Nuclear Spin Superradiance

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1 Introduction

Nuclear spins may exhibit several coherent phenomena, occurring at nuclear magnetic resonance frequencies, which have their direct counterparts in resonant atomic systems. For instance, nuclear free induction is an analog of atomic free induction and the spin echo is an analog of the photon echo. These analogies are due to the fact that an ensemble of identical spins forms a collective of finite-level objects, similar to a resonant system of atoms or molecules. Such a finite-level nonequilibrium system can, under special conditions, behave as a collection of coherent radiators.

Similarly to the case of resonant atoms, one may distinguish two principally different ways of dealing with spin assemblies. One situation would be when spins are near their equilibrium state, with a nonequilibrium perturbation produced by a resonant alternating field. This is a typical situation of nuclear magnetic resonance. Another possibility could be if spins are initially prepared in a strongly nonequilibrium state, e.g. being polarized against a constant external magnetic field. In the latter case, the polarized spins resemble a system of inverted resonant atoms.

One of the most interesting effects exhibited by a system of inverted atoms is superradiance, when the radiation intensity is approximately proportional to the number of atoms squared, while the duration of a superradiant pulse is inversely proportional to this number. The possibility of atomic superradiance was predicted by Dicke, and nowadays it is well studied both theoretically and experimentally. There exists a vast literature on the subject, whose description can be found in the recent books. A natural question that arises is: If atomic and spin systems are so similar, then could a kind of spin superradiance be realized?

For spins to behave coherently, there must exist a cause correlating their motion. In the case of atoms, such a correlating mechanism is caused by the photon exchange through the common radiation field. This exchange results in the formation of effective interatomic correlations collectivizing atomic radiation. For spins, however, their magneto-dipole radiation is too weak to develop noticeable spin correlations. This concerns both nuclear as well as electron spins. Such photon-exchange interactions are negligible as compared to disordering dipolar spin interactions. How then might the spin motion be collectivized?

It is, of course, possible to force an ensemble of spins to develop coherence by imposing an initial condition of longitudinal magnetization, and using an rf pulse to produce a macroscopic transverse magnetization. But this would result in the standard free nuclear induction, with the coherence being lost during the dephasing time $T_2$. However, free nuclear induction, although being a coherent process, is not superradiance with one of the main characteristic features of a short radiation time of a superradiant pulse $\tau_p \ll T_2$, shorter than the transverse dephasing time. To make the superradiant pulse time $\tau_p$ so short, some internal nonlinear correlating mechanism has to be involved.

Mutual spin correlations can arise owing to a feedback field formed by a resonant electric circuit coupled to the spin system and tuned to the Zeeman transition frequency of spins. The role of such a coupling in magnetic resonance experiments has been analysed by Bloembergen and Pound. They showed that, in the presence
of an electric resonant circuit, coupled to a spin system, the signal of nuclear induction can be damped in a time much smaller than $T_2$. Since this shortening of the induction damping time is due to collective effects, caused by the spin coupling through the resonator feedback field, the resulting process may be termed collective induction. This effect has been observed in a number of substances, as has been reviewed\textsuperscript{7}. The process already possesses one of the prerequisites of superradiance, i.e. a short radiation time $\tau_p < T_2$. Usually, the induction signal starts at $t = 0$, having there its maximum intensity, while a superradiant pulse is always separated from the time origin by a delay time $t_0 > 0$. The signal of collective induction can also be peaked at a delay time well separated from $t = 0$. However, there is a principal difference between collective induction and superradiance: In collective induction, coherence is induced by external sources, while in superradiance coherence develops in a self-organized way owing to internal correlations. The self-organized, spontaneous, nature of superradiance is one of its most important features\textsuperscript{3–5}. In general, one includes into the class of superradiant the processes that are triggered by initial external pulses, but with a compulsory requirement that the imposed initial coherence be very weak, playing just the role of a trigger. Thus, collective induction, though being a collective coherent process, is not yet superradiance.

The principal characteristics of superradiance, which apply both to atomic as well as to spin systems, can be summarized as follows:

1. Superradiance is collective, coherent radiation by an ensemble of radiators.

2. It is a spontaneous process developing in a self-organized way.

3. The maximal intensity of a superradiant burst is proportional to the number of radiators in the power larger than one.

4. The duration of a superradiant pulse is essentially shorter than the dephasing time $T_2$.

5. A superradiant pulse has a peak at a finite delay time.

In atomic systems, one may distinguish different types of superradiance, all having the same characteristic features, including a short pulse time $\tau_p < T_2$ and finite delay time $t_0 > 0$. First of all, superradiance can occur as a transient process or as a lasting repeated effect, depending on whether there is no external nonresonant pumping permanently applied to the system or, respectively, there exists such a pumping supporting atomic inversion. In the first case, the system is prepared in an inverted state, after which no nonresonant pumping is involved. Then, if the relaxation process is gently promoted at $t = 0$ by an external pulse imposing weak initial coherence on the system, triggered superradiance may develop. When no initial coherence is thrust on the system, but superradiance appears as a completely self-organized effect starting from a purely incoherent state, then this is named pure superradiance. In the case, when the system is subject to the action of a nonresonant pumping constantly supporting atomic inversion, the regime of pulsing superradiance may arise, with a long series of superradiant bursts.
It has been of great interest to find out if a kind of spin superradiance, occurring in spin systems similarly to this phenomenon in atomic assemblies, could be realized experimentally and, if so, how should it be described theoretically. Despite several similarities between spin and atomic ensembles, they are, nevertheless, rather different. For example, spontaneous radiation, starting the process of pure atomic superradiance, is absent in spin assemblies. Therefore, one of the most intriguing questions has been if pure spin superradiance can exist and, if so, what would be its origin.

The phenomenon of spin superradiance, being realized, could be of interest by its own and, in addition, it could be employed for several applications, as is discussed in section 11. For example, these applications could include:

(i) \textit{Investigation of materials characteristics} by measuring the relaxation parameters that are specific for spin superradiance and do not arise in other types of spin relaxation.

(ii) \textit{Fast repolarization of targets} used in various scattering experiments of high-energy physics.

(iii) \textit{Construction of spin masers} producing coherent radiation at radiofrequencies.

(iv) \textit{Creation of sensitive detectors} of weak external pulses, based on the mechanism of triggered spin superradiance.

(v) \textit{Method of information processing}, derived from the feasibility of regulating the number of and the intervals between superradiant bursts in the regime of punctuated spin superradiance.

\section{Experimental Observation}

Historically, the regime of \textit{pulsing spin superradiance} was observed first\textsuperscript{8-10}. This was done for a ruby crystal $\text{Al}_2\text{O}_3$, with the $\text{Cr}^{3+}$ paramagnetic admixture. The active nuclei were $^{27}\text{Al}$, with spins $I = 5/2$. If the ruby crystal is oriented in an external magnetic field such that a fully resolved quadrupole structure of its five $\Delta m = \pm 1$ NMR transitions can be observed, and a resonant circuit is tuned to a selected NMR line, then $^{27}\text{Al}$ spins form a fictitious two-level system. In experiments, the resonant circuit was tuned to the central $\{-\frac{1}{2}, \frac{1}{2}\}$ line. The density of $^{27}\text{Al}$ nuclei was $\rho_{\text{Al}} = 4.4 \times 10^{22}$ cm$^{-3}$ and that of $\text{Cr}^{3+}$, $\rho_{\text{Cr}} = 8.6 \times 10^{18}$ cm$^{-3}$. The measurements were performed in a static magnetic field of about $B_0 = 1.1$ T, which for the $^{27}\text{Al}$ magnetogyric ratio $7 \times 10^7$ s$^{-1}$T$^{-1}$ makes the NMR frequency $8 \times 10^7$ Hz. The temperature range was from 1.6 to 4.2 K. The ringing time of the resonant circuit was $\tau = 10^{-6}$ s, and the quality factor $Q$ was varied between 60 and 200. The coil filling factor was $\eta = 0.55$. The transverse spin damping time was $T_2 = 3 \times 10^{-5}$ s. The spin population inversion of the $^{27}\text{Al}$ nuclear spins was achieved by dynamic nuclear polarization, by means of powerful microwave radiation supplied to the sample in the vicinity of a selected $\text{Cr}^{3+}$ electron spin resonance line. The
corresponding longitudinal spin pumping time was $T_1^* = 0.1$ s. The emission of a long train of superradiant bursts was observed, with the delay time of the first pulse being $t_0 = 2 \times 10^{-3}$ s and its duration being $\tau_p = 2.6 \times 10^{-5}$ s in the case of the quality factor $Q = 60$, while $\tau_p = 8 \times 10^{-6}$ s for $Q = 200$.

