Origin of Pure Spin Superradiance

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The question addressed here is: What originates pure spin superradiance in a polarized spin system placed inside a resonantor? The term "pure" means that no initial coherence is imposed on spins, and its appearance manifests a purely self-organized collective effect. An accurate solution of evolution equations for a microscopic model is given. The results show that the resonator Nyquist noise does not play any role in starting spin superradiance, but the emergence of the latter is initiated by local spin fluctuations due to nonsecular dipole interactions.

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A polarized spin system prepared in a nonequilibrium state returns to equilibrium through the spin-spin and spin-lattice relaxation mechanisms. The spin relaxation can be drastically accelerated if the nonequilibrium magnetic system is placed inside a coil of a resonance electric circuit with the natural frequency tuned to the precession frequency of spin magnetic moments [1]. The strong shortening of the relaxation time is caused by the coherence between individual rotating spins, which develops as a result of the interaction between the rotating magnetization and the resonator feedback field. This coherent phenomenon is analogous to the Dicke superradiance [2] occurring in atomic and molecular systems, so Bloembergen and Pound [1] also called this fast collective damping in spin systems the radiation damping. Friedberg and Hartmann [3] noted that, in fact, the whole process in spin systems involves no radiation at all, but merely nonradiative transfer of energy from the sample to the coil and back. Nevertheless, the term superradiance has been accepted for the transient coherent phenomenon in spin systems, when it develops as a self-organized process, similar to the Dicke superradiance. One more justification for using the term spin superradiance is that this effect is always accompanied by coherent magnetodipole radiation, though the corresponding radiation intensity is very weak as compared to the easily measured current power [4].

The term *pure spin superradiance* is used in order to stress that this is a purely self-organized process, when coherence develops from an absolutely incoherent state. This is to be distinguished from *triggered spin superradiance,* during which collective effects also play an important role, but the process starts from a coherent initial state, so that the imposed initial coherence triggers the development of a correlated state, in the same way as triggered optical superradiance [5] happens in atomic and molecular systems. When spin superradiance is caused by nuclear spins, it can be called the *nuclear spin superradiance.* A system of ion spins in a resonator cavity can, in principle, be also a source of spin superradiance.

The nuclear spin superradiance has been recently observed in a series of experiments [6–11] with different substances: from Al nuclear spins in ruby (Al_2O_3) and from proton spins in propanediol ($C_3H_8O_2$), butanol (C_4H_9OH), and ammonia (NH_3). The interpretation of the pure spin superradiance in these experiments is commonly based on the following picture. A system of polarized spins is placed in a constant magnetic field directed opposite to the sample polarization. The sample is put inside the coil of a passive electric circuit whose natural frequency is tuned to the Zeeman frequency of spins. The fluctuating magnetic field formed by the thermal Nyquist noise of the resonance circuit starts moving spins from their position of unstable equilibrium. The motion of spins induces an electric current in the circuit, which creates a stronger magnetic field acting back on spins. Under the action of the feedback field, spins move faster increasing even more the resonater feedback field, and so on. This avalanche-type process results in a fast spin relaxation.

However, in this, generally correct, picture there is one suspicious point, namely, that the beginning of the process is originated by the thermal Nyquist noise of the resonator. If one attentively reads the classical paper by Bloembergen and Pound [1], then one finds there the estimate for the thermal damping, due to the thermal noise in resonator, showing that this damping is so negligibly small for macroscopic systems that it can never produce the initial thermal relaxation.

Thus we confront the alternative: either the common belief that this is the resonator thermal noise which initiates the pure spin superradiance is a delusion or Bloembergen and Pound are wrong. To resolve this paradox and to answer the question "what actually is the origin of pure spin superradiance" is the aim of the present paper.

