Superradiant Operation of Spin Masers

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Abstract

The theory of spin superradiance, developed earlier for nuclear magnets, is generalized to a wider class of spin systems, such as granular magnets and molecular magnets. The latter may possess strong single-site magnetocrystalline anisotropy, whose role in nonlinear spin dynamics is analysed. Transient as well as pulsing superradiant regimes are described. These coherent regimes may be employed in the operation of spin masers.
1 Introduction

Among different masers, generating radiation at microwave and radio frequencies [1], there is a separate class of spin masers, in which the radiation process is due to moving spins. The role of a resonator for such masers is played by a resonant electric circuit coupled with a spin system. The peculiarity of spin masers is that their magnetodipole radiation is rather weak and practically does not propagate into free space but mainly is taken up by a resonant coil surrounding the spin sample [2]. Nevertheless, because of a deep physical similarity with other types of masers, radiating spin systems are called spin masers [3].

Resonant optical systems can display the effect of superradiance, which is a coherent spontaneous emission, when the radiation intensity is approximately proportional to the number of radiators squared [4,5]. Spin systems may also exhibit this phenomenon [6] which, because of its direct analogy with optical atomic superradiance, is termed spin superradiance. The latter, in the same way as the former, can be of two major types, transient and pulsing. The transient superradiance occurs as a single sharp superradiant burst that may be accompanied by several small quickly diminishing oscillations. This regime happens when the system is prepared in an inverted strongly nonequilibrium state, after which it is not influenced by any additional external fields. The pulsing superradiance corresponds to a long train of superradiant bursts, which can be realized if the system is subject to a permanent pumping mechanism. Both types of superradiance were experimentally observed for several spin systems, including the transient [7–10] and pulsing [11–13] spin superradiance. The theory of spin superradiance was developed [14–20], being based on microscopic Hamiltonians and allowing for the first correct description of purely self-organized superradiance. Numerical simulations of spin superradiance, being a kind of computer experiments, were also realized [21–27].

These studies of spin superradiance were based on the Hamiltonian describing an ensemble of polarized nuclear spins interacting through dipolar forces. Such a matter, for short, can be called a nuclear magnet. The influence of hyperfine forces on nuclear spin superradiance was also considered [28–30]. This attention to nuclear magnets has been due to the fact that the experiments with spin superradiance [7–13] had been accomplished for nuclear spins. In principle, this effect could be realized for electronic spins as well, with a similar theoretical description of electron spin superradiance [28,31,32].

In the present paper, the theory of spin superradiance is generalized to a wider class of materials, such as granular magnets and molecular magnets, which can possess higher spins and strong single-site magnetic anisotropy. The theory is also improved by taking a more accurate account of retardation effects.

2 Materials Characteristics

The experiments on spin superradiance till now have been accomplished with nuclear magnets. The first observation of transient spin superradiance [7,8] and its confirmation [9] were done for propanediol C₃H₈O₂. An active substance here is the ensemble of proton spins with
the density $\rho_H \sim 4 \times 10^{22} \text{ cm}^{-3}$. The spins were polarized, by means of dynamic nuclear polarization, parallel to an external magnetic field $B_0 \sim 1$ T, which corresponds to the Zeeman frequency $\omega_0 \sim 10^8$ Hz. The material was kept at low temperature $T \sim 0.1$ K, which resulted in strong suppression of the nuclear spin-lattice relaxation. The corresponding longitudinal relaxation time was $T_1 \sim 10^5$ s. The transverse dephasing time, due to dipolar interactions, was $T_2 \sim 10^{-5}$ s. The sample was coupled to a resonant electric circuit with a quality factor $Q \sim 100$ and a ringing time $\tau \sim 10^{-6}$ s.

In other experiments on transient spin superradiance [10] butanol C$_4$H$_9$OH and ammonia NH$_3$ were used. These materials are rich with protons of density $\rho_H \sim 10^{23}$ cm$^{-3}$. An external magnetic field $B_0 \sim 1$ T defined the Zeeman frequency $\omega_0 \sim 10^8$ Hz. The experiments were carried out at low temperature, resulting in the long spin-lattice relaxation time $T_1 \sim 4 \times 10^4$ s up to $10^5$ s. The transverse dephasing time was $T_2 \sim 10^{-5}$ s. The quality factor of the resonant electric circuit was $Q \sim 30$, and the ringing time $\tau \sim 5 \times 10^{-7}$ s.

The experiments on pulsing spin superradiance [11–13] employed the ruby crystal Al$_2$O$_3$. The active nuclei here are $^{27}$Al, with spin $I = 5/2$ and density $\rho_{Al} \sim 4 \times 10^{22}$ cm$^{-3}$. The crystal was oriented in an external magnetic field $B_0 \sim 1$T so that a fully resolved structure of its five $\Delta m = \pm 1$ transitions could be observed. Then, if a resonant circuit is tuned to a selected transition line, $^{27}$Al spins form an effective two-level system. In experiments, the circuit was tuned to the central $\{-\frac{5}{2}, \frac{5}{2}\}$ line, with a transition frequency $\omega_0 \sim 10^8$ Hz. At low temperatures $T \sim 1$ K, the spin-lattice relaxation time was $T_1 \sim 10^5$ s. The transverse relaxation time was $T_2 \sim 10^{-5}$ s. The quality factor of the resonant circuit was $Q \sim 100$, with the ringing time $\tau \sim 10^{-6}$ s. The inversion of spin polarization was permanently supported by means of dynamic nuclear polarization with the pumping rate $\gamma_1^* = 10$ s.

Wishing to extend the possibility of realizing spin superradiance for other types of spin systems, the first such materials that come to mind are granular magnets. These are composed of magnetic nanoparticles of diameters between 10 Å to 10$^4$ Å. Each nanoparticle is a superparamagnetic cluster of an effective spin $S$ that can be sufficiently high. There exists a large variety of magnetic nanoparticles [33,34] which can be formed by simple metals, such as Ni, Fe, Co, and Hg or their oxides, as NiO and Fe$_2$O$_3$. Many magnetic nanoparticles are made of different alloys, such as NiFe$_2$O$_4$, Nd$_2$Fe$_{14}$B, Pr$_2$Fe$_{14}$B, Tb$_2$Fe$_{14}$B, Dy$_2$Fe$_{14}$B, Pr$_2$Co$_{14}$B, Sm$_1$Fe$_{11}$Ti$_1$, Sm$_1$Fe$_{10}$V$_2$, Sm$_2$Fe$_{17}$N$_{23}$, Sm$_2$Fe$_{17}$C$_{22}$, Sm$_2$Co$_{17}$, and SmCo$_5$. An ensemble of magnetic nanoparticles could be polarized in an external magnetic field, after which, inverting the latter, one would get an inverted nonequilibrium system. The following process should be similar to that developing in an ensemble of nuclear spins. The main disadvantage of granular magnets is that composing them nanoparticles vary in size and shape. It is practically hardly possible to make a system of nanoparticles being almost identical. And an essential variation of the properties of radiating objects leads to a large inhomogeneous broadening, which hinders the feasibility of achieving a good level of coherence.

Another class of composite objects, possessing nonzero spin, are magnetic molecules [35]. These molecules can form crystalline materials where all magnetic clusters are well defined with the same shape, size, and orientation, because of which the inhomogeneous broadening has to be very low. Such materials, composed of magnetic molecules are termed molecular magnets. There are many different magnetic molecules [36,37] having in their
ground state nonzero total spins $S$. For example, the dodecanuclear manganese cluster $[\text{Mn}_{12}\text{O}_{12}(\text{CH}_3\text{COO})_{16}(\text{H}_2\text{O})_{4}]$·2CH$_3$COOH·4H$_2$O is a molecule with spin $S = 10$. It has a rather strong single-site anisotropy characterized by a parameter $D \approx 0.7$ K, which makes the anisotropy barrier $DS^2 \approx 70$ K. At low temperatures, lower than the blocking temperature $T_B \approx 3$ K, the magnetization of a molecular crystal is preserved during the relaxation time $\approx 10^7$ s. The size of a molecule is about 10 Å and the distance between neighbouring molecules in the molecular crystal is about 14 Å. The interaction between molecules is through dipolar forces, with an energy $\approx 0.1$ K or $10^{10}$ Hz. More information on the properties of this molecule can be found in Refs. [38–48].