The first observation of pure spin superradiance was accomplished in Dubna\textsuperscript{11,12}. A dielectric propanediol $\text{C}_3\text{H}_8\text{O}_2$, with a paramagnetic admixture of Cr$^{5+}$, was used as an active substance. This material possesses a high concentration of protons, with density $\rho_H = 4 \times 10^{22}$ cm$^{-3}$. The proton spins $I = 1/2$ played the role of radiators. The admixture of Cr$^{5+}$, with the density $\rho_{\text{Cr}} = 1.8 \times 10^{20}$ cm$^{-3}$, was employed for the purpose of dynamic nuclear polarization of proton spins. Experiments with the same material propanediol were repeated in Saint Petersburg\textsuperscript{13}. In all these experiments\textsuperscript{11−13}, electric circuits were used as resonators, with quality factors $Q$ between 100 and 600. The filling factor was $\eta = 0.6$. The volume of samples varied from 0.5 cm$^3$ to 12 cm$^3$. For external static magnetic fields $B_0$ in the interval between 0.5 T and 2.64 T, with the proton magnetogyric ratio $2.675 \times 10^8$ s$^{-1}$T$^{-1}$, the NMR proton frequencies were between $1.3 \times 10^8$ Hz and $7.1 \times 10^8$ Hz. The experiments were carried out at low temperatures of 0.05 K to 0.1 K. The inverted proton polarization reached 90%. Cooling of the sample resulted in strong suppression of the nuclear spin-lattice relaxation. The related longitudinal relaxation time was $T_1 = 1.8 \times 10^5$ s at $B_0 = 0.5$ T and $T_1 = 1.8 \times 10^6$ s at $B_0 = 2.64$ T. The transverse dephasing time was $T_2 = 0.85 \times 10^{-5}$ s. After preparing an inverted sample, with the proton polarization directed against an external static magnetic field $B_0$, the latter was scanned with the velocity about $5 \times 10^{-3}$ T$\cdot$s$^{-1}$. When in the process of this slow adiabatic scanning the field value $B_0$ reached that one for which the proton NMR frequency coincided with the circuit natural frequency, the resonance condition was met. Then a powerful superradiant burst was produced, with the duration time about $\tau_p = 1.3 \times 10^{-6}$ s.

Similar experiments, observing pure spin superradiance, were accomplished in Bonn\textsuperscript{14}, with the target materials butanol $\text{C}_4\text{H}_9\text{OH}$ and ammonia $\text{NH}_3$. These materials are also rich with protons, with density $\rho_H \approx 3 \times 10^{23}$ cm$^{-3}$. By means of dynamic nuclear polarization, proton polarizations of up to 99% were achieved. The samples were cooled to low temperatures, the spin-lattice relaxation time was $T_1 \approx 3.6 \times 10^4$ s for protons in butanol and $T_1 \approx 10^5$ s for protons in ammonia. The resonant electric circuit, with the quality factor $Q = 33$, had a resonance frequency of $1.6 \times 10^8$ Hz.

In different experiments, slightly different setups were employed. A typical arrangement\textsuperscript{11,12} is shown in Fig. 1. The characteristic superradiant pulse\textsuperscript{11,12} is presented in Fig. 2.

A series of numerical simulations were accomplished\textsuperscript{15,16}, which could be considered as computer experiments modelling the behaviour of spins in conditions typical of experimental observations. These computer simulations confirmed the existence of both pure as well as triggered spin superradiance, being in good agreement with experiments.
3 Basic Model

Since the phenomenon of spin superradiance exists in nature, it is necessary to develop its theoretical description. The simplest idea would be to invoke for this purpose the Bloch equations. However, these are designed so that they treat the magnetization of a sample as a macroscopic vector, which presupposes the existing coherence from the very beginning. If at the initial time coherence of spins is absent, the Bloch equations would never display it. Therefore, these equations can describe only triggered spin superradiance, when coherence is thrust upon spins at the initial time by assuming the presence of nonzero transverse magnetization. But pure spin superradiance, being a self-organized coherent process cannot in principle be treated by the Bloch equations. A discussion of this problem as well as the related references can be found in a review\textsuperscript{17}. In order to be able to describe all possible regimes of spin dynamics, it is necessary to resort to microscopic models. A theory of nuclear spin superradiance, based on realistic Hamiltonians typical of nuclear spin assemblies in condensed matter\textsuperscript{1,2,18} has been previously developed\textsuperscript{19–24}.

A system of $N$ nuclear spins, enumerated by an index $i = 1, 2, \ldots, N$, is characterized by their spin operators $\hat{\mathbf{I}}_i$. The Hamiltonian of nuclear spins in a solid can be written as a sum

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

of the Zeeman term and of a part due to many-body spin interactions. The Zeeman term contains

$$\hat{H}_i = -\hbar \gamma_n \mathbf{B} \cdot \hat{\mathbf{I}}_i,$$

where $\mathbf{B}$ is the total magnetic field and $\gamma_n$ is the nuclear magnetogyric ratio. The total magnetic field

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

consists of a static magnetic field $B_0$ along the $z$-axis and of a transverse field being a sum of an external field $B_1$ and of a field $H$ produced by an electric coil which the sample is immersed into. The corresponding orientation of the coordinate axes with respect to the sample is explained in Fig. 3. The static magnetic field is directed so that

$$\gamma_n B_0 < 0.$$ \hfill (4)

In general, the nuclear magnetogyric ratio can be positive as well as negative. If it is positive, then inequality (4) tells that the static field is negative. In this case, the initial polarization of the sample nuclei is called inverted if it is positive, that is directed against the static magnetic field.

The basic force acting between nuclear spins is due to the dipolar interactions which may be described by the Hamiltonian

$$\hat{H}_{ij} = \sum_{\alpha \beta} D_{ij}^{\alpha \beta} \hat{\mathbf{I}}_i^\alpha \hat{\mathbf{I}}_j^\beta,$$ \hfill (5)
where the upper indices $\alpha$ and $\beta$ denote the coordinate components $\alpha, \beta = x, y, z$, while

$$D^{\alpha\beta}_{ij} = \frac{\hbar^2 \gamma_n^2}{r_{ij}^3} \left( \delta_{\alpha\beta} - 3n_{ij}^\alpha n_{ij}^\beta \right)$$

(6)

is the dipolar tensor, in which

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

The dipolar tensor satisfies the equalities

$$\sum_\alpha D^{\alpha\alpha}_{ij} = 0, \quad \sum_j (\delta_{ij} - n_{ij} n_{ij}) D^{\alpha\beta}_{ij} = 0,$$

(7)

of which the first is exact and the second is asymptotically valid for a macroscopic sample.

For practical purpose, it is convenient to work with the ladder spin operators

$$\hat{I}_j^\pm \equiv \hat{I}^x_j \pm i\hat{I}^y_j,$$

called the raising and lowering spin operators, respectively. Then, with the help of the notation

$$a_{ij} \equiv D^{zz}_{ij}, \quad b_{ij} \equiv \frac{1}{4} \left( D^{xx}_{ij} - 2iD^{xy}_{ij} - D^{yy}_{ij} \right), \quad c_{ij} \equiv \frac{1}{2} \left( D^{xz}_{ij} - iD^{yz}_{ij} \right),$$

(8)

the Zeeman term (2), for the total field (3), becomes

$$\hat{H}_i = -\hbar \gamma_n B_0 \hat{I}_i^z - \frac{1}{2} \hbar \gamma_n (B_1 + H) \left( \hat{I}_i^+ + \hat{I}_i^- \right)$$

(9)

and the dipolar interaction (5) takes the form

$$\hat{H}_{ij} = a_{ij} \left( \hat{I}_i^z \hat{I}_j^z - \frac{1}{2} \hat{I}_i^+ \hat{I}_j^- \right) + b_{ij} \hat{I}_i^+ \hat{I}_j^+ + b_{ij}^* \hat{I}_i^- \hat{I}_j^- +$$

$$+ 2c_{ij} \hat{I}_i^+ \hat{I}_j^+ + 2c_{ij}^* \hat{I}_i^- \hat{I}_j^-.$$

(10)

Equations (9) and (10) will be used below to derive the evolution equations.

The electric circuit, coupled with the spin sample, is characterized by resistance $R$, inductance $L$, and capacity $C$. The coil, surrounding the sample, has $n$ turns of cross-section area $A_c$ over a length $l$. The electric current $j$ of the circuit is given by the Kirchhoff equation

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

(11)

in which $\tilde{E}$ is an electromotive force, if any, and $\Phi$ is a magnetic flux

$$\Phi = \frac{4\pi}{c} nA_c \eta M_x$$

(12)
formed by the $x$-component of the magnetization density
\[
M_x = \frac{\hbar \gamma_n}{V} \sum_i < \hat{I}_i^x > .
\] (13)

Here the brackets $< \ldots >$ imply statistical averaging. The filling factor $\eta$ is approximately equal to $\eta \approx V / V_c$, where $V$ is the sample volume, while $V_c \equiv A_c l$ is the coil volume.

The electric current in the circuit is formed by moving transverse spins. In its turn, the current, circulating over the coil, produces a feedback magnetic field
\[
H = 4\pi \frac{n j}{c l} .
\] (14)

Hence, the Kirchhoff equation (11) can be rewritten for the feedback field (14). To this end, it is useful to employ the notation for the natural circuit frequency
\[
\omega \equiv \frac{1}{\sqrt{LC}} \quad \left( L \equiv 4\pi \frac{n^2 A_c}{c^2 l} \right)
\] (15)

and for the circuit ringing time
\[
\tau \equiv \frac{1}{\gamma} \quad \left( \gamma \equiv \frac{R}{2L} \right)
\] (16)

The related circuit damping
\[
\gamma = \frac{\omega}{2Q} \quad \left( Q \equiv \frac{\omega L}{R} \right)
\] (17)

is connected with the quality factor $Q$. Employing these notations, together with that for the reduced electromotive force
\[
h \equiv \frac{c \tilde{E}}{n A_c \gamma} ,
\] (18)

one obtains from the Kirchhoff equation (11) the result
\[
\frac{dH}{dt} + 2\gamma H + \omega^2 \int_0^t H(t') \ dt' = \gamma h - 4\pi \eta \frac{dM_x}{dt}
\] (19)

for the feedback magnetic field (14).