The solution of the formulated problem meets the following difficulty. As follows from the analysis of Bloembergen and Pound [1], the homogeneous approach provided by the Bloch equation is not sufficient for

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correctly describing the process, but inhomogeneous local fields that produce a microscopic relaxation mechanism are essential. The phenomenological Bloch equation, even being solved in a reasonably accurate approximation [12], can describe only the triggered spin superradiance, when an initial coherence is imposed by assuming nonzero initial conditions for transverse magnetization. To take into account inhomogeneous local fields providing a microscopic relaxation mechanism means the necessity of dealing with a microscopic model. Writing the equations of motion for spin components we get a $3N$-dimensional system of nonlinear differential equations for N spins, plus the Kirchhoff equation for an electric circuit. If one invokes any approach to solve this system of equations based on the uniform mean-field approximation, then one immediately returns to a homogeneous picture equivalent to the Bloch equation, thus losing information on local fields. When the number of spins N is not too large, say, $N \sim 10-10^3$, then it is possible to resort to numerical calculations [4,13]. However, such computer simulations are able to give only a qualitative description, since the number of spins involved is incomparably smaller than what one has in real samples with $N \sim 10^{23}$. Below, an analytical solution of microscopic equations is presented.

A system of nuclear spins is described [14] by the Hamiltonian

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

with dipole spin interactions

$$H_{ij} = (\mu^2/r_{ij})\left[\vec{S}_i \cdot \vec{S}_j - 3\left(\vec{S}_i \cdot \vec{n}_{ij}\right)\left(\vec{S}_j \cdot \vec{n}_{ij}\right)\right], \tag{2}$$

where μ is a nuclear magneton and

$$\vec{n}_{ij} \equiv \vec{r}_{ij}/r_{ij}, \quad \vec{r}_{ij} \equiv \vec{r}_i - \vec{r}_j, \quad r_{ij} \equiv |\vec{r}_{ij}| .$$

The total magnetic field

$$\vec{B} = H_0 \vec{e}_z + H \vec{e}_x \tag{3}$$

consists of a constant external field H_0 and an alternating field H of a resonator coil. The latter has n turns of a cross-section area A_c over a length l. The resonance electric circuit includes a resistance R, inductance L, and capacity C. The alternating resonator field

$$H = (4\pi n/cl)j \tag{4}$$

is formed by an electric current satisfying the Kirchhoff equation

$$L\frac{dj}{dt} + Rj + \frac{1}{C}\int_0^t j(\tau)\,d\tau = -\frac{d\Phi}{dt} + E_f , \tag{5}$$

in which E_f is an electromotive force and

$$\Phi = \frac{4\pi}{c} n A_c \eta \rho \frac{\mu}{N} \sum_{i=1}^{N} \langle S_i^x \rangle$$

is a magnetic flux through the coil, $\eta \equiv V/V_c$ being a filling factor, $V_c \equiv lA_c$ the coil volume, and $\rho \equiv N/V$ the density of spins.

Define the resonator natural frequency $\omega \equiv 1/\sqrt{LC}$, ringing width $\gamma_3 \equiv R/2L$, and dimensionless fields

$$h \equiv \mu H/\hbar\gamma_3, \quad f \equiv c\mu E_f/nA_c\hbar\gamma_3^2. \tag{6}$$

Introduce the parameter

$$\alpha_0 \equiv \pi\eta\rho\mu^2/\hbar\gamma_3 \tag{7}$$

characterizing the strength of coupling between the spin system and the resonator. Let us also use the notation

$$u \equiv \frac{1}{N}\sum_{i=1}^{N}\langle S_i^- \rangle, \quad s \equiv \frac{1}{N}\sum_{i=1}^{N}\langle S_i^z \rangle \tag{8}$$

for the mean spin components, where $\langle \cdots \rangle$ implies statistical averaging. Then the Kirchhoff equation (5) takes the form

$$\frac{dh}{dt} + 2\gamma_3 h + \omega^2 \int_0^t h(\tau)\,d\tau = -2\alpha_0(u^* + u) + \gamma_3 f . \tag{9}$$