The octanuclear iron molecular cluster $[\text{Fe}_8\text{O}_2(\text{OH})_{12}(\text{OH})_{12}(\text{tacn})_{6}]^{8+}$, where "tacn" stands for the organic ligand triazacyclononane, also has a high spin $S = 10$. Its magnetic anisotropy is $D \approx 0.3$ K, hence the anisotropy barrier is $DS^2 \approx 30$ K. The blocking temperature is $T_B \approx 1$ K, below which the relaxation time for a molecular crystal is about $10^5$ s. The size of a molecule and the intermolecular distance in a crystal are close to those for the molecule Mn$_{12}$ mentioned above. Other characteristics of the molecule can be found in Refs. [36,37,49–53].

The molecule $[\text{Cr(CNMnL)}_6](\text{ClO}_4)_9$, where L is a neutral pentadentate ligand, has in its ground state the total spin $S = 27/2$ (see [37]). The molecule $[(\text{PhSiO}_2)_6\text{Cu}_6(\text{O}_2\text{SiPh})_6]$ possesses the spin $S = 3$ (see [54]). And the molecule $K_6[V^{4+}_{13}\text{As}_6\text{O}_{42}(\text{H}_2\text{O})]·8\text{H}_2\text{O}$ has the spin $S = 1/2$ and no magnetic anisotropy [55,56]. There is a number of other molecules [37] with nonzero total ground-state spin. Some molecules may have zero spin in their ground state but a finite spin in excited states [57–59].

The relaxation of the total magnetization of a crystal, consisting of many magnetic molecules, occurs because of the axial degeneracy of spin direction in each molecule. At zero external magnetic field, the spin of a single molecule can be directed either up or down, so that the equilibrium state of an ensemble of these molecules corresponds to zero total magnetization. At temperatures higher than the blocking temperature $T_B$, the relaxation is rather fast and is due to thermal fluctuations. At low temperatures below $T_B$, the relaxation is very slow, so that the polarization of a molecular magnet can be blocked for months. At such low temperatures, the relaxation is characterized as quantum spin tunneling between the degenerate states. To understand the low-temperature behaviour of magnetic molecules, it is necessary to take into account both the internal effects, caused by atoms composing each molecule [60–62], as well as external interactions between the molecules forming a crystal [63–65]. Taking account of dipole interactions between molecules is crucial for correctly describing the relaxation in a molecular magnet [42,51,52,56,66].

In this way, there exists a large variety of different objects, such as nuclei, granules, and molecules, each of which can be considered as an entity, like a particle with an effective spin that can vary in a wide diapason. The main interactions between such magnetic particles, forming a solid, are dipole interactions. A sample, composed of these particles, can be polarized and at low temperatures the polarization can be preserved for a very long time. A specific feature of magnetic particles with high spin is the presence of the single-site magnetic anisotropy, which one has to take into account when considering collective spin dynamics. The problem, to be addressed in the following sections, is how coherent spin radiation can
arise is such materials made of objects with high spins and magnetic anisotropy. A special attention will be paid to the possibility of realizing spin superradiance that is a self-organized process developing without imposing on the spin system an initial transverse coherence.

3 Types of Coherence

Before passing to the development of a generalized theory of spin superradiance, a few words are to be said for concretizing the term "coherence" that will be repeatedly used in what follows. Generally, one uses this term in two different meanings. One widespread usage implies under a coherent state of a many-particle system just a pure quantum state characterized by the same wave function for all particles. Then an incoherent state is a mixed state described by a density matrix [67]. In the theory of nuclear magnetic resonance [68,69], coherence usually means the existence of transverse magnetization and, more generally, the existence of non-diagonal matrix elements. One also tells [68,69] that the existence of the transverse magnetization means phase coherence, as opposed to amplitude coherence associated with a nonzero longitudinal polarization. To formalize these definitions for a system of \( N \) spins, let us introduce the notation

\[
S^z \equiv \frac{1}{N} \sum_{i=1}^{N} S^z_i, \quad S^{\pm} \equiv \frac{1}{N} \sum_{i=1}^{N} S^{\pm}_i,
\]

where \( S^z_i \) is the \( z \)-component of the spin operator and \( S^{\pm}_i \) are the raising and, respectively, lowering spin operators. Denote the statistical averaging by the angle brackets \( < \ldots > \). Then a nonzero \( < S^z > \neq 0 \) means longitudinal coherence, or diagonal coherence, or amplitude coherence. While a nonzero \( < S^{\pm} > \neq 0 \) signifies the existence of transverse coherence, or nondiagonal coherence, or phase coherence.

Another possibility could be to tell that a nonzero \( < S^z > \neq 0 \) corresponds to state coherence. When the whole system is in pure state, then \( < S^z > = \pm S \), which can be named pure coherent state, to distinguish it from the case of partially coherent state, when \( 0 < |< S^z >| < S \). This definition of state coherence is in agreement with the quantum-mechanical understanding of a coherent state as of a pure state. It also agrees with the definition of coherent states in quantum field theory as of eigenvalues of field operators [70]. In many applications, the operator \( S^z \) characterizes a population difference. Hence, nonzero \( < S^z > \) may be associated with population coherence. Being related to the determination of eigenfunctions of operators, the state coherence may also be called quantum coherence.

Since the raising and lowering operators describe transitions between quantum states, the existence of nonzero \( < S^{\pm} > \neq 0 \) can be ascribed to transition coherence. This type of coherence is closely connected with the existence of coherent radiation, because of which it may be termed radiation coherence. Also, this recalls the classical understanding of coherence as of synchronous motion of several objects, which makes it sometimes possible to use the name of classical coherence. Synchronous motion is often termed as the motion in phase. That is why the term of phase coherence is appropriate here.

These types of coherence are not strictly correlated with each other. A spin system may possess one of them or both, or neither. But often they are complimentary to each other, as
it happens in the process of superradiance. Then, at the initial time, the spin system has to be prepared in a well polarized state, thus, displaying state coherence. In addition, this state must be strongly nonequilibrium. Transition coherence, at the initial time, should be absent. It has to develop in a self-organized way owing to mutual spin correlations, which can be realized through a feedback field. Usually, the maximal transition coherence in superradiance develops at the moment when the state coherence is minimal.

4 Spin Hamiltonian

Consider an ensemble of \( N \) spins \( S_i \) enumerated by the index \( i = 1, 2, \ldots, N \). These spins can correspond either to nuclei, or to granules, or to molecules. The Hamiltonian of the system,

\[
H = \sum_i H_i + \frac{1}{2} \sum_{i \neq j} H_{ij},
\]

contains the terms \( H_i \), related to individual spins, and the terms \( H_{ij} \) describing spin interactions. An individual term

\[
H_i = -\mu_0 B \cdot S_i - D(S_i^z)^2
\]

includes the Zeeman energy and a part characterizing magnetic anisotropy. In the case of nuclei, \( \mu_0 = g_S \mu_N = \hbar \gamma_S \), where \( g_S \) is the Landé factor; \( \mu_N \), nuclear magneton; \( \gamma_S \), gyromagnetic ratio. For nuclei, \( \mu_0 \) can be positive as well as negative. When \( S \) is an electron spin, \( \mu_0 = -g_S \mu_B = -\hbar \gamma_S \), where \( \mu_B \) is the Bohr magneton. Then \( \mu_0 \) is negative. The anisotropy term, with an anisotropy constant \( D \), is nontrivial only for spins higher than \( 1/2 \). Positive \( D > 0 \) implies an easy-axis anisotropy, while \( D < 0 \) means an easy-plane anisotropy. The pair terms in the Hamiltonian (1) describe dipolar interactions

\[
H_{ij} = \sum_{\alpha \beta} C_{ij}^{\alpha \beta} S_i^\alpha S_j^\beta
\]

through the dipolar tensor

\[
C_{ij}^{\alpha \beta} = \frac{\mu_0^2}{r_{ij}^3} \left( \delta_{\alpha \beta} - 3n_i^\alpha n_j^\beta \right),
\]

in which \( \alpha, \beta = x, y, z \) and

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

The dipolar tensor enjoys the properties

\[
\sum_\alpha C_{ij}^{\alpha \alpha} = 0, \quad \sum_{j(\neq i)} C_{ij}^{\alpha \beta} = 0,
\]

of which the first is exact and the second is asymptotically exact for a macroscopic sample with a large number of spins \( N \gg 1 \). The total magnetic field

\[
\mathbf{B} = B_0 \mathbf{e}_z + (B_1 + H) \mathbf{e}_x
\]
consists of an external longitudinal magnetic field $B_0$, transverse magnetic field $B_1$, and a feedback field $H$ of the resonant electric circuit. In what follows, the longitudinal magnetic field is assumed to be constant and directed so that

$$\mu_0 B_0 < 0 .$$

(7)

In particular, for electronic spins, $B_0 > 0$, since $\mu_0$ is negative.