The feedback equation (19) can be presented in a form that extremely useful to exploit for analysing the evolution equations20. By envolving the method of Laplace transforms and introducing the transfer function
\[
G(t) = \left( \cos \omega t - \frac{\gamma}{\omega} \sin \omega t \right) e^{-\gamma t} ,
\]

with $\tilde{\omega} \equiv \sqrt{\omega^2 - \gamma^2}$, one may present eq. (19) as the integral
\[
H = \int_0^t G(t-t') \left[ \gamma h(t') - 4\pi \eta \dot{M}_x(t') \right] \ dt',
\] (20)

where the dot over $\dot{M}_x$ means, as usual, time derivative.
4 Evolution Equations

The equations of motion for the nuclear spin operators are given by the corresponding Heisenberg equations. From these one aims at deriving the equations for the following statistical averages. The function

\[ u \equiv \frac{1}{I} < \hat{I}^-_i > \]  

defines the rotation of transverse spin components. The degree of coherence in this rotation is described by

\[ w \equiv \frac{1}{I^2} < \hat{I}^+_i > < \hat{I}^-_i > = |u|^2 . \]  

And the average

\[ s \equiv \frac{1}{I} < \hat{I}^z_i > \]

gives the longitudinal spin polarization, which is analogous to the population difference in optic systems. Keeping in mind that wavelengths at radio-frequencies are much larger than mean distances between spins and usually even essentially larger than linear sizes of a sample, one may employ the uniform approximation, assuming that the functions (21) to (23) do not depend on spatial variables.

In order to obtain the evolution equations for the quantities (21) to (23), one averages the Heisenberg equations for the corresponding spin operators. In doing so, one encounters the known problem of the resulting equations being not closed but containing, in addition to \( u \), \( w \), and \( s \), also binary averages of spin operators. The standard way of closing these equations would be by employing the mean-field decoupling, when the binary average \( < \hat{I}^\alpha_i \hat{I}^\beta_j > \), with \( i \neq j \), is factorized onto the product \( < \hat{I}^\alpha_i > < \hat{I}^\beta_j > \). Such a decoupling, however, ends with the Bloch-type equations having the same deficiency of never exhibiting coherence if at the initial time \( w(0) = 0 \). This is because the mean-field decoupling completely neglects spin correlations leading to local spin fluctuations. Taking account of such fluctuations is a necessary prerequisite for describing pure spin superradiance.

Another possibility of closing the hierarchy of spin evolution equations could be by writing additional equations for the binary correlators \( < \hat{I}^\alpha_i \hat{I}^\beta_j > \) and by decoupling only the higher-order correlators, retaining untouched the binary ones. Unfortunately, this drastically increases the number of equations and so complicates the problem that it becomes practically untreatable even for equilibrium magnets\(^25\). The more so for nonequilibrium systems presented by nonlinear differential equations.

To render the set of spin evolution equations closed so that to keep the problem treatable and, at the same time, to take into account local spin fluctuations, one can apply the method of stochastic decoupling\(^19\)–\(^21\),\(^24\). The main idea of the latter is to separate in the evolution equations the terms describing long-range correlations from the terms related to short-range fluctuations. Employing the method of restricted traces, one may define two types of statistical averages, one incorporating only
long-range correlations and another one involving short-range fields that are treated as stochastic variables. In what follows, the first type of averaging is denoted by the single angle brackets $< \ldots >$, while the second type, by the double brackets $\langle \ldots \rangle$. Then, the binary spin correlators are presented as

$$< \hat{I}_i^\alpha \hat{I}_j^\beta > = < \hat{I}_i^\alpha > < \hat{I}_j^\beta > \quad (i \neq j) ,$$

where the averaging does not touch stochastic variables which enter the evolution equations in the following linear combinations:

$$\xi_0 \equiv \frac{1}{\hbar} \sum_{j \neq i} \left( a_{ij} \hat{I}_j^z + c_{ij} \hat{I}_j^+ + c_{ij}^* \hat{I}_j^- \right) ,$$

$$\xi \equiv - \frac{i}{\hbar} \sum_{j \neq i} \left( \frac{1}{2} a_{ij} \hat{I}_j^- - 2 b_{ij} \hat{I}_j^+ - 2 c_{ij} \hat{I}_j^z \right) ,$$

with the coefficients defined in eq. (8). Stochastic variables (25) describe local fluctuating fields which act on neighbour spins forcing them to move. The existence of such fluctuating fields should lead to the appearance of dipolar dynamic broadening\textsuperscript{26}. To completely define the problem, it is necessary to prescribe the rules of calculating stochastic averages over the random variables (25). This can be done by considering the stochastic variables (25) as describing a white-noise process characterized by the stochastic averages

$$\langle \langle \xi_0 (t) \rangle \rangle = \langle \xi (t) \rangle = 0 , \quad \langle \langle \xi_0 (t) \xi_0 (t') \rangle \rangle = 2 \Gamma_3 \delta (t - t') ,$$

$$\langle \langle \xi_0 (t) \xi (t') \rangle \rangle = \langle \langle \xi (t) \xi (t') \rangle \rangle = 0 , \quad \langle \langle \xi^* (t) \xi (t') \rangle \rangle = 2 \Gamma_3 \delta (t - t') ,$$

in which $\Gamma_3$ is the width of dynamic broadening due to local dipole interactions of nuclei. This is a sort of inhomogeneous broadening existing additionally to the homogeneous broadening yielding the longitudinal, $\Gamma_1 \equiv T^{-1}_1$, and transverse, $\Gamma_2 \equiv T^{-1}_2$, relaxation widths.

To obtain the resulting evolution equations in a compact form, it is convenient to introduce the notation

$$f \equiv - i \gamma_n \left( B_1 + H \right) + \xi \quad (27)$$

for an effective force acting on a spin. The transverse external magnetic field $B_1$ may, in general, contain a static as well as an alternating term, as

$$B_1 = h_1 + h_2 \cos \omega t , \quad (28)$$

where, for simplicity, the frequency of the alternating field is assumed to be in resonance with the frequency of the electric circuit. Recall that the magnetic field $H$, produced by the coil, is caused by a feedback field and, possibly, by a field of an electromotive force, which may be presented as

$$h(t) = h_0 \cos \omega t . \quad (29)$$
Finally, the NMR frequency is denoted by
\[ \omega_0 \equiv |\gamma_n B_0| \tag{30} \]
Then the evolution equations for the functions (21) to (23) acquire the form
\[ \frac{du}{dt} = -i(\omega_0 + \xi_0 - i\Gamma_2)u + f_s \tag{31} \]
\[ \frac{dw}{dt} = -2\Gamma_2 w + (u^*f + f^*u)s \tag{32} \]
\[ \frac{ds}{dt} = -\frac{1}{2}(u^*f + f^*u) - \Gamma_1(s - \zeta) \tag{33} \]
where \( \zeta \) is an average stationary value for a \( z \)-component of a spin. Generally, \(-1 \leq \zeta \leq 1\). When there is no external pumping, then \( \zeta = -1 \). In the presence of pumping, e.g. by means of dynamic nuclear polarization, the pumping parameter \( \zeta > -1 \) and can reach the value \( \zeta = 1 \). Equations (31) to (33) compose a nonlinear system of stochastic differential equations describing all dynamic properties of nuclear spins.

The system of evolution equations (31) to (33) looks yet rather complicated. Fortunately, there are several small parameters that allow one to simplify the consideration by employing the \textit{scale separation approach} \cite{19,21,27}, which is a generalization of the Krylov-Bogolubov averaging technique \cite{28} to stochastic and partial differential equations. The existing small parameters are connected with the relatively small values of the relaxation widths \( \Gamma_1 \) and \( \Gamma_2 \) as compared to the NMR frequency (30), so that
\[ \frac{\Gamma_1}{\omega_0} \ll 1 , \quad \frac{\Gamma_2}{\omega_0} \ll 1 . \tag{34} \]
Clearly, the quantity
\[ \Gamma_0 \equiv \pi \eta \rho_n \gamma_2^2 \hbar I \left( \rho_n \equiv \frac{N}{V} \right) \tag{35} \]
is also small with respect to \( \omega_0 \), as well as the dynamic broadening width \( \Gamma_3 \), that is
\[ \frac{\Gamma_0}{\omega_0} \ll 1 , \quad \frac{\Gamma_3}{\omega_0} \ll 1 . \tag{36} \]
All amplitudes of the applied transverse fields are assumed to be small too, which implies that the values
\[ \nu_0 \equiv \gamma_n h_0 , \quad \nu_1 \equiv \gamma_n h_1 , \quad \nu_2 \equiv \gamma_n h_2 \tag{37} \]
satisfy the inequalities
\[ \frac{|\nu_0|}{\omega_0} \ll 1 , \quad \frac{|\nu_1|}{\omega_0} \ll 1 , \quad \frac{|\nu_2|}{\omega_0} \ll 1 . \tag{38} \]
The electric circuit, coupled to the spin system, is supposed to be of good quality, having a high quality factor, which means that the ringing width is small compared to the natural circuit frequency,

\[ \frac{\gamma}{\omega} \ll 1 \quad (Q \gg 1) \]  

(39)

The last, though not the least, is that the electric circuit has to be tuned to the NMR frequency (30), hence the resonance condition for the detuning \( \Delta \) must be valid:

\[ \frac{|\Delta|}{\omega_0} \ll 1 \quad (\Delta \equiv \omega - \omega_0) \]  

(40)

In this way, the circuit plays the role of a resonator.