To derive the evolution equation for the variables (8), we proceed as follows. Write the Heisenberg equations for the corresponding spin components with the standard notation $\omega_0 \equiv \mu H_0/\hbar$ for the Zeeman frequency. Decouple the double spin correlators in the manner described by ter Haar [15], in order to preserve the terms containing the homogeneous spin-spin relaxation $\gamma_2 \equiv T_2^{-1}$, which can be done by using second-order perturbation theory. For generality, we may also include the term describing the spin-lattice relaxation $\gamma_1 \equiv T_1^{-1}$. These steps are known and clear. The most difficult problem is how to treat the local spin fields

$$\delta_i = \frac{1}{\hbar}\sum_{j(\neq i)}^{N}\left\langle \tfrac{3}{2} a_{ij} S_j^z + c_{ij} S_j^+ + c_{ij}^* S_j^- \right\rangle,$$
$$\varphi_i = -\frac{2}{\hbar}\sum_{j(\neq i)}^{N}\left\langle b_{ij} S_j^+ + c_{ij} S_j^z \right\rangle, \tag{10}$$

caused by the dipole interactions

$$a_{ij} = \frac{\mu^2}{r_{ij}^3}(1 - 3\cos^2\vartheta_{ij}),$$
$$b_{ij} = -\frac{3\mu^2}{4r_{ij}^3}\sin^2\vartheta_{ij}\exp(-i2\varphi_{ij}), \tag{11}$$
$$c_{ij} = -\frac{3\mu^2}{4r_{ij}^3}\sin^2(2\vartheta_{ij})\exp(-i\varphi_{ij}),$$

where ϑ_{ij} and φ_{ij} are the spherical angles of $\vec{n}_{ij}$. Note that in a uniform approximation the local fluctuating fields (10) are zero because of the properties of the dipole interactions (11). To get a closed set of equations, at the same time retaining the information on the presence of fluctuating fields (10), we may replace the latter by stochastic fields, $\delta_i \longrightarrow \varphi_0$, $\varphi_i \longrightarrow \varphi$, the first of which, in compliance with (10), is real and the second is complex. The distribution of these random fields is such that the averaging over it, which we shall denote by $\langle\langle \cdots \rangle\rangle$, gives

$$\langle\langle \varphi_0 \rangle\rangle = \langle\langle \varphi \rangle\rangle = 0, \quad \langle\langle \varphi_0^2 \rangle\rangle = \frac{1}{2}\langle\langle |\varphi|^2 \rangle\rangle = \gamma_*^2, \quad (12)$$

where the dispersion γ_*, in accordance with (10), is of the order of γ_2. In this way, for the spin components (8) we obtain the system of stochastic equations

$$\frac{du}{dt} = i(\omega_0 - \varphi_0 + i\gamma_2)u - i(\gamma_3 h + \varphi)s,$$
$$\frac{ds}{dt} = \frac{i}{2}(\gamma_3 h + \varphi)u^* - \frac{i}{2}(\gamma_3 h + \varphi^*)u - \gamma_1(s - \zeta),$$
$$\frac{d|u|^2}{dt} = -2\gamma_2|u|^2 - i(\gamma_3 h + \varphi)su^* + i(\gamma_3 h + \varphi^*)su. \quad (13)$$

The structure of (13) is transparent: $\gamma_3 h + \varphi$ is the total effective field acting on spins; h is the resonator field defined by (9); φ_0 and φ model random local fields with a distribution whose particular form is not important since all we need is the property (12). If φ_0 and φ were absent, then (13) would be reduced to the Bloch equation.