For what follows, it is convenient to pass to raising and lowering operators

$$S_i^\pm \equiv S_i^x \pm i S_i^y ,$$

which are Hermitian conjugated with each other. Also, introduce the notation

$$a_{ij} \equiv C^{zz}_{ij} , \quad b_{ij} \equiv \frac{1}{2} \left( C^{xx}_{ij} + i C^{yy}_{ij} \right) ,$$
$$c_{ij} \equiv \frac{1}{4} \left( C^{xx}_{ij} + 2i C^{xy}_{ij} - C^{yy}_{ij} \right) .$$

(8)

Then the individual term (2) can be written as

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

(9)

And the interaction term (3) takes the form

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

(10)

Thus, the spin system is described by the Hamiltonian (1), with the terms (9) and (10).

5 Electric Circuit

The external fields $B_0$ and $B_1$ are assumed to be given. What is not yet defined is the resonator feedback field $H$, produced by a resonant electric circuit. Let the circuit be characterized by resistance $R$, inductance $L$, and capacity $C$. The spin sample is inserted into a coil of $n$ turns, length $l$ and cross-section area $A_c$. The electric current in the circuit is determined 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 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

$$M_x = \frac{\mu_0}{V} \sum_i < S_i^x > ,$$

(13)

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

The current, circulating over the coil, produces a magnetic field

$$H = \frac{4\pi n}{c l} j ,$$

(14)

with $c$ being the light velocity. Hence, Eq. (11) may be rewritten for the field (14). Introduce the circuit natural frequency

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

(15)

and the circuit ringing time

$$\tau \equiv \frac{1}{\gamma} \quad \left( \gamma \equiv \frac{R}{2L} \right) ,$$

(16)

with the related circuit damping

$$\gamma = \frac{\omega}{2Q} \quad \left( Q \equiv \frac{\omega L}{R} \right) ,$$

(17)

where $Q$ is the quality factor. Define the reduced electromotive force

$$h \equiv \frac{c \bar{E}}{n A_c \gamma} .$$

(18)

Then the Kirchhoff equation (11) can be transformed to the equation

$$\frac{dH}{dt} + 2\gamma H + \omega^2 \int_0^t H(t') \, dt' = \gamma h - 4\pi \eta \frac{dM_x}{dt}$$

(19)

for the feedback magnetic field produced by the coil.

The feedback equation (19) can be presented in another equivalent form [15–18] which is very useful for solving the evolution equations. For this purpose, we apply to Eq. (19) the method of Laplace transforms and use the transfer function

$$G(t) = \left( \cos \tilde{\omega}t - \frac{\gamma}{\tilde{\omega}} \sin \tilde{\omega}t \right) e^{-\gamma t} ,$$

with $\tilde{\omega} \equiv \sqrt{\omega^2 - \gamma^2}$. Then from the feedback equation (19) we obtain

$$H = \int_0^t G(t - t') \left[ \gamma h(t') - 4\pi \eta \dot{M}_x(t') \right] \, dt' ,$$

(20)

where $\dot{M}_x \equiv dM_x/dt$. 

8
6 Evolution Equations

First, we write the Heisenberg equations

\[
\frac{i\hbar}{\alpha} \frac{dS_{i\alpha}}{dt} = [S_{i\alpha}, \hat{H}]
\]

for the spin operators, using the commutation relations

\[
[S_{i+}, S_{j-}] = -2\delta_{ij} S_{i}^{z}, \quad [S_{i-}, S_{j+}] = \pm \delta_{ij} S_{i}^{z}, \quad [S_{i+}, (S_{j}^{z})^{2}] = \delta_{ij} \left( S_{i-} S_{i}^{z} + S_{i}^{z} S_{i-} \right).
\]

For the lowering operator, this gives

\[
\frac{i\hbar}{\alpha} \frac{dS_{i-}}{dt} = -\mu_0 B_0 S_{i}^{-} + \mu_0 (B_1 + H) S_{i}^{z} - D (S_{i}^{-} S_{i}^{z} + S_{i}^{z} S_{i}^{-}) +
\]

\[
+ \sum_{j(\neq i)} \left[ a_{ij} \left( S_{i}^{-} S_{j}^{z} + \frac{1}{2} S_{i}^{z} S_{j}^{-} \right) + b_{ij} S_{i}^{-} S_{j}^{-} + b_{ij}^{*} \left( S_{i}^{-} S_{j}^{z} - 2 S_{i}^{z} S_{j}^{z} \right) - 2 c_{ij}^{*} S_{i}^{z} S_{j}^{z} \right].
\] (21)

The equation for the raising operator is obtained from Eq. (21) by Hermitian conjugation. The equation for \( S_{i}^{z} \) is

\[
\frac{i\hbar}{\alpha} \frac{dS_{i}^{z}}{dt} = \frac{1}{2} \mu_0 (B_1 + H) \left( S_{i}^{-} - S_{i}^{+} \right) +
\]

\[
+ \sum_{j(\neq i)} \left[ \frac{1}{4} a_{ij} \left( S_{i}^{-} S_{j}^{z} - S_{i}^{z} S_{j}^{-} \right) + b_{ij}^{*} S_{i}^{z} S_{j}^{z} - b_{ij} S_{i}^{z} S_{j}^{-} + c_{ij}^{*} S_{i}^{z} S_{j}^{z} - c_{ij} S_{i}^{z} S_{j}^{-} \right].
\] (22)

To analyze these equations, we employ the scale separation approach [14,17,18,71]. Notice, first, that in Eqs. (21) and (22), we may separate the terms

\[
\xi_0 \equiv \frac{1}{\hbar} \sum_{j(\neq i)} \left( a_{ij} S_{j}^{z} + b_{ij}^{*} S_{j}^{z} + b_{ij} S_{j}^{-} \right),
\]

\[
\xi \equiv -\frac{i}{\hbar} \sum_{j(\neq i)} \left( \frac{1}{2} a_{ij} S_{j}^{-} - 2 b_{ij}^{*} S_{j}^{z} - 2 c_{ij}^{*} S_{j}^{z} \right),
\] (23)

which play the role of local fields acting on spins. Statistical averages of these fields, with the usage of the uniform approximation, are zero owing to the equalities

\[
\sum_{j(\neq i)} a_{ij} = \sum_{j(\neq i)} b_{ij} = \sum_{j(\neq i)} c_{ij} = 0
\]

following from the properties (5). At the same time, statistical averages of the spin operators are certainly nonzero. Therefore the local fields (23), acting on a short scale, can be treated as operator variables of nature different from the spin operators. Such local fields may be modelled by random variables [68,69,72]. Thus, we have two types of variables in the system, spin operators \( S_{i}^{-}, S_{i}^{+}, S_{i}^{z} \), and stochastic fields \( \xi_0, \xi, \xi^{*} \). The former are responsible for long-range global phenomena while the latter, for short-range local effects. The stochastic
fields describe fast fluctuations in the local surrounding of each spin. The existence of such fluctuations yields inhomogeneous dynamic broadening.