The existence of the small parameters makes it possible, by analysing the right-hand sides of the evolution equations (31) to (33), to conclude that the function \( u \) has to be classified as fast, as compared to the slow functions \( w \) and \( s \). Conversely, the functions \( w \) and \( s \) are quasi-invariants with respect to \( u \). In addition, one may derive an explicit expression for the resonator magnetic field \( H \) by iterating its integral representation with the solution of eq. (31), where only the main term in the right-hand side is retained. This iteration gives

\[ \gamma_n H = i\alpha(u - u^*) + \beta \cos \omega t, \]  

(41)

where the first term, with the coupling function

\[ \alpha \equiv \frac{\Gamma_0\omega_0}{\gamma} \left(1 - e^{-\gamma t}\right), \]  

(42)

is caused by the feedback coupling of spins with the resonator, and the second term, with the coupling

\[ \beta \equiv \frac{\nu_0}{2} \left(1 - e^{-\gamma t}\right), \]  

(43)

is due to an electromotive force. The time dependence of the coupling functions (42) and (43) describes the retardation in the resonator action on spins. Expressions (42) and (43) are written here for the case of small detuning, such that \( |\Delta| \ll \gamma \). The latter assumption is not principal but is employed solely for the simplification of formulas. Relation (41) holds true for any detuning satisfying the resonance condition (40).

Substituting relation (41) in eqs. (31) to (33) and using the notation

\[ f_1 \equiv -i\nu_1 - i(\nu_2 + \beta) \cos \omega t + \xi, \]  

(44)

one comes to the evolution equations

\[ \frac{du}{dt} = -[i(\omega_0 + \xi_0) + \Gamma_2 - \alpha s] u + f_1 s - \alpha s u^*, \]  

(45)

\[ \frac{dw}{dt} = -2(\Gamma_2 - \alpha s)w + (u^* f_1 + f_1^* u) s - \alpha s \left[(u^*)^2 + u^2\right], \]  

(46)
\[
\frac{ds}{dt} = -\alpha w - \frac{1}{2} (u^* f_1 + f_1^* u) - \Gamma_1 (s - \zeta) + \frac{1}{2} \alpha \left[ (u^*)^2 + u^2 \right]. \tag{47}
\]

Following further the scale separation approach, eq. (45) for the fast function \( u \) can be solved, with \( w \) and \( s \) being quasi-invariants. The solution obtained is substituted in the right-hand sides of eqs. (46) and (47), which are averaged over the period \( 2\pi/\omega_0 \) of fast oscillations as well as over stochastic variables defined in eq. (25). Then, introducing the effective attenuation width

\[
\Gamma_{\text{eff}} \equiv \Gamma_3 - \alpha \nu_1^2 \frac{s}{\omega_0^2} - \frac{\nu_1 (\nu_2 + \beta) \Gamma}{2 \omega_0^2} e^{-\Gamma t} + \frac{(\nu_2 + \beta)^2}{4 \Gamma} \left( 1 - e^{-\Gamma t} \right), \tag{48}
\]

where

\[
\Gamma \equiv \Gamma_2 - \alpha s + \Gamma_3,
\]

reduces eqs. (46) and (47) for slow functions to the evolution equations

\[
\frac{dw}{dt} = -2(\Gamma_2 - \alpha s)w + 2\Gamma_{\text{eff}} s^2, \tag{49}
\]

\[
\frac{ds}{dt} = -\alpha w - \Gamma_{\text{eff}} s - \Gamma_1 (s - \zeta). \tag{50}
\]

In the form of the widths (48), it is again assumed that the detuning is small, such that \( |\Delta| \ll |\gamma| \ll \omega_0 \), which is not principal but just simplifies cumbersome expressions.

The evolution equations (49) and (50) are the main equations describing the dynamics of strongly nonequilibrium nuclear spins. The analysis of these equations makes it possible to study various regimes of spin motion.

## 5 Nyquist Noise

The problem of principal importance is: What is the origin of pure spin superradiance? In other words, what initiates the motion of spins when no coherence is thrust upon the system at \( t = 0 \) and there are no external fields pushing spins in the transverse direction?

Keeping in mind the analogy with atomic assemblies, one may remember that in an inverted system of atoms the relaxation process begins with atomic spontaneous radiation, which is a quantum process. After the seed radiation field appears in the system, atomic correlations start arising through the interatomic photon exchange. As soon as these correlations become sufficiently intensive, coherence develops. Then, the quantum stage of spontaneous emission changes for the coherent stage, when atoms are correlated and emit coherently, which results in superradiance.

The collectivization of spins can be produced by means of the resonator feedback field. But for this field to arise, the spins, first, have to start their motion. Spontaneous emission for spins is absent. Then, what could initiate the motion of spins? What mechanism would be the origin of pure spin superradiance?
There existed a widespread delusion that the thermal Nyquist noise of the resonant electric circuit could be the initiating cause of spin rotation. At this point, it is necessary to stress that the role of the thermal Nyquist noise in producing a fluctuating torque on the magnetization was in detail discussed by Bloembergen and Pound in their classic paper\(^6\). They showed that the average thermal damping is inversely proportional to the sample volume, because of which this damping could be noticeable only for a single spin, but for a macroscopic sample of many spins \(N \gg 1\) the coil thermal damping should be negligibly small. This analysis seems to have been completely forgotten by later workers who have proclaimed the leading role of the coil thermal noise in initiating spin rotation.

To explicitly illustrate the role of the thermal Nyquist noise, one has to consider the effective attenuation width (48), including the influence of the electromotive force related to the resonance mode of the thermal noise\(^{19-21}\). When there are no external transverse magnetic fields, that is when \(\nu_1 = \nu_2 = 0\), the effective width (48) reads

\[
\Gamma_{\text{eff}} = \Gamma_3 + \frac{\nu_0^2}{16\Gamma} \left(1 - e^{-\Gamma t}\right) \left(1 - e^{-\gamma t}\right)^2.
\] (51)

The electromotive force, corresponding to the resonator thermal noise, can be written as

\[
\tilde{E} = \tilde{E}_0 \cos \omega t.
\] (52)

The related magnetic field, defined in eqs. (18) and (29), has the amplitude

\[
h_0 = \frac{c\tilde{E}_0}{nA_e\gamma}.
\] (53)

From here, it follows that

\[
h_0^2 = 8\pi \frac{\eta \rho_0 \tilde{E}_0^2}{\gamma RN}.
\] (54)

With the definition (37), one gets the quantity

\[
\nu_0^2 = \frac{8\Gamma_0 \tilde{E}_0^2}{h\gamma T R N},
\] (55)

which characterizes in eq. (51) the average thermal damping due to the Nyquist noise.

First of all, eq. (51) shows that the thermal damping at \(t = 0\) is exactly zero because of the temporal factors. The latter become essential only for \(t > T_2, \tau\). Therefore, the influence of the thermal noise at short times should be suppressed by these temporal factors.

Moreover, the quantity (55) is inversely proportional to the number of spins. That is, the thermal damping is inversely proportional to \(N\), and thus it should be negligibly small for a macroscopic sample with \(N \gg 1\). This is in a complete agreement with the conclusion of Bloembergen and Pound\(^6\).

To be more precise, it is possible to explicitly calculate the quantity (55) substituting there the square amplitude of the electromotive force due to the thermal
noise\textsuperscript{7}, which is
\[ \tilde{E}_0^2 = \frac{\hbar \omega}{2\pi} \gamma R \coth \frac{\hbar \omega}{2k_B T}. \] (56)

At radio-frequencies, the inequality
\[ \frac{\omega}{\omega_T} \ll 1 \quad \left( \omega_T \equiv \frac{k_B T}{\hbar} \right) \] (57)
is valid. As a result, eq. (56) simplifies to
\[ \tilde{E}_0^2 \simeq \frac{\hbar}{\pi} \gamma R \omega_T. \] (58)

Thence, eq. (55) becomes
\[ \nu_0^2 = \frac{8\Gamma_0 \omega_T}{\pi IN}. \] (59)

Expression (51) shows that the thermal damping has to be compared to the dynamic broadening width $\Gamma_3$, which can be of order of $\Gamma_2$. When the inequality
\[ \frac{\nu_0^2}{\Gamma_2^2} \ll 1 \] (60)
holds true, the thermal damping can be safely neglected, playing no role in spin relaxation, as compared to the dynamic broadening.

To estimate the factor (59) and, respectively, to check the inequality (60), one can accept the values typical of experiments on proton spin superradiance\textsuperscript{11–13}. Then, for the filling factor $\eta = 0.6$, proton density $\rho_n = 4 \times 10^{22} \text{ cm}^{-3}$, and the proton magnetogyric ratio $\gamma_n = 2.675 \times 10^8 \text{ s}^{-1}\text{T}^{-1}$, the linewidth (35) is $\Gamma_0 = 2.846 \times 10^4 \text{ Hz}$. At temperature $T = 0.1 \text{ K}$, the thermal frequency is $\omega_T = 1.309 \times 10^{11} \text{ Hz}$. Under the static magnetic field $B_0 = 1 \text{ T}$, the proton NMR frequency is $\omega_0 = 2.675 \times 10^8 \text{ Hz}$. The ratio $\omega/\omega_T \sim 10^{-3}$ is small. With the transverse relaxation width $\Gamma_2 = 1.176 \times 10^5 \text{ Hz}$, one obtains
\[ \frac{\nu_0^2}{\Gamma_2^2} = \frac{1.37 \times 10^6}{N}. \]

For a sample volume of about 1 $\text{ cm}^3$ and $N \sim 10^{23}$, one has $\nu_0^2/\Gamma_2^2 \sim 10^{-17}$. This ratio is so much tiny that, clearly, the thermal Nyquist noise is not able to play any role in spin relaxation in a macroscopic sample. It is only for the number of spins $N \leq 10^6$, when the thermal noise would be noticeable. When considering macroscopic samples, with $N \gg 10^6$, the resonator thermal noise is to be neglected.