To consider the case of pure spin superradiance, the initial conditions for the system of Eqs. (9) and (13) are to be taken as

$$h(0) = 0, \quad u(0) = 0, \quad s(0) = z_0. \quad (14)$$

The electromotive force $E_f = E_0 \cos\omega t$ in (5) corresponds to the resonance mode of the thermal Nyquist noise. The driving force in (6) is

$$f = f_0 \cos\omega t, \quad f_0 \equiv c\mu E_0 / n A_c \hbar \gamma_3^2. \quad (15)$$

The system of equations (9) and (13) can be solved by a method [16] combining the guiding-center approach [17] and the method of averaging [18]. The idea is straightforward: First, we classify the variables as fast or slow. To this end, we take into account the usual inequalities $\gamma_1 \ll \omega_0$, $\gamma_2 \ll \omega_0$, $\gamma_3 \ll \omega$, and consider the quasiresonance case, when $|\Delta| \ll \omega_0$, where $\Delta \equiv \omega - \omega_0$ is detuning. Then, we notice right away that the variables h and u can be treated as fast, and s and $|u|^2$ as slow. Keeping the latter as fixed parameters

$$s = z, \quad |u| = v, \quad (16)$$

we get for the fast variables a system of linear equations, which, therefore, is not too difficult to solve. The found solutions for fast variables are to be substituted into the equations for slow variables, and the right-hand sides of these equations are to be averaged over the period $2\pi/\omega_0$ of fast oscillations and also over the distribution of stochastic fields. This procedure results in the equations

$$\frac{dz}{dt} = g\gamma_2 w - \gamma_1(z - \zeta) - \gamma_f z,$$
$$\frac{dw}{dt} = -2\gamma_2 w - 2g\gamma_2 wz + 2\gamma_f z^2 \quad (17)$$

for the slow variables, where

$$w \equiv v^2 - \frac{2\gamma_*^2}{\omega_0^2} z, \quad g \equiv \alpha_0\left(\frac{\gamma_3}{\gamma_2}\right)\frac{\pi(\gamma_2 - \gamma_3)^2}{(\gamma_2 - \gamma_3)^2 + \Delta^2},$$

and the attenuation

$$\gamma_f \equiv \frac{f_0^2\gamma_3^4}{32\omega_0^2(\Delta^2 + \gamma_2^2)}\left\{\left(1 + \frac{8\pi^2}{3}\right)\gamma_2 - 2\pi\Delta + \frac{\omega_0 z}{\Delta^2 + \gamma_2^2}[(\alpha - 2\pi\beta)(\Delta^2 - \gamma_2^2) + 2\gamma_2\Delta(\beta + 2\pi\alpha)]\right\}, \quad (18)$$

in which

$$\alpha \equiv \alpha_0\left(\frac{\gamma_3}{\omega_0}\right)\frac{\pi(\gamma_2 - \gamma_3)^2}{(\gamma_2 - \gamma_3)^2 + \Delta^2},$$
$$\beta \equiv \alpha_0\left(\frac{\gamma_3}{\omega_0}\right)\frac{\pi(\gamma_2 - \gamma_3)\Delta}{(\gamma_2 - \gamma_3)^2 + \Delta^2}$$

is due to the action of the driving field (15).

The amplitude of the electromotive force related to the thermal Nyquist noise [19], at temperature T satisying the inequality $\hbar\omega \ll k_B T$, is given by $E_0^2 = \gamma_3 R k_B T/\pi$. Hence for the amplitude of the driving field (15) we have

$$f_0^2 = 8\alpha_0 k_B T / \pi\hbar\gamma_3 N. \quad (19)$$

Let us accept the values of parameters characteristic of experiments [7–11] with proton spins $\omega_0 \sim \omega \sim 10^8\ \mathrm{sec}^{-1}$, $\gamma_1 \sim 10^{-5}\ \mathrm{sec}^{-1}$, $\gamma_2 \sim 10^5\ \mathrm{sec}^{-1}$, $\gamma_3 \sim 10^6\ \mathrm{sec}^{-1}$, $T \sim 0.1$ K, and $N \sim 10^{23}$. Then $f_0 \sim 10^{-10}$ and the thermal attenuation (18) is $\gamma_f \sim 10^{-16}\ \mathrm{sec}^{-1}$. Such an insignificant quantity, of course, plays no role, as compared to all other damping parameters, and has to be neglected in (17).