To make the problem closed, it is necessary to define the stochastic averages over the random fields (23). The latter can be treated as white noise with the stochastic averages

\[ \langle \xi_0(t) \rangle = 0, \quad \langle \xi(t) \rangle = 0, \quad \langle \xi_0(t) \xi_0(t') \rangle = 2 \gamma_3 \delta(t - t'), \]
\[ \langle \xi(t) \xi(t') \rangle = 2 \gamma_3 \delta(t - t'), \quad \langle \xi^*(t) \xi(t') \rangle = 2 \gamma_3 \delta(t - t'), \] (24)

where \( \gamma_3 \) is the width of dynamic broadening. The method of modelling local fields by random variables can be called randomization of local fields.

Averaging over spin operators, which are responsible for long-range phenomena, we may employ the mean-field approximation

\[ \langle S^\alpha_i S^\beta_j \rangle = \langle S^\alpha_i \rangle \langle S^\beta_j \rangle \quad (i \neq j). \] (25)

This kind of approximation can be used only for the pairs of spins at different sites \( i \neq j \), since \( S^\alpha_i \) and \( S^\beta_j \) commute in such a case. But the spin operators do not commute for \( i = j \). Hence the averages \( \langle S^\alpha_i S^\beta_i \rangle \) cannot be factorized as above. Notice that for spin \( S = 1/2 \), the anisotropy term in the Hamiltonian does not contribute to the equations of motion, since \( (S^z_i)^2 = 1/4 \). The problem of spin decoupling, more general than condition (25), has been considered by several authors [73–76]. The term in Eq. (21), caused by the magnetic anisotropy, can be decoupled as follows:

\[ \langle S^z_i S^z_i \rangle = \left( 2 - \frac{1}{S} \right) \langle S^z_i \rangle \langle S^z_i \rangle. \] (26)

This presentation enjoys correct limiting properties. For \( S = 1/2 \), it nullifies; while for \( S \to \infty \), when spins behave classically, a simple factorization occurs. The decouplings (25) and (26) do not take account of spin-spin correlations, which can be incorporated into the evolution equations by including the term describing spin attenuation, with the related spin-spin relaxation width \( \gamma_2 \). To allow for the influence of lattice, account must be taken of the spin-lattice relaxation, with the corresponding relaxation parameter \( \gamma_1 \).

Let us average the equations of motion (21) and (22) over the spin operators, not touching the stochastic fields (23). In so doing, we introduce the following definitions. The variable

\[ x \equiv \frac{1}{S} < S^-_i > \] (27)

describes the rotation of transverse spin components. As is discussed in Section 3, this average is connected with the arising transition coherence, or transverse coherence, or radiation coherence. The degree of such a coherence can be characterized by the real function

\[ y \equiv \frac{1}{S^2} < S^+_i > < S^-_i > = |x|^2. \] (28)

And the longitudinal spin polarization is given by

\[ z \equiv \frac{1}{S} < S^z_i >. \] (29)
Recall that the radiation of spins happens at radio-frequencies whose wavelengths are much larger than the mean distance between spins. Therefore, it is admissible to employ the uniform approximation, assuming that the functions (27) to (29) do not depend on spatial variables.

Note that, instead of resorting to the uniform approximation for the functions (27) to (29), it would be possible to work with the following arithmetical averages: transition function

\[ x \equiv \frac{1}{NS} \sum_{i=1}^{N} < S_i^- > , \]

coherence intensity

\[ y \equiv \frac{1}{N^2 S^2} \sum_{i \neq j}^{N} < S_i^+ S_j^- > , \]

and longitudinal polarization, or spin polarization

\[ z \equiv \frac{1}{NS} \sum_{i=1}^{N} < S_i^z > . \]

The resulting equations would be absolutely the same.

Let us define the Zeeman frequency

\[ \omega_0 \equiv \frac{1}{\hbar} |\mu_0 B_0| , \] (30)

the effective transition frequency

\[ \bar{\omega}_0 \equiv \omega_0 - (2S - 1)Dz , \] (31)

and introduce the notation

\[ f \equiv - \frac{i}{\hbar} \mu_0 (B_1 + H) + \xi \] (32)

for an effective force acting on spins. Then from Eqs. (21) and (22), we obtain the evolution equations for the functional variables (27) to (29):

\[ \frac{dx}{dt} = -i (\bar{\omega}_0 + \xi_0 - i\gamma_2) x + f z , \] (33)

\[ \frac{dy}{dt} = -2\gamma_2 y (x^* f + f^* x) z , \] (34)

\[ \frac{dz}{dt} = -\frac{1}{2} (x^* f + f^* x) - \gamma_1 (z - \sigma) . \] (35)

These equations are assumed to be complimented by the initial conditions

\[ x(0) = x_0 , \quad y(0) = y_0 , \quad z(0) = z_0 . \]
Equations (33) to (35) are stochastic differential equations, since they contain the stochastic variables $\xi_0$ and $\xi$. Getting stochastic equations is the price for making them closed. These also are nonlinear equations because of the resonator feedback field entering through the effective force (32). This resonator field is given by Eq. (20), where one has to substitute the magnetization density

$$M_x = \frac{1}{2} \rho \mu_0 S (x^* + x) \quad (\rho \equiv \frac{N}{V}).$$

Due to the integral form (20), Eqs. (33) to (35) plus (20) compose a system of stochastic nonlinear integro-differential equations.

## 7 Stochastic Averaging

The system of equations (33) to (35) plus (20) looks rather complicated. Nevertheless, it can be essentially simplified by invoking the method of stochastic averaging [17,18,71], which is a generalization of multiscale averaging techniques to stochastic differential equations. The applicability of the method is due to the existence of several small parameters.

The spin-lattice and spin-spin relaxation parameters are assumed to be small as compared to the Zeeman frequency,

$$\frac{\gamma_1}{\omega_0} \ll 1, \quad \frac{\gamma_2}{\omega_0} \ll 1. \quad (36)$$

The interaction energy of spins with the resonator field is proportional to the natural width

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

Since $\gamma_2 \sim n_0 \rho \mu_0^2 S^2 / \hbar$, where $n_0$ is the number of nearest neighbors, then

$$\gamma_0 \sim \frac{\pi \eta}{n_0 S} \gamma_2 < \gamma_2.$$

Hence, the natural width (37) is small, as well as the width of dynamic broadening,

$$\frac{\gamma_0}{\omega_0} \ll 1, \quad \frac{\gamma_3}{\omega_0} \ll 1. \quad (38)$$

Here we do not explicitly consider hyperfine interactions, surmising that their influence can be included in the values of the related relaxation parameters. In order to estimate the impact of these interactions, e.g. on the magnitude of the dynamic broadening, it is necessary to discriminate the cases when the radiating objects are nuclear spins or electronic spins. Recall that the spins of magnetic molecules are of electronic nature. The dynamic broadening of electronic spins, caused by dipolar and hyperfine interactions, respectively, is $\gamma_3 \sim \rho_e \mu_e^2$ and $\gamma'_3 \sim \rho_n \mu_n \mu_e$, where $\rho_e$ and $\rho_n$ are the densities of electronic or nuclear spins, with $\mu_e \equiv g_S \mu_B S$ and $\mu_n \equiv g_I \mu_N I$ being the electronic and nuclear moments. Similarly, denoting by capital letters the relaxation parameters for nuclear spins, we have the dynamic...
broadening, due to dipolar or hyperfine interactions as $\Gamma_3 \sim \rho_n \mu_n^2$ and $\Gamma_3' \sim \rho_e \mu_e \mu_n$. From here, the following relations are valid:

$$\frac{\gamma'_3}{\gamma_3} \sim \frac{\rho_n \mu_n}{\rho_e \mu_e}$$

Taking into account the equalities

$$\frac{\mu_e}{\mu_n} = \frac{gs \mu_B S}{g_1 \mu_N I}, \quad \frac{\mu_B}{\mu_N} = \frac{m_p}{m_e} = 1836,$$

we see that the hyperfine interactions do not play an important role for electronic spins but can be rather important in the case of nuclear spins [28–30].