6 \textbf{Incoherent Stage}

At the initial stage, the main role in starting spin relaxation is played by the dynamic broadening due to local spin fluctuations. Omitting in the effective width (48) the
terms of second order with respect to small parameters and neglecting the resonator thermal noise by setting \( \beta \to 0 \), one has

\[
\Gamma_{\text{eff}} = \Gamma_3 + \frac{\nu^2}{4\Gamma} \left( 1 - e^{-\Gamma t} \right) .
\]

(61)

This equation shows that at the very beginning of the process, when \( t \to 0 \), then \( \Gamma_{\text{eff}} \to \Gamma_3 \). That is, the dynamic broadening width \( \Gamma_3 \) makes the major contribution to the effective relaxation width, eq. (61).

For asymptotically small times \( t \to 0 \), the coupling function (42) is close to zero, \( \alpha \to 0 \). Then the evolution equations (49) and (50) can be reduced to the form

\[
\frac{dw}{dt} = -2\Gamma_2 w + 2\Gamma_3 s^2 , \quad \frac{ds}{dt} = -\Gamma_1^*(s - \zeta^*) ,
\]

(62)
in which

\[
\Gamma_1^* \equiv \Gamma_1 + \Gamma_3 , \quad \zeta^* \equiv \frac{\Gamma_1}{\Gamma_1^*} \zeta .
\]

(63)

The solutions to eqs. (62) are

\[
w = w_0 e^{-2\Gamma_2 t} + 2\Gamma_3 \int_0^t s^2(t') e^{-2\Gamma_2(t-t')} \, dt' , \quad s = \zeta^* + (s_0 - \zeta^*) e^{-\Gamma_1^* t} ,
\]

(64)

which demonstrates the character of spin motion at short times \( t \to 0 \). At this stage, the motion of spins is yet completely incoherent, since, because of retardation, the coupling with the resonator has not yet been switched on. Assuming that \( \Gamma_1 \ll \Gamma_3 \), hence \( \zeta^* \ll 1 \), one may simplify the solution (64) as

\[
w \simeq w_0 e^{-2\Gamma_2 t} + \frac{\Gamma_3 s_0^2}{\Gamma_2 - \Gamma_3} \left( e^{-2\Gamma_3 t} - e^{-2\Gamma_2 t} \right) , \quad s \simeq s_0 e^{-\Gamma_1^* t} .
\]

The initial incoherent stage of spin relaxation can also be called the quantum stage, as far as the relaxation is caused by quantum effects of spin-spin interactions and there are yet no collective effects that could lead to the development of coherence.

The duration of the incoherent quantum stage lasts till the quantum time \( t_q \), when the coupling function (42) reaches the value \( \Gamma_2 \), so that

\[
\alpha(t_q) = \Gamma_2 ,
\]

(65)

and when taking account of collective effects, due to the coupling with the resonator, becomes important. It is also easy to notice that the difference \( \Delta = \Gamma_2 - \alpha s \) in eq. (49) may change its sign at the quantum time \( t_q \), which means that the generation of coherent radiation would begin. Combining eqs. (42) and (65) gives for the quantum time

\[
t_q = \tau \ln \frac{g}{g - 1} ,
\]

(66)

where the notation of the effective coupling parameter

\[
g \equiv \frac{\Gamma_0 \omega_0}{\Gamma_2 \gamma} = 2Q \frac{\Gamma_0}{\Gamma_2} .
\]

(67)
is introduced. The quantum time (66) is positive only for \( g > 1 \). If \( g = 1 \), then \( t_q \to \infty \). This indicates that the quantum stage is finite and may change to a collective stage only if the coupling parameter (67) is \( g > 1 \). If the spin-resonator coupling is weak, and \( g \leq 1 \), there exists solely the quantum stage, and the collective stage never comes into being. For sufficiently strong coupling, eq. (66) gives

\[
    t_q \simeq \frac{\tau}{g} \quad (g \gg 1).
\]  

(68)

The values of solutions (64) at the end of the quantum stage, i.e. at \( t_q \), can be estimated assuming that \( t_q \ll T_2 \) and \( t_q \ll T_3 \equiv \Gamma_3^{-1} \). Then eqs. (64) yield

\[
    w(t_q) \simeq w_0 + 2\delta_3 s^2, \quad s(t_q) \simeq s_0,
\]  

(69)

where

\[
    \delta_3 \equiv \frac{\Gamma_3}{g\gamma} = \frac{\Gamma_2 \Gamma_3}{\Gamma_0 \omega_0}. \tag{70}
\]

This is a small parameter, since \( \Gamma_0 \sim \Gamma_3 \) while \( \Gamma_2 \ll \omega \). But, no matter how small \( \delta_3 \) is, it can be principally important if \( w_0 = 0 \).

7 Transient Superradiance

After the incoherent quantum stage, the coupling function (42) increases, switching on collective effects resulting in the appearance of spin superradiance\(^{19-21}\). An accurate description of this process can be made by solving the evolution equations (49) and (50) numerically. It is also possible to present an explicit analytical description of how superradiance develops for the transient stage, when the time is larger than the quantum time \( t_q \) but yet much smaller than the spin-lattice relaxation time \( T_1 \). Then, the term with \( \Gamma_1 \) in eq. (50) can be omitted. If there are no strong external transverse fields, then one should set \( \nu_1 \to 0 \) and \( \nu_2 \to 0 \). Assuming that the coupling function (42) has reached its maximal value, one gets \( \alpha \approx g\Gamma_2 \). The dynamic broadening width \( \Gamma_3 \) is not larger than \( \Gamma_2 \). Hence, for sufficiently large coupling parameter \( g \), one may neglect \( \Gamma_3 \) as compared to \( g\Gamma_2 \) \( \gg \Gamma_3 \). Under these conditions, eqs. (49) and (50) read

\[
    \frac{dw}{dt} = -2\Gamma_2(1-gs)w, \quad \frac{ds}{dt} = -g\Gamma_2 w. \tag{71}
\]

Initial conditions for eqs. (71) should be taken at \( t = t_q \).

Equations (71) can be solved exactly\(^{19-21}\) yielding

\[
    w = \left( \frac{\Gamma_p}{g\Gamma_2} \right)^2 \text{sech}^2 \left( \frac{t-t_0}{\tau_p} \right), \quad s = -\frac{\Gamma_p}{g\Gamma_2} \tanh \left( \frac{t-t_0}{\tau_p} \right) + \frac{1}{g}. \tag{72}
\]

Here \( \Gamma_p \) is a pulse width, \( \tau_p = \Gamma_p^{-1} \) is a pulse time, and \( t_0 \) is a delay time. These parameters are the integration constants that are to be defined from initial conditions.
\( w(t_q) \) and \( s(t_q) \) taken at the boundary of the transient stage, that is at the quantum time \( t_q \). Employing eqs. (69) results in

\[
\Gamma_p^2 = \Gamma_g^2 + (g \Gamma_2)^2 \left( w_0 + 2\delta_3 s_0^2 \right), \quad \Gamma_g \equiv \Gamma_2(1 - gs_0), \quad \Gamma_p \tau_p = 1, \quad (73)
\]

while the delay time is

\[
t_0 = t_q + \frac{\tau_p}{2} \ln \left| \frac{\Gamma_p - \Gamma_g}{\Gamma_p + \Gamma_g} \right|. \quad (74)
\]

Solutions (72) demonstrate that the coherence intensity \( w(t) \) has the shape of a burst of width \( \tau_p \) and peaked at the delay time \( t_0 \).

If the spin-resonator coupling \( g \) is sufficiently weak or the initial spin polarization \( s_0 \) is sufficiently small, so that \( gs_0 \leq 1 \), than the value of the delay time (74) shifts to the quantum region, becoming \( t_0 \leq t_q \). This means that the radiation intensity would not have the shape of a coherent burst, but is rather a decreasing function of time, typical of nuclear induction. In the case of an external pulse thrusting onto the spins an essential initial coherence, such that \( g^2 w_0 > 1 \), the radiation time \( \tau_p \) is small, \( \tau_p < T_2 \), which is characteristic of collective induction. When \( gs_0 > 1 \) and \( g^2 w_0 > 1 \), the signal of collective induction is peaked at a finite delay time \( t_0 > 0 \). As is explained in the Introduction, collective induction, with coherence being induced by an initial external source, is in principal different from superradiance that is a process of spontaneously arising coherence.

