \sim 10^{-10}, & \quad \frac{\gamma_2}{\omega_0} \sim 10^{-3}, & \quad \frac{\gamma_2}{\omega_0} \sim 10^{-3}, & \quad \frac{\gamma_3}{\omega} \sim 10^{-2}.
\end{aligned}
\end{equation}
The relaxation parameter $\gamma_N$ due to the resonator Nyquist noise, as compared to the spin–spin relaxation parameter $\gamma_2$, is

$$\frac{\gamma_N}{\gamma_2} \sim 10^{-21}.$$ 

The picture obtained analytically is in agreement with the numerical simulations [12–14]. Some results of the latter are shown in Figs.1-4, where $K_{coh} \equiv P_{coh}/P_{inc}$ is a coherence coefficient being the ratio of the coherent part, $P_{coh}$, of the current power, $P$, to its incoherent part, $P_{inc}$, and $P_z \equiv -z(t)$. These figures illustrate the qualitative changes of the processes under the variation of some parameters. For qualitative understanding, the absolute values of units in figures are not important, so they are not specified (for details see [12–14]).

3 Conclusion

The main results of the analytical solution for the problem of spin relaxation in nuclear magnets can be formulated as follows.

1. Taking account of direct spin–dipole interactions is important for the correct description of relaxation processes.

2. Nonsecular dipole interactions play the major role in starting spin relaxation, when no initial coherence is thrust upon the system.

3. The Nyquist noise of a resonator does not influence the relaxation processes in macroscopic samples.

4. There are seven qualitatively different regimes of relaxation, three of which are triggered by primary coherence imposed upon the system and four others are initiated by local dipole fields.

5. The regimes that are purely self-organized, when no initial coherence is imposed, cannot be described by the standard Bloch equations. These regimes are: free relaxation, collective relaxation, weak superradiance, and pure superradiance.
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Figure Captions

Fig. 1

Coherence coefficient $K_{coh}$, current power $P$, and the $z$-projection of spin polarization $P_z$ as functions of time, in arbitrary units, for two different coupling parameters, $g_1$ and $g_2$, with the relation $g_1/g_2 = 10$. The solid line is for $g_1$; the dashed, for $g_2$.

Fig. 2

The same as in Fig. 1, for two different Zeeman frequencies, $\omega_{01}$ and $\omega_{02}$, related by the ratio $\omega_{01}/\omega_{02} = 5$. The solid line is for $\omega_{01}$; the dashed, for $\omega_{02}$.

Fig. 3

The same functions as in Fig. 1, for different initial spin polarizations, with the relation $z_{01}/z_{02} = 2$. The solid line is for $z_{01}$; the dashed, for $z_{02}$.

Fig. 4

The same functions as in Fig. 1, for different initial transverse magnetizations, with the relation $v_{01}/v_{02} = 0.5$, when $z_0 = 0$. The solid line is for $v_{01}$; the dashed, for $v_{02}$.