The external transverse magnetic field is taken in the form

$$B_1 = h_0 + h_1 \cos \omega t .$$  \hspace{1cm} (39)

And let the resonant part of the reduced electromotive force (18) be presented as

$$h = h_c \cos \omega t .$$  \hspace{1cm} (40)

Introduce the notation

$$\nu_0 \equiv \frac{\mu_0 h_0}{h}, \quad \nu_1 \equiv \frac{\mu_0 h_1}{2h}, \quad \nu_c \equiv \frac{\mu_0 h_c}{2h} .$$  \hspace{1cm} (41)

The amplitudes of the fields (39) and (40) are supposed to be small, in the sense that

$$\left| \frac{\nu_0}{\omega_0} \right| \ll 1, \quad \left| \frac{\nu_1}{\omega_0} \right| \ll 1, \quad \left| \frac{\nu_c}{\omega_0} \right| \ll 1 .$$  \hspace{1cm} (42)

The influence of stochastic fields has to be considered as weak, since the stochastic averages (24) are proportional to the dynamic broadening width that, according to Eq. (38), is small. In this way, the effective force (32) can be treated as weak.

Finally, the resonant circuit is assumed to be of good quality, implying that

$$\frac{\gamma}{\omega} \ll 1 \quad (Q \gg 1) .$$  \hspace{1cm} (43)

The existence of the listed small parameters shows that the transition function $x$, defined by Eq. (33), is to be considered as fast, compared to the slow functions $y$ and $z$, satisfying Eqs. (34) and (35). Conversely, $y$ and $z$ are temporal quasi-invariants with respect to $x$.

The resonator field $H$, in the first approximation, may be found by iterating Eq. (20) with the solution of Eq. (33) of zero order with respect to small parameters, that is with $x \simeq x_0 \exp(-i\tilde{\omega}t)$, where $z$ is a quasi-invariant. This iteration yields

$$\frac{\mu_0 H}{h} = i(\alpha x - \alpha^* x^*) + 2\beta \cos \omega t ,$$  \hspace{1cm} (44)
where $\alpha$ is the coupling function of spins with the resonator feedback field,

$$\alpha = \gamma_0 \tilde{\omega}_0 \left[ 1 - \exp \{-i(\omega - \tilde{\omega}_0)t - \gamma t\} \right] + \gamma_0 \tilde{\omega}_0 \left[ 1 - \exp \{i(\omega + \tilde{\omega}_0)t - \gamma t\} \right] , \tag{45}$$

and $\beta$ is the coupling function of spins with the electromotive force,

$$\beta = \nu_c \left[ 1 - e^{-\gamma t} \right] . \tag{46}$$

Clearly, the action of the feedback field can be efficient only in the case of a resonant coupling, which requires the resonance condition

$$\frac{\tilde{\Delta}}{\omega} \ll 1 , \quad \tilde{\Delta} \equiv \omega - |\tilde{\omega}_0| . \tag{47}$$

Note that the effective frequency (31) can, in general, be positive as well as negative. The spin-feedback coupling (45) simplifies if the resonance is good, which means that $|\tilde{\Delta}| < \gamma$. In such a case, Eq. (45) acquires the simple form

$$\alpha \approx \frac{\gamma_0 \tilde{\omega}_0}{\gamma^2 + \Delta^2} \left( 1 - e^{-\gamma t} \right) . \tag{48}$$

More generally, the coupling function (45) is complex, with its real and imaginary parts being

$$\text{Re} \alpha = \frac{\gamma_0 \tilde{\omega}_0}{\gamma^2 + \Delta^2} \left[ 1 - \left( \cos \tilde{\Delta} t - \frac{\Delta}{\gamma} \sin \tilde{\Delta} t \right) e^{-\gamma t} \right] ,$$

$$\text{Im} \alpha = -\frac{\gamma_0 |\tilde{\omega}_0|}{\gamma^2 + \Delta^2} \left[ \frac{\Delta}{\gamma} - \left( \sin \tilde{\Delta} t + \frac{\Delta}{\gamma} \cos \tilde{\Delta} t \right) e^{-\gamma t} \right] ,$$

where the resonance condition (47) is used.

Expression (44) is to be substituted into Eqs. (33) to (35). With this in mind, we define the collective frequency

$$\Omega \equiv \tilde{\omega}_0 - z \text{Im} \alpha \tag{49}$$

and the collective attenuation

$$\Gamma \equiv \gamma_2 - z \text{Re} \alpha . \tag{50}$$

These quantities depend on time through the slow variables $z$ and $\alpha$, because of which one may say that the frequency and attenuation experience dynamic shift. In the case of good resonance, when $\alpha$ is given by Eq. (48), expressions (49) and (50) are

$$\Omega = \tilde{\omega}_0 = \omega_0 - (2S - 1)Dz , \quad \Gamma = \gamma_2 - \alpha z . \tag{51}$$

Also, we define the effective force

$$f_1 \equiv -i \nu_0 - 2i(\nu_1 + \beta) \cos \omega t + \xi . \tag{52}$$
Then Eqs. (33) to (35) rearrange to

\[ \frac{dx}{dt} = -i(\Omega + \xi_0 - i\Gamma)x + f_1z - \alpha zx^* , \]  
\[ \frac{dy}{dt} = -2\Gamma y + (x^* f_1 + f_1^* x)z - \alpha z \left[(x^*)^2 + x^2\right] , \]  
\[ \frac{dz}{dt} = -\alpha y - \frac{1}{2}(x^* f_1 + f_1^* x) - \gamma_1(z - \sigma) + \frac{1}{2} \alpha \left[(x^*)^2 + x^2\right] . \]

The time derivatives of \( \alpha \) and \( \beta \) are proportional to \( \gamma \), because of which these functions are to be treated as slow, compared to \( x \). Hence, \( \alpha \) and \( \beta \), as well as \( \Omega \) and \( \Gamma \), are temporal quasi-invariants with respect to \( x \).

The solution to Eq. (53), with the quasi-invariants kept fixed, reads

\[ x = x_0 e^{-i(\Omega + \Gamma)t} \exp \left\{ -i \int_0^t \xi_0(t') dt' \right\} + 
+ z \int_0^t f_1(t') e^{-i(\Omega + \Gamma)(t-t')} \exp \left\{ -i \int_{t'}^t \xi_0(t'') dt'' \right\} dt' . \]  

The counterrotating term of Eq. (53), containing \( x^* \), gives to the form (56) a small addition of order \( \gamma_2 / \omega_0 \), because of which the latter is omitted.

Define the quantities

\[ \tilde{\Gamma} \equiv \Gamma + \gamma_3 \, , \quad \delta \equiv \omega - |\Omega| \, , \]  

which in the case of good resonance are

\[ \tilde{\Gamma} \approx \gamma_2 - \alpha z + \gamma_3 \, , \quad \delta \approx \omega - |\bar{\omega}_0| = \bar{\Delta} \, . \]

Averaging Eq. (56) over the stochastic variable \( \xi_0 \), we get

\[ \langle x \rangle = -\frac{\nu_0 z}{\Omega - i\tilde{\Gamma}} + \left( \frac{\nu_1 + \beta}{\delta + i\tilde{\Gamma}} \right) e^{-i\omega t} + \left[ x_0 + \int \frac{\nu_0 z}{\Omega - i\tilde{\Gamma}} - \left. \left( \frac{\nu_1 + \beta}{\delta + i\tilde{\Gamma}} \right) \right| e^{-i(\Omega + \Gamma)t} \right] . \]

The fast solution (56) has to be substituted into Eqs. (54) and (55) for the slow variables, whose right-hand sides are to be averaged over stochastic fields and over time in the infinite interval. Take the initial condition for the transition function \( x \) in the real form

\[ x_0 = \frac{1}{S} < S^x_t(0) > , \]

which is not principal but just slightly simplifies the following formulas. Also, keep in mind that the collective attenuation is small, compared to the collective frequency,

\[ \left| \frac{\Gamma}{\Omega} \right| \ll 1 \, . \]
From here, since $\gamma_3 \ll \omega_0$, it follows that $|\tilde{\Gamma}| \ll |\Omega|$. And let us introduce the effective attenuation

$$J \equiv \gamma_3 - \alpha \frac{\nu_1}{\Omega^2} z - \frac{\nu_0 (\nu_1 + \beta) \tilde{\Gamma}}{\Omega^2} e^{-\tilde{\Gamma}t} + \frac{(\nu_1 + \beta)^2 \tilde{\Gamma}}{\delta^2 + \tilde{\Gamma}^2} (1 - e^{-\tilde{\Gamma}t}) .$$

(58)

In the latter, the terms related to the action of the constant transverse field are of second and third order in small parameters. Omitting these terms gives

$$J = \gamma_3 + \frac{(\nu_1 + \beta)^2 \tilde{\Gamma}}{\Gamma^2 + \delta^2} (1 - e^{-\tilde{\Gamma}t}) .$$

Averaging the right-hand sides of Eqs. (54) and (55) over the stochastic fields and time, we come to the evolution equations

$$\frac{dy}{dt} = -2(\gamma_2 - \alpha z)y + 2Jz^2 ,$$

(59)

$$\frac{dz}{dt} = -\alpha y - Jz - \gamma_1 (z - \sigma) ,$$

(60)

whose solutions are called the guiding centers.