For solutions (72) to describe a superradiant burst, the requirements discussed in the Introduction must be satisfied. In order that the initial coherence, given by \( w_0 \), would not essentially influence the pulse width \( \Gamma_p \), it should be that

\[
g^2 w_0 < 1. \quad (75)
\]

This guaranties a basically spontaneus character of the process. Then, the pulse time \( \tau_p \) is to be sufficiently short and the delay time finite,

\[
\tau_p < T_2, \quad t_q < t_0 < \infty. \quad (76)
\]

The second of these inequalities yields

\[
gs_0 > 1, \quad w_0 + 2\delta_3 s_0^2 > 0. \quad (77)
\]

Combining all conditions (75) to (77), one sees that there exist two types of transient spin superradiance, whose classification, taking account of the smallness of the parameter \( \delta_3 \ll 1 \), can be accomplished as follows:

(i) **Triggered superradiance**, when

\[
gs_0 > 1 + \sqrt{1 - g^2 w_0}, \quad w_0 \neq 0.
\]

Here, a weak external pulse imposes an initial coherence on spins, but, being rather weak, this pulse plays just the role of a trigger, while the following development of coherence is mainly governed by internal properties.
(ii) Pure superradiance, when

\[ g s_0 > 2, \quad w_0 = 0. \]

This is a purely self-organized process, with no coherence prescribed from external sources.

In the case of pure spin superradiance, the characteristic quantities (73) and (74), having regard to \( \delta_3 \ll 1 \), become

\[
\Gamma_p \simeq (g s_0 - 1) \Gamma_2 \left[ 1 + \left( \frac{g s_0}{g s_0 - 1} \right)^2 \delta_3 \right], \quad \tau_p \simeq \frac{T_2}{g s_0 - 1} \left[ 1 - \left( \frac{g s_0}{g s_0 - 1} \right)^2 \delta_3 \right],
\]

\[
t_0 \simeq \frac{\tau}{g} + \frac{T_2}{2(g s_0 - 1)} \ln \left| \frac{2}{\delta_3} \left( 1 - \frac{1}{g s_0} \right)^2 \right|. \tag{78}
\]

Although the parameter \( \delta_3 \) is small, it cannot be neglected, since its value is crucial for defining the delay time \( t_0 \). As is seen, if \( \delta_3 \to 0 \), then \( t_0 \to \infty \). The parameter \( \delta_3 \), according to the notation (70), is proportional to the dynamic broadening width \( \Gamma_3 \) due to local spin fluctuations. As far as the latter fluctuations are the main cause of starting spin relaxation, it is not surprising that the related broadening defines the delay time.

From eqs. (72) it follows that the maximum of the coherent burst occurs at \( t = t_0 \), when

\[
w(t_0) = \left( s_0 - \frac{1}{g} \right)^2, \quad s(t_0) = \frac{1}{g}. \tag{79}
\]

For longer times \( t \gg t_0 \), the superradiant signal exponentially diminishes, and the spin polarization returns to an almost inverted value,

\[
w \simeq 4w(t_0)e^{-2\Gamma_p t}, \quad s \simeq -s_0 + \frac{2}{g}. \tag{80}
\]

The larger is the spin-resonator coupling \( g \), the more complete is the spin inversion. The effect of superradiant spin inversion can be employed for the ultrafast repolarization of nuclear targets used in scattering experiments\textsuperscript{14,29}.

The shape of a superradiant pulse, described by \( w(t) \) in eqs. (72), is in very good agreement with experiments\textsuperscript{11–13}. Thus, the measured signal, presented in Fig. 2, ideally fits the calculated function \( w(t) \), as is discussed in the papers\textsuperscript{11,12}. A concrete relation between the measured intensity of the signal and \( w(t) \) will be explained in Sec. 10.

It is worth emphasizing that the radiation intensity and radiation time of the studied pulse have the properties typical of a superradiant burst. To illustrate this, it is sufficient to recall that the initial inverted spin polarization \( s_0 = N_+/N \) is the ratio of the number of inverted spins to their total number in the sample. Inverted spins play the role of radiators, whose number is \( N_+ \). For large spin-resonator coupling \( g \gg 1 \), one has \( w(t_0) \sim s_0^2 \) and \( \tau_p \sim 1/s_0 \), which is clear from the consideration above. Therefore

\[
w(t_0) \sim N_+^2, \quad \tau_p \sim N_+^{-1},
\]
which is characteristic of superradiance.

When there is no constant pumping by means of dynamic nuclear polarization, there appears one dominant superradiant burst given by eqs. (72). In an inhomogeneous sample, after the first burst, secondary maser oscillations can arise, whose accurate description requires numerical investigation\textsuperscript{15–17,30–32}.

It is interesting to mention that the signals of nuclear spin echo can also be essentially amplified by the presence of a resonant electric circuit coupled with a spin system, which has been observed in experiment\textsuperscript{33}.

8 Pulsing Superradiance

Another regime of spin superradiance can be achieved if the inversion of nuclear spins is constantly supported by a pumping mechanism, for instance, by dynamic nuclear polarization. Then, a series of superradiant bursts can be produced, as is experimentally observed\textsuperscript{8–10}. Such a regime, with a series of superradiant pulses that may repeatedly arise during a rather long time, can be termed pulsing superradiance. The theoretical description of this regime is based on the evolution equations (49) and (50), which require numerical computations\textsuperscript{17,22,24}. However, for the late stage, when $t \gg T_2, \tau$, and the system is close to a stationary state, it is possible to solve the problem analytically.

At the late stage, when $t \gg \tau$, the coupling function (42) becomes $\alpha = g\Gamma_2$. As is explained in Section 5, the resonator thermal noise can always be neglected, that is $\beta \to 0$. When there are no strong external transverse fields, then $\nu_1 \to 0$ and $\nu_2 \to 0$. For the simplicity, the dynamic broadening width $\Gamma_3$ can also be neglected as compared to a large $g\Gamma_2$. Then the evolution equations (49) and (50) can be written as a two-dimensional dynamical system

\begin{equation}
\frac{dw}{dt} = v_1, \quad \frac{ds}{dt} = v_2,
\end{equation}

with the velocity fields

\begin{equation}
v_1 = -2\Gamma_2(1 - gs)w, \quad v_2 = -g\Gamma_2 w - \Gamma_1^*(s - \zeta).
\end{equation}

In the presence of dynamic nuclear polarization, $\Gamma_1^*$ is a pumping rate and $\zeta > 0$ is a pumping parameter.

An accurate theoretical analysis of eqs. (81) is given below. The velocity fields are taken in the simplified form (82). This is done for pedagogical reasons, in order not to become entangled into cumbersome formulas, but for emphasizing the principal ideas and techniques, on which such an analysis is based. Using the same methods, one could accomplish an analysis of the evolution equations (49) and (50), including the dynamic broadening and external transverse fields, which would result in much more intricate equations.

An important quantity characterizing local stability of motion is the local expansion rate\textsuperscript{34}

\begin{equation}
\Lambda(t) \equiv \frac{1}{t} \text{Re} \int_0^t \text{Tr} \hat{X}(t') \, dt',
\end{equation}

with

\begin{equation}
\hat{W} = \frac{1}{2} \frac{\partial^2}{\partial s^2} + \frac{1}{2} \frac{\partial^2}{\partial \zeta^2} + 2g\Gamma_2(1 - gs)w
\end{equation}
whose definition involves the Jacobian expansion matrix $\dot{X}$ that in the considered case is

$$\dot{X}(t) \equiv \begin{bmatrix} \frac{\partial v_1}{\partial w} & \frac{\partial v_1}{\partial s} \\ \frac{\partial v_2}{\partial w} & \frac{\partial v_2}{\partial s} \end{bmatrix}.$$  

If, at the moment $t$, the rate (83) is positive, $\Lambda(t) > 0$, this means that the phase volume of the dynamical system expands, while if $\Lambda(t) < 0$, then the phase volume contracts. The eigenvalues of the expansion matrix $\dot{X}$ are the local characteristic exponents

$$X^\pm = -\frac{1}{2} \left\{ \Gamma_1^* + 2\Gamma_2(1 - gs) \pm \sqrt{[\Gamma_1^* - 2\Gamma_2(1 - gs)]^2 - 8g^2\Gamma_2^2w} \right\},$$

whose real parts define the local Lyapunov exponents

$$\lambda^\pm(t) \equiv \text{Re} \ X^\pm(t).$$

The signs of the latter show whether the motion at the moment $t$ is stable or not.

In the theory of dynamical systems\cite{35}, one usually considers asymptotic stability related to the limit $t \to \infty$. The stationary, or fixed, points are given by the zeros of the velocity fields. The equations $v_1 = v_2 = 0$, with the velocities (82), yield two stationary solutions: one fixed point is

$$w_1^* = 0, \quad s_1^* = \zeta$$

and another is

$$w_2^* = \frac{\Gamma_1^*}{g^2\Gamma_2} (g\zeta - 1), \quad s_2^* = \frac{1}{g}.$$  

As is clear, among several solutions, only those could have sense whose values are in the region of validity of the considered functions. This region, as follows from eqs. (22) and (23), is limited by the inequalities

$$0 \leq w \leq 1, \quad -1 \leq s \leq 1.$$  

Thence, it is evident that the fixed point (85) may have sense only if $g\zeta \geq 1$. When $g\zeta = 1$, both fixed points (84) and (85) coincide. On the manifold of system parameters, the value $g\zeta = 1$ is termed a bifurcation point.