As is evident, the derived equations (59) and (60) for the guiding centers are incomparably easier to analyse than the initial equations (33) to (35). Since Eqs. (59) and (60) are nonlinear, their solutions can display rather nontrivial behaviour, especially when the resonant transverse field or electromotive force are present. Spin dynamics, in the presence of such transverse injected fields is known to be quite complicated [13,77,78]. But even when external fields are absent, the nonlinear spin dynamics may demonstrate very interesting effects.

Before analysing the evolution equations (59) and (60), let us pay attention to the physical conditions under which these equations have been obtained. The principal point here is the resonance condition (47). It is only when the spin system is in resonance with the coupled electric circuit that we could expect the appearance of noticeable transition coherence, which would develop in a self-organized way. This coherence, of course, can be induced by external transverse fields. However recall that our major aim is to consider superradiant regimes when the transition coherence and, respectively, radiation coherence, arises spontaneously, without being stimulated by intensive external fields.

There can be several possibilities for realizing the resonance condition (47). First of all, for systems with spin one-half, there is no magnetic anisotropy so that the effective transition frequency (31) coincides with the Zeeman frequency, $\tilde{\omega}_0 = \omega_0$. Then we have the same situation as for nuclear magnets [14–20] with $S = 1/2$.

If we are dealing with higher spins, then, nevertheless, there is the possibility of reducing the problem to an effective spin-one-half system. This can be done by tuning the resonant electric circuit to one of the transition frequencies of admissible $2S$ transitions and by supporting the population of the upper level with the help of a permanent nonresonant pumping, as it was accomplished for the nuclear spin $I = 5/2$ of $^{27}$Al in experiments [11–13]. In that case solely the regime of pulsing spin superradiance can be achieved.
For higher spins, the transition from the state with the spin projection $S$ to that with the projection $-S$ corresponds to a multiphoton transition through $2S - 1$ intermediate levels. If the longitudinal external magnetic field is sufficiently strong, such that $(2S - 1)D \ll \omega_0$, then again $\tilde{\omega}_0 \approx \omega_0$, and the considered $2S$-photon transition is not much different from the one-photon transition, with the same theory [14–20] being applicable.

The worst case occurs when we are interested in a multiphoton transition in a spin sample with high magnetic anisotropy, such that $(2S - 1)D \gg \omega_0$. Then the effective transition frequency (31) is $\tilde{\omega}_0 \approx -(2S - 1)Dz$, which changes with time together with $z$. Because of this, to keep the resonance condition (47) valid, one should respectively vary with time the natural resonator frequency $\omega$. It is feasible, in principle, to imagine such a sliding resonance, when the circuit characteristics, as inductance and capacity, are changing in time so that to preserve the approximate equality $\omega \approx \tilde{\omega}_0$, following the varying $\tilde{\omega}_0$. But it looks that such a sliding resonance would be difficult to realize experimentally. However, even not this is the major obstacle. When the magnetic anisotropy is the prevailing part in the effective frequency (31), so that $\tilde{\omega}_0 \approx -(2S - 1)Dz$, then the spin-resonator coupling (48) is proportional to $-Dz$. As a result, $\alpha z \sim -Dz^2$, which is always negative for the easy-axis anisotropy, with $D > 0$. Hence, the collective attenuation $\Gamma$, given in Eq. (51), is always negative. This means, according to Eqs. (53), (54), or (59), that the function $y$ decreases with time. Therefore, there is no generation of coherent radiation. For this to occur, the collective frequency $\Gamma$ must, at least for some period of time, become negative. As is evident from Eqs. (54) and (59), a negative attenuation $\Gamma$ leads to the generation of coherent radiation, but a positive $\Gamma$ leads to the decay of transition coherence. Thus, a too strong easy-axis magnetic anisotropy suppresses transverse coherence, hindering the development of multiphoton spin superradiance.

In the case of an easy-plane anisotropy, with $D < 0$, the quantity $\alpha z \sim |D|z^2$ is positive. Hence, $\Gamma = \gamma_2 - \alpha z$ could be negative. However, a strong $x-y$-plane anisotropy implies that it is difficult to noticeably polarize spins in the $z$-direction. That is, $z$ is always small, which prevents making $\alpha z$ large, such that $\alpha z > \gamma_2$. Therefore a strong single-site anisotropy is an obstacle for achieving a well-developed transition coherence.

### 8 Quantum Stage

Let us assume that there are no strong external fields imposing on the spin system an essential transverse coherence, so that at the initial stage the motion of transverse spins is not coherent. This is the most interesting case to consider how the transverse coherence develops in a self-organized way from the initially incoherent motion. The period of time, when there are yet no collective correlations but only quantum spin interactions are present can be called the quantum stage.

At the very beginning of the process, when $\gamma t \ll 1$, the spin-resonator coupling function (48) is yet close to zero, and we may set $\alpha \rightarrow 0$. If there are no transverse external resonator fields, which implies that $\nu_1 = \nu_c = 0$, hence $\beta = 0$, then the effective attenuation (58) reduces to $J = \gamma_3$. In that case, when collective effects, due to the correlation of spins
through the feedback field have not yet been developed, the evolution equations (59) and (60) are yet quite simple, having the form

\[
\frac{dy}{dt} = -2\gamma_2 y + 2\gamma_3 z^2, \quad \frac{dz}{dt} = -(\gamma_1 + \gamma_3)z + \gamma_1 \sigma.
\]  

(61)

Then at short time \( t \to 0 \), we have

\[
y \simeq \left( y_0 - \frac{\gamma_3 z_0^2}{\gamma_2} \right) e^{-2\gamma_2 t} + \frac{\gamma_3 z_0^2}{\gamma_2}, \quad z \simeq \left( z_0 - \frac{\gamma_1 \sigma}{\gamma_1 + \gamma_3} \right) e^{-(\gamma_1 + \gamma_3)t} + \frac{\gamma_1 \sigma}{\gamma_1 + \gamma_3}.
\]  

(62)

With time, spin correlations increase owing to the resonator feedback field. Strengthening spin correlations result in the developing transverse coherence. When the transition coherence is well developed, collective phenomena come into play and the dynamic behaviour of spins becomes qualitatively different from that at the initial stage. The change in the features of spin motion occurs, of course, gradually. However, it is possible to define the moment of time separating these two regimes of motion. The qualitative change in spin dynamics occurs when the collective attenuation width (50) changes its sign because of the growing spin-resonator coupling. Then the quantum stage transfers to the coherent stage. The crossover time \( t_c \), separating these stages is given by the equation

\[
\Gamma(t_c) = 0.
\]  

(63)

The latter, in the case of good resonance, takes the form

\[
\alpha(t_c) z(t_c) = \gamma_2.
\]  

(64)

Introducing the effective spin-resonator coupling parameter

\[
g = \frac{\gamma\gamma_0 \tilde{\omega}_0}{\gamma_2(\gamma_2 + \Delta^2)},
\]  

(65)

in which

\[
\tilde{\omega}_0 \equiv \omega_0 - (2S - 1)Dz_0, \quad \tilde{\Delta} \equiv \omega - |\tilde{\omega}_0|,
\]  

(66)

we obtain from Eq. (64) the crossover time

\[
t_c = \tau \ln \left( \frac{gz_0}{gz_0 - 1} \right).
\]  