Because of the existence of two stationary solutions, the limit

$$\Lambda^* \equiv \lim_{t \to \infty} \Lambda(t) = \text{Re} \ \lim_{t \to \infty} \text{Tr} \ \dot{X}(t)$$

of the local expansion rate (83), for which $\Lambda^* = \lambda^+ + \lambda^-$, with the Lyapunov exponents

$$\lambda^\pm \equiv \text{Re} \ \lim_{t \to \infty} X^\pm(t),$$

possesses two different values. The local expansion rates at the first and second fixed points, respectively, are

$$\Lambda_1^* = -\Gamma_1^* - 2\Gamma_2(1 - g\zeta), \quad \Lambda_2^* = -\Gamma_1^*.$$  

(86)
According to the principle of minimal expansion, the smaller the local expansion rate, the more stable is a dynamic state. The values show that when \( g\zeta < 1 \), the fixed point (84) is more stable, while when \( g\zeta > 1 \), the stationary solution (85) becomes more stable.

The limits \( X^\pm \equiv \lim_{t \to \infty} X^\pm(t) \) of the characteristic exponents, at the corresponding fixed points, are

\[
X_1^+ = -\Gamma^*_1, \quad X_1^- = -2\Gamma_2(1 - g\zeta), \\
X_2^\pm = -\frac{\Gamma^*_1}{2} \left[ 1 \pm \sqrt{1 - 8 \frac{\Gamma_2}{\Gamma^*_1} (g\zeta - 1)} \right].
\] (87)

The related real parts \( \lambda^\pm = \text{Re} X^\pm \) define the Lyapunov exponents. Analysing eqs. (87) yields the following classification of the fixed points (84) and (85).

When \( g\zeta < 1 \), then the Lyapunov exponents for the stationary solution (84) are negative, \( \lambda_1^+ < 0 \). This tells that the fixed point (84) is a stable node. For the stationary solution (85), one has \( \lambda_2^+ < 0 \) but \( \lambda_2^- > 0 \), which means that the fixed point (85) is a saddle. Hence, for \( g\zeta < 1 \), spin dynamics tends to the stationary solution (84). The relaxation rates are given by \( \lambda_1^+ = X_1^+ \) defined in eqs. (87), from which it follows that the spin motion is incoherent.

In the case \( g\zeta = 1 \), the stationary solution is unique, since the fixed points (84) and (85) coincide. The related Lyapunov exponents are also the same: \( \lambda_1^+ = \lambda_2^- < 0 \), \( \lambda_1^- = \lambda_2^- = 0 \). The value \( g\zeta = 1 \) is associated with a bifurcation point.

With the increasing pumping, when

\[
1 < g\zeta < 1 + \frac{\Gamma^*_1}{8\Gamma_2},
\]

one gets \( \lambda_1^+ < 0 \), \( \lambda_1^- > 0 \), while \( \lambda_2^\pm < 0 \). This indicates that the features of the fixed points have been changed — the fixed point (84) is now a saddle, while that of eq. (85) has turned to a stable node. For realistic nuclear spins, \( \Gamma^*_1 \ll \Gamma_2 \). Therefore, the considered region of \( g\zeta \) is very narrow. The relaxation rates are close to \( \lambda_2^+ \approx -\frac{1}{2}\Gamma^*_1 \); hence the spin motion is to be incoherent. The coherence intensity \( w_2^* \), given in eq. (85), although is not zero exactly, but, since \( \Gamma^*_1 \ll \Gamma_2 \) and \( g > 1 \), is very small, not much differing from zero.

Increasing the pumping further, so that

\[
g\zeta > 1 + \frac{\Gamma^*_1}{8\Gamma_2},
\]

changes the picture qualitatively. Then the fixed point (84) continues to be a saddle, since \( \lambda_1^+ < 0 \), \( \lambda_1^- > 0 \), but the fixed point (85) turns into a stable focus, as far as its characteristic exponents become complex,

\[
X_2^\pm = -\frac{\Gamma^*_1}{2} \pm i\omega_\infty, \\
\omega_\infty \equiv \frac{\Gamma^*_1}{2} \sqrt{8 \frac{\Gamma_2}{\Gamma^*_1} (g\zeta - 1) - 1}. 
\] (89)
This is the regime of pulsing spin superradiance, when there appears a long series of superradiant bursts lasting about the time $T_1^* = (\Gamma_1^*)^{-1}$. At the intermediate stage, this pulsing is not periodic, but becomes approximately periodic as time increases. The asymptotic frequency (89) defines the time interval $T_\infty \equiv 2\pi/\omega_\infty$ between superradiant pulses at the late stage, when the spin evolution is close to the stationary solution (85). This interpulse time for sufficiently strong pumping, with $g\zeta \gg 1$, is

$$T_\infty \equiv \frac{2\pi}{\omega_\infty} \simeq \pi \sqrt{\frac{2T_1^*T_2}{g\zeta}}. \quad (90)$$

The time (90), by varying the related parameters, can be varied widely. For example, keeping in mind the parameters typical of the proton spins$^{11-13}$, one has $\Gamma_0 = 2.85 \times 10^4$ Hz, $\omega_0 = 2.675 \times 10^8$ Hz, $\Gamma_2 = 1.18 \times 10^5$ Hz. With the quality factor $Q = 100$, the ringing width is $\gamma = 1.34 \times 10^6$ Hz. The spin-resonator coupling (67) is $g = 48$. For the pumping characteristics $\zeta = 1$ and $\Gamma_1^* = 10$ Hz, the interpulse time (90) is $T_\infty = 0.59 \times 10^{-3}$ s. The number of pulses can be estimated as $T_1^*/T_\infty \sim 100$.

A detailed description of the regime of pulsing spin superradiance, for arbitrary times $t > 0$, can be accomplished with the help of numerical calculations$^{17,22,24}$. The phenomenon of pulsing spin superradiance may be employed for creating pulsing spin masers$^{22}$.

A regime, similar to pulsing spin superradiance, can also be realized without dynamic nuclear polarization. For this purpose, one could change in the following way the experimental setup used for realizing the transient spin superradiance. In the latter regime, as is described in Section 7, if the spin-resonator coupling $g$ is sufficiently strong, a sharp superradiant burst occurs at the delay time $t_0$. After the time $t_0 + \tau_p$, the spin polarization, according to eq. (80), becomes practically inverted, provided $g \gg 1$. If at this moment, one inverts the static magnetic field $B_0$ or acts on spins by a resonant $\pi$-pulse, or just turns the sample $180^\circ$ about an axis perpendicular to $B_0$, then again a strongly nonequilibrium state of almost completely inverted spins is prepared. After the time $t_0$, counted from the moment when the newly inverted state is created, another superradiant burst will arise. Then, one could again either invert the static magnetic field or invert the magnetization by a resonant $\pi$-pulse, or turn the sample to arrange a novel nonequilibrium inverted state of spins. After this, one more superradiant burst will appear. Such a procedure can be repeated as many times as necessary for producing a required number of sharp superradiant pulses. The achieved regime may be named punctuated spin superradiance. The principal difference of this regime from the pulsing spin superradiance is the possibility of controlling the number of pulses as well as the interpulse time. The term punctuation here implies the possibility of varying the time intervals between pulses and to form groups of pulses, containing different numbers of the latter. In that way, it is feasible to compose a code, similar to the Morse alphabet. Hence, punctuated spin superradiance can be used for processing information, which may be employed, for instance, in quantum computers.
9 Hyperfine Interactions

Real solids, in addition to nuclei, always contain electrons. It is therefore important to understand the influence of hyperfine spin-spin interactions between nuclei and electrons on nuclear spin superradiance.

A system of nuclear and electronic spins is described by the Hamiltonian

$$\hat{H} = \hat{H}_{\text{nuc}} + \hat{H}_{\text{ele}} + \hat{H}_{\text{hyp}},$$

(91)

in which the nuclear term $\hat{H}_{\text{nuc}}$ is given by eq. (1). The electronic spin Hamiltonian is

$$\hat{H}_{\text{ele}} = -\frac{1}{2} \sum_{i \neq j} J_{ij} \hat{S}_i \cdot \hat{S}_j + \hbar \gamma_e \sum_i B \cdot \hat{S}_i,$$

(92)

where $J_{ij}$ is an exchange interaction potential, $\hat{S}_i$ is an electron spin, and $\gamma_e$ is the electron magnetogyric ratio. The part of the Hamiltonian (91) describing hyperfine interactions is

$$\hat{H}_{\text{hyp}} = A \sum_i \hat{S}_i \cdot \hat{I}_i + \frac{1}{2} \sum_{i \neq j} \sum_{\alpha \beta} A_{ij}^{\alpha \beta} \hat{S}_i^{\alpha} \hat{I}_j^{\beta}.$$  

(93)

Here the first term is the Fermi contact hyperfine interaction of nuclei with $s$-electrons, characterized by the energy

$$A = \frac{8\pi}{3} \hbar^2 \gamma_e \gamma_n |\psi(0)|^2,$$

(94)

with $\psi(r)$ being the electron wave function. The second term in eq. (93) presents the dipolar hyperfine interactions, with

$$A_{ij}^{\alpha \beta} = -\hbar^2 \frac{\gamma_e \gamma_n}{r_{ij}^3} \left( \delta_{\alpha \beta} - 3 n_{ij}^{\alpha} n_{ij}^{\beta} \right).$$  

(95)

The consideration of the problem with the Hamiltonian (91) can be accomplished by employing the same methods as have been detailed in the previous sections. The main technical difference is that the consideration becomes more cumbersome, as now it is necessary to deal with six evolution equations for spins, three of which are for nuclear spins and three other, for electronic spins. The seventh equation is the Kirchhoff equation (19), in which the magnetization density now is a sum

$$M_x = \frac{\hbar}{V} \sum_i \left( \gamma_n < \hat{I}_i^x > - \gamma_e < \hat{S}_i^x > \right)$$

(96)

of the terms due to nuclear and electronic spins. The evolution equations can be again solved by invoking the scale separation approach. The most important conclusions resulting from the presence of hyperfine interactions are as follows.