(67)

The latter, to be positive and finite, requires that

\[
gz_0 > 1 \quad (0 < t_c < \infty).
\]  

(68)

When condition (68) is not valid, then the quantum stage will be never replaced by the coherent one. For instance, if \( gz_0 = 1 \), then \( t_c \to \infty \). When the initial spin polarization is
sufficiently high and the spin-resonator coupling (65) is strong, then the crossover time (67) is

\[ t_c \simeq \frac{\tau}{g z_0} \quad (g z_0 \gg 1). \]

For the coherent regime to appear, it is necessary that the crossover time (67) be shorter than the following relaxation times,

\[ t_c \ll \left\{ T_1 \equiv \frac{1}{\gamma_1}, T_2 \equiv \frac{1}{\gamma_2}, T_3 \equiv \frac{1}{\gamma_3} \right\}. \tag{69} \]

In the other case, spin polarization would relax before the transition coherence could arise. Then the solutions (62) at the crossover time can be written as

\[ y(t_c) \simeq y_0 + 2 \gamma_3 t_c z_0^2, \quad z(t_c) \simeq z_0 + \gamma_1 t_c \sigma. \tag{70} \]

If the single-site anisotropy is strong, such that \((2S - 1)Dz_0 \gg \omega_0\), then \(\tilde{\omega}_0 \sim -z_0\). Since the spin-resonator coupling (65) is proportional to \(\tilde{\omega}_0\), then \(g \sim -z_0\), from where it follows that \(g z_0 \sim -z_0^2\), which is always nonpositive. Hence, condition (68) cannot be held true. This means that the coherent stage can never appear. In order that the transition coherence could develop, the external magnetic field \(B_0\) has to be sufficiently strong so that to suppress the destructive role of the magnetic anisotropy.

The dynamic broadening width \(\gamma_3\) in Eqs. (61) has been assumed to be a constant. In general, it can be a function of time. For example, considering the dynamics of the spin polarization at the beginning of the process, when \(t \to 0\), and when \(\gamma_1 \ll \gamma_3\), we have the equation

\[ \frac{dz}{dt} = -\gamma_3 z \quad (t \to 0). \]

Setting here

\[ \gamma_3(z) \simeq \frac{\gamma_s z_0}{2(z_0 - z)} \quad (z \to z_0) \]

results in the square-root relaxation \([66]\)

\[ z \simeq z_0(1 - \sqrt{\gamma_s t}) \]

observed in molecular magnets \([45]\) at short times, below the blocking temperature. Thus, for \(\text{Mn}_{12}\) one has \(\gamma_s \approx 10^{-2}\) s.

At longer times, below the blocking temperature, the relaxation of magnetization in molecular magnets follows a stretched exponential law \([37,42,44,49,51,52]\). This can be derived if

\[ \gamma_3(t) = \frac{\kappa}{t} (\gamma_s t)^\kappa \quad (0 < \kappa \leq 1), \]

which gives

\[ z = z_0 \exp \left\{-(\gamma_s t)^\kappa \right\}. \]

The power \(\kappa \approx 0.5\) for \(T < T_B\) and \(\kappa = 1\) for \(T > T_B\). The relaxation parameter \(\gamma_s\) depends on the applied magnetic field.
Recall that all the consideration of this section concerns the situation when collective effects have not yet come into play, so that the spin-resonator coupling is negligible, \( \alpha \approx 0 \). But if the spin system is coupled to a resonator and condition (68) is valid, then there exists a finite crossover time (67) when the quantum stage is followed by the coherent one.

9 Coherent Stage

After the crossover time (67), the coupling function (48) fastly increases to the value \( \alpha \approx g\gamma_2 \). This means that collective effects come into play and the transition coherence starts developing. At the transient stage, when time is larger than \( t_c \) but much shorter than \( T_1 \), one may neglect the longitudinal relaxation, omitting \( \gamma_1 \) in Eq. (60). Superradiance occurring at this transient stage, when

\[
t_c \leq t \ll T_1 ,
\]

is the transient superradiance.

If there are no transverse external fields, then the effective attenuation (58) is \( J = \gamma_3 \). For a sufficiently large coupling parameter (65), such that \( g\gamma_2 \gg \gamma_3 \), one may also neglect in Eqs. (59) and (60) the terms containing \( \gamma_3 \). Then these equations read

\[
\begin{align*}
\frac{dy}{dt} &= -2\gamma_2(1-gz)y , \\
\frac{dz}{dt} &= -g\gamma_2 y .
\end{align*}
\]

Equations (72) can be solved exactly [15–18] yielding

\[
\begin{align*}
y &= \left( \frac{\gamma_p}{g\gamma_2} \right)^2 \text{sech}^2 \left( \frac{t - t_0}{\tau_p} \right) , \\
z &= -\frac{\gamma_p}{g\gamma_2} \tanh \left( \frac{t - t_0}{\tau_p} \right) + \frac{1}{g} ,
\end{align*}
\]

where the pulse width \( \gamma_p \), the pulse time \( \tau_p \), with the relation \( \gamma_p \tau_p = 1 \), and the delay time \( t_0 \) are the integration constants to be defined from the initial conditions taken at the crossover time \( t_c \). Equating the functions (73) to the values (70) gives for the pulse width the equation

\[
\gamma_p^2 = \gamma_g^2 + (g\gamma_2)^2(y_0 + 2\gamma_3 t_c z_0^2) , \quad \gamma_g = \gamma_2(1-gz_0) , \quad \gamma_p \tau_p \equiv 1 ,
\]

and for the delay time, we get

\[
t_0 = t_c + \frac{\tau_p}{2} \ln \left| \frac{\gamma_p - \gamma_g}{\gamma_p + \gamma_g} \right| .
\]

Solutions (73) describe a superradiant pulse. Since, by definition, superradiance is a self-organized process, we consider the case when there is no strong transverse coherence imposed on the system at the initial time. This implies the inequality

\[
g^2 y_0 < 1 .
\]

In order that the delay time (75) would be finite, and larger than the crossover time, so that

\[
t_c < t_0 < \infty ,
\]
it is necessary that
\[ y_0 + 2\gamma_3 t_c z_0^2 > 0, \quad g z_0 > 1. \] (78)

One more condition on superradiance is that the pulse time be shorter than the spin-spin dephasing time,
\[ \tau_p < T_2. \] (79)

Then, taking into account that \( \gamma_3 t_c \ll 1 \), we may distinguish two superradiant regimes: *triggered superradiance*, when
\[ g z_0 > 1 + \sqrt{1 - g^2 y_0}, \quad y_0 \neq 0, \] (80)
and *pure superradiance*, when
\[ g z_0 > 2, \quad y_0 = 0. \] (81)

One of the necessary conditions for superradiance to occur is \( g z_0 > 1 \), which is the same condition (68) for the existence of the crossover time (67). This condition, in view of Eqs. (65) and (66), can be written as
\[ \omega_0 z_0 > (2S - 1) D z_0^2 + \frac{\gamma_2 (\gamma^2 + \tilde{\Delta}^2)}{\gamma z_0}, \]
which tells us that the longitudinal magnetic field \( B_0 \) has to be sufficiently strong and the initial spin polarization must be positive, \( z_0 > 0 \).

For the pure spin superradiance, when \( y_0 = 0 \), taking into consideration that \( \gamma_3 t_c \ll 1 \), the pulse width and time can be presented as
\[ \gamma_p = (g z_0 - 1) \gamma_2 (1 + \gamma_3 \tilde{t}_c), \quad \tau_p = \frac{T_2}{g z_0 - 1} (1 - \gamma_3 \bar{t}_c), \] (82)
and the delay time (75) takes the form
\[ t_0 = t_c + \frac{\tau_p}{2} \ln \left| \frac{2}{\gamma_3 \bar{t}_c} \right|, \] (83)
where the notation
\[ \tilde{t}_c \equiv t_c e^{2\gamma t_c} = \tau \left( \frac{g z_0}{g z_0 - 1} \right)^2 \ln \left( \frac{g z_0}{g z_0 - 1} \right) \]
is used. When \( \gamma t_c \ll 1 \), which is equivalent to the inequality \( g z_0 \gg 1 \), then \( \tilde{t}_c \simeq t_c \).