Local spin fluctuations of electrons lead to the increase of the nuclear dynamic broadening, which results in the sum

$$\tilde{\Gamma}_3 = \Gamma_3 + \Gamma'_3$$

(97)
of the widths caused by nuclear dipolar interactions, $\Gamma_3$, and by electron-nuclear hyperfine interactions, $\Gamma'_3$. The relation between these widths is given by the ratio
\[
\frac{\Gamma'_3}{\Gamma_3} = \frac{\rho_e \mu_e}{\rho_n \mu_n},
\] (98)
in which $\rho_e$ and $\rho_n$ are the electron and nuclear densities, while $\mu_e \equiv \hbar \gamma_e S$ and $\mu_n \equiv \hbar \gamma_n I$ are the electron and nuclear magnetic moments, respectively. When the electron and nuclear densities are close to each other, then, because $\mu_e / \mu_n \sim 10^3$, the ratio (98) can be large compared to unity. Therefore, the width (97) can be essentially increased by the hyperfine interactions. This results in the shortening of the delay time $t_0$ of a superradiant burst, as well as in shortening the pulse time $\tau_p$.

In the electronic subsystem, there may appear long-range magnetic order due to the exchange interaction of electronic spins. If so, the NMR frequency shifts to the value
\[
\omega_n = \omega_0 + \frac{A}{\hbar} S m_z,
\] (99)
where $m_z$ is the $z$-projection of the average electronic magnetization normalized to unity, $A$ being the parameter (94). Then, the frequency (99) will enter all formulas instead of $\omega_0$.

Long-range magnetic order of electrons leads to the renormalization of the coupling with the resonator according to the rule
\[
\tilde{g} = g \left( 1 + \frac{\rho_e \mu_e A}{\rho_n \mu_n \hbar \omega_n} m_z \right).
\] (100)
This may result because of the large ratio $\mu_e / \mu_n \sim 10^3$ in an essential increase of the coupling (100), as compared to $g$. The electronic subsystem plays the role of an additional resonator, which enhances the effective coupling of nuclear spins with the resonant electric circuit$^{24}$.

10 Radiation Intensity

To complete the treatment, it is worth emphasizing once again why, actually, the studied phenomenon can be called spin superradiance. An ensemble of coherently moving nuclear spins, as is clear, generates the magnetodipole radiation with the total intensity
\[
I(t) = \frac{2}{3c^3} \left| \tilde{M}(t) \right|^2,
\] (101)
where
\[
\tilde{M}(t) = \hbar \gamma_n \sum_i < I_i(t) >
\]
is the total magnetization of nuclei. The quantity of interest is the radiation intensity averaged over fast oscillations. For the intensity (101), this yields
\[
\overline{I}(t) = \frac{2}{3c^3} \mu_n^2 \omega_0^4 N^2 w(t),
\] (102)
where, as always, $\mu_n \equiv h\gamma_n I$ is the nuclear magnetic moment. As is seen, the radiation intensity is proportional to the number of spins squared, which is characteristic of superradiance.

Though coherently moving spins do produce superradiance, the radiation intensity (102) is not high. At the peak of a superradiant burst, $w(t_0)$ is given by eq. (79). For $g \gg 1$ and $s_0 \approx 1$, one has $w(t_0) \approx 1$. Then the maximal radiation intensity, for proton spins, with $\mu_n = 1.411 \times 10^{-26} J T^{-1}$, $\omega_0 = 2.675 \times 10^8$ Hz, and $N \approx 10^{23}$, is $\mathcal{T}(t_0) \approx 2.5 \times 10^{-5}$ W.

Despite so weak a radiation intensity, it can be easily detected. This is because what is directly measured is the power of current

$$P(t) = Rj^2(t),$$

which is generated in a coil by the radiating spins. Using the relation of the induced current with the resonator magnetic field (14), one gets

$$j^2(t) = \frac{V_c}{4\pi L} H^2(t).$$

The field $H$ can be found from eq. (41). Setting in this equation $\beta = 0$, i.e. neglecting thermal noise, and averaging over fast oscillations gives

$$\overline{H^2(t)} = \frac{2}{\gamma_n^2} \alpha^2(t) w(t).$$

Then, for the averaged current power (103), one finds

$$\overline{P(t)} = g\Gamma_2 I\hbar \omega N \left(1 - e^{-\gamma t}\right)^2 w(t).$$

(104)

This expression contains, as compared to eq. (102), an additional dependence on the feedback retardation. But for $t \gg \tau$, the radiation intensity (102) and the current power (104) differ one from another only by a numerical factor

$$\frac{\overline{P(t)}}{\overline{T(t)}} \simeq \frac{3g\Gamma_2 \lambda^3}{16\pi^2 \Gamma_0 V_c} = \frac{3Q\lambda^3}{8\pi^2 V_c},$$

(105)

in which $\lambda = 2\pi c/\omega$ is the radiation wavelength. For an NMR frequency $\omega = 2.675 \times 10^8$ Hz, one has $\lambda = 0.71 \times 10^3$ cm. The factor (105) can reach rather large values. Thus, if $Q = 100$ and $V_c = 10$ cm$^3$, this factor is of order $10^8$. This is why even a low radiation intensity can be easily measured.

11 Applications

Nuclear spin superradiance is collective spontaneous radiation by nuclear spins at a frequency of nuclear magnetic resonance. This phenomenon is an analog of atomic superradiance occurring at optical frequencies. Being a novel coherent phenomenon
at NMR frequencies, it may find various applications, among which it is possible to suggest the following:

1) **Investigation of materials characteristics**
   Spin relaxation in materials is commonly employed for studying relaxation parameters describing intrinsic properties of matter. Nuclear spin superradiance is accomplished by a self-organized coherent spin relaxation, which is drastically different from other types of spin relaxation. Another kind of relaxation may provide additional information on the properties of materials. Thus, the main mechanism originating pure spin superradiance is the existence of local spin fluctuations caused by the interaction of nuclear spins with each other, by means of dipolar forces, and with electronic spins, through hyperfine forces. Therefore, studying specific features of pure spin superradiance supplies information about these local fluctuations.

2) **Fast repolarization of targets**
   The relaxation of nuclear spins, in the process of superradiance, happens much faster than the usual spin-dephasing time $T_2$. If the initial spin polarization is sufficiently high and the spin-resonator coupling is sufficiently strong, spins, after a superradiant burst, change their orientation becoming almost completely inverted. Such an ultrafast inversion of spins can be employed for quick repolarization of polarized solid-state targets used in scattering experiments.

3) **Construction of spin masers**
   A superradiant spin system is a source of coherent radiation at radiofrequencies. Being a source of coherent radiation, it is analogous to lasers operating at optical frequencies. Spin masers could find applications similar to those of optical lasers. A spin maser can function in a transient regime, emitting a single burst typical of pure or triggered spin superradiance, and also it can work in a lasting regime based on pulsing spin superradiance.

4) **Creation of sensitive detectors**
   Spin superradiance can be triggered by a very weak external pulse of intensity corresponding to $w_0$. The latter depends exponentially on the delay time $t_0$. Therefore, measuring the delay time of a superradiant burst provides an accurate evaluation of the triggering intensity. Then, triggered spin superradiance may serve as a sensitive mechanism for detecting weak external signals.

5) **Methods of information processing**
   In the regime of punctuated superradiance, the intervals between superradiant bursts as well as the number of pulses can be regulated. This makes it feasible to compose a kind of the Morse Code alphabet and, respectively, to develop a technique of processing information. Such a method of information processing could be used in quantum computers.

In conclusion, it is necessary to note that the developed theory, as well as possible applications, are appropriate not only for nuclear spins but, in general, for any spin system. For instance, all this can immediately be extended to electronic spins. The
related phenomenon is *electron spin superradiance*. Other types of materials could be the so-called molecular magnets formed of magnetic molecules. The latter often have high spins, thus possessing several quantum transitions. In that case, the resonant electric circuit has to be tuned to one of the admissible transition frequencies. The resulting effect would be *molecular spin superradiance*, whose intensity could be much higher than that of nuclear spins. Nuclear spin superradiance is just a representative from a class of phenomena called *spin superradiance*. These phenomena, being realized with different materials, should exhibit a rich variety of specific features that could be not solely interesting but also useful for various applications.
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Figure Captions

**Fig. 1.** Typical experimental setup for detecting spin superradiance. The region surrounded by the dashed curve signifies a cryostat. Among the two coils below, the left one plays the role of antenna, and the right one is a part of the resonant electric circuit. The studied sample is shown as a dark bar inside the resonant coil. The upper left block is an oscilloscope, and the lower one is a plotter.

**Fig. 2.** Voltage signal corresponding to a superradiant pulse as a function of time, measured in units of $10^{-7}$ s, for two initial spin polarizations, $s_0 = 0.52$ (lower curve) and $s_0 = 0.57$ (upper curve).

**Fig. 3.** Orientation of the coordinate axes with respect to the sample that is inserted into the coil of a resonant electric circuit.