At the delay time \( t = t_0 \), the coherence intensity \( y \) is maximal, which follows from the solutions (73) giving
\[ y(t_0) = \left( z_0 - \frac{1}{g} \right)^2 (1 + 2\gamma_3 t_c), \quad z(t_0) = \frac{1}{g}. \] (84)

After this, the transition coherence fastly decays. Thus, for \( t \gg t_0 \), one has
\[ y \simeq 4y(t_0) e^{-2\gamma p t}, \quad z \simeq -z_0 + \frac{2}{g}. \] (85)
For large $g \gg 1$, the initial spin polarization becomes almost completely inverted. This effect can be used for fast polarization reversal in polarized solid-state targets employed in scattering experiments [10,79]. The polarization reversal under a superradiant burst is illustrated in Fig. 1, which results from numerical simulations [21–23].

10 Pulsing Regime

After the transient superradiant burst, occurring at the delay time $t_0$, the transition coherence dies out, unless the inversion of spin polarization is supported by a permanent pumping. This pumping can be accomplished by means of dynamic nuclear polarization. In the latter case, the regime of pulsing spin superradiance can be achieved. To study this regime, we consider a spin system without external transverse fields. Then the effective attenuation (58) is $J = \gamma_3$. At long times, such that $\gamma t \gg 1$, the coupling function (48) reaches its maximal value $\alpha \approx g\gamma_2$. In that case, Eqs. (59) and (60) can be written as

$$ \frac{dy}{dt} = v_1 , \quad \frac{dz}{dt} = v_2 , \quad (86) $$

with the right-hand sides

$$ v_1 = -2\gamma_2(1 - gz)y + 2\gamma_3z^2 , \quad v_2 = -g\gamma_2y - \gamma_3z - \gamma_1^*(z - \sigma) . \quad (87) $$

In the presence of the permanent polarization pumping, the pumping rate $\gamma_1^*$ takes the place of the spin-lattice relaxation parameter $\gamma_1$, while $\sigma \in [-1, 1]$ is a pumping parameter.

To understand the behaviour of the solutions $y$ and $z$ at long times $t \to \infty$, let us find the stationary points of Eqs. (86), which are given by the equations $v_1 = v_2 = 0$. In general, these equations give us two stationary points $y_1^*$, $z_1^*$ and $y_2^*$, $z_2^*$, also called fixed points. Among these, however, only those have sense, which satisfy the physical restrictions

$$ 0 \leq y \leq 1 , \quad -1 \leq z \leq 1 . \quad (88) $$

Also, the actual solutions of Eq. (86) will tend to that fixed point which is stable, whose properties can be derived by means of the Lyapunov stability analysis. The stability of fixed points depends on the values of the spin-resonator coupling (65) and of the effective pumping parameter

$$ \sigma^* \equiv \frac{\gamma_1^*\sigma}{\gamma_1^* + \gamma_3} . \quad (89) $$

The value of this parameter depends on the relation between the pumping rate $\gamma_1^*$ and the dynamic broadening width $\gamma_3$. Since the effects of temperature can be modelled through a thermal bath of random fluctuations [80], the temperature dependence can be incorporated in $\gamma_3$.

When the pumping is weak, such that

$$ g\sigma^* \leq -1 , \quad (90) $$

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then the stable fixed point corresponds to the solutions
\[ y_1^* \simeq \frac{(\gamma_1^* + \gamma_3)\gamma_3}{g^2\gamma_1^*\gamma_2} |1 + g\sigma^*| , \quad z_1^* \simeq \sigma \left(1 + \frac{\gamma_3}{g\sigma\gamma_1^*}\right). \] (91)

The related characteristic exponents are
\[ X_1^+ \simeq -\gamma_1^* - \frac{\gamma_1^* - 2\gamma_2}{g\sigma\gamma_2} \gamma_3 , \quad X_1^- \simeq -2\gamma_2(1 - g\sigma) - \gamma_3. \] (92)

The fixed point (91) is a stable node. The coherence intensity \( y_1^* \sim \gamma_3/g\gamma_2 \) is small.

When the spin-resonator coupling is weak, so that \(|g\sigma^*| \ll 1\), then the stationary point is given by the solutions
\[ y_1^* \simeq \gamma_1^* \gamma_3 (\sigma^*)^2 \left(1 + \frac{\gamma_1^* - \gamma_3}{\gamma_1^* + \gamma_3} g\sigma^*\right) , \quad z_1^* \simeq \sigma^* \left(1 - \frac{\gamma_3}{\gamma_1^* + \gamma_3} g\sigma^*\right). \] (94)

The corresponding characteristic exponents
\[ X_1^+ \simeq -\gamma_1^* - \gamma_3 \left(1 - \frac{4\gamma_2}{\gamma_1^* - 2\gamma_2 + \gamma_3} g\sigma^*\right) , \quad X_1^- \simeq -2\gamma_2 \left(1 - \frac{\gamma_1^* - 2\gamma_2 - \gamma_3}{\gamma_1^* - 2\gamma_2 + \gamma_3} g\sigma^*\right) \] (95)
show that the fixed point (94) is, as early, a stable node. The limiting value \( y_1^* \ll 1 \). In these both cases of either Eq. (90) or Eq. (93), the solutions monotonically tend to the stationary point \( y_1^*, z_1^* \).

When the spin-resonator coupling as well as pumping are sufficiently strong, so that
\[ g\sigma^* \geq 1 , \] (96)
then the solutions of the evolution equations (86) tend to another fixed point
\[ y_2^* \simeq \frac{\gamma_1^* + \gamma_3}{g^2\gamma_2} (g\sigma^* - 1) , \quad z_2^* \simeq \frac{1}{g} \left(1 - \frac{\gamma_3}{g\sigma\gamma_1^*}\right). \] (97)

Now, the characteristic exponents are complex valued,
\[ X_2^ \pm \simeq -\frac{1}{2} \gamma_1^* - \frac{1}{2} \gamma_3 \left(1 + \frac{2\gamma_2}{g\sigma\gamma_1^*}\right) \pm i\omega_\infty , \] (98)
containing the asymptotic frequency
\[ \omega_\infty \simeq \sqrt{2g\sigma\gamma_1^*\gamma_2}. \] (99)

The fixed point (97) is a stable focus. The limiting value of the coherence intensity \( y_2^* \sim \gamma_1^*/g\gamma_2 \) is small. However, in the way to the fixed point (97), the spin system exhibits a
series of superradiant bursts, which can be demonstrated by solving Eqs. (86) numerically [20,30,71]. This is the regime of pulsing spin superradiance. The superradiant bursts are not equidistant in time, and become approximately periodic only at \( t \to \infty \), with the asymptotic period

\[
T_\infty \equiv \frac{2\pi}{\omega_{\infty}} \simeq \pi \sqrt{\frac{2T_1^* T_2}{g\sigma}},
\]

where \( \gamma_1^* T_1^* \equiv 1 \). Each superradiant burst, occurring in the time interval \( 0 < t < T_1^* \), possesses a high level of transition coherence, but after \( T_1^* \) the amplitude of the coherence intensity \( y \) diminishes, tending to \( y_2^* \).

In conclusion, superradiant regimes may be employed in the operation of spin masers. There are three possible such regimes: triggered superradiance, pure superradiance, and pulsing superradiance. In materials with higher spins, as molecular magnets, it is necessary to take into account single-site magnetic anisotropy. If this anisotropy is strong, it hinders the possibility of achieving a high level of transition coherence in multiphoton transitions. However, by a sufficiently strong external magnetic field the destructive role of magnetic anisotropy can be suppressed. Another possibility is to tune the resonant electric circuit to one of the quantum transition lines of a high spin. When a sole transition line is selected by the resonant tuning, the situation becomes similar to the case of an effective two-level system.

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Figure Caption

Fig. 1. Intensity of transition coherence $y$ in arbitrary units (upper curve) and the longitudinal spin polarization $z$ (lower curve) as functions of time measured in units of $T_2$, obtained from numerical simulations for 300 spins.