This result shows, in agreement with Bloembergen and Pound [1], that the Nyquist noise of the resonator can never produce the initial thermal relaxation, and thus cannot originate the pure spin superradiance.

Omitting in (17) the negligibly small γ_f and taking into account that $\gamma_1 \ll \gamma_2$, we come to

$$\frac{dz}{dt} = g\gamma_2 w, \quad \frac{dw}{dt} = -2\gamma_2 w(1 + gz). \quad (20)$$

According to (14), the initial conditions are $z(0) = z_0$ and $v(0) = 0$. Equations (20) are exactly integrable yielding

$$z = \frac{\gamma_0}{g\gamma_2}\tanh\left(\frac{t - t_0}{\tau_0}\right) - \frac{1}{g},$$
$$v^2 = \left(\frac{\gamma_0}{g\gamma_2}\right)^2 \mathrm{sech}^2\left(\frac{t - t_0}{\tau_0}\right) + \frac{2\gamma_*^2}{\omega_0^2} z; \quad (21)$$

here γ_0 is the radiation width given by

$$\gamma_0^2 = \Gamma_0^2 - 2(g\gamma_2)^2 \varepsilon_* z_0 , \tag{22}$$

where

$$\Gamma_0 \equiv \gamma_2(1 + gz_0), \quad \varepsilon_* \equiv (\gamma_*/\omega_0)^2,$$

the radiation time $\tau_0 = \gamma_0^{-1}$, and the delay time is

$$t_0 = \frac{\tau_0}{2} \ln \left| \frac{\gamma_0 - \Gamma_0}{\gamma_0 + \Gamma_0} \right| . \tag{23}$$

The criterion for the occurrence of spin superradiance is the validity of the inequalities

$$0 < t_0 < \infty, \quad \tau_0 < T_2 . \tag{24}$$

Invoking (22) and (23) and bearing in mind that $\varepsilon_* \ll 1$, we find that (24) is equivalent to

$$z_0 < z_p \equiv -2/g, \quad \varepsilon_* > 0 . \tag{25}$$

As long as $|z_0| < \frac{1}{2}$, the first of the inequalities (25) requires that $g \geq 4$. In this way, the pure spin superradiance occurs when the initial spin polarization z_0 is negative, with an absolute value surpassing the threshold $|z_p| = 2/g$, when the coupling of the spin system with a resonator is sufficiently strong, $g \geq 4$, and if there exist local random fields with a nonzero dispersion $\gamma_* > 0$.

To emphasize the crucial importance of the local fields, let us notice that if one puts $\varepsilon_* \to 0$, then $\gamma_0 \to |\Gamma_0|$ and $|t_0| \to \infty$. That is, without these fields the pure spin superradiance is impossible. To make the essential dependence of the delay time on ε_* apparent, we may write (23) for the case of strong coupling, when $g|z_0| \gg 1$, then

$$t_0 \simeq \frac{T_2}{2g|z_0|} \ln \left| \frac{2z_0}{\varepsilon_*} \right| .$$

From here it is evident that $t_0 \to \infty$ as $\varepsilon_* \to 0$. For the parameters typical of experiments [7–11], we have $t_0 \sim 10^{-6} - 10^{-5}$ sec. So, these are the local random fields that are responsible for starting the process of self-organization leading to the pure spin superradiance.

One more question is worth answering: Which part of the local fields is more important for initiating the pure spin superradiance? Recall that the stochastic fields entering into the evolution equations (13) are related to two types of local fields defined in (10). As follows from (13), the term δ_i in (10), corresponding to φ_0, only shifts the rotation frequency, while the term φ_i, corresponding to φ, starts moving the spin z component even when the resonator feedback field h is yet absent. The term φ_i in (10) is due to the dipole interactions b_{ij} and c_{ij} defined in (11). These interactions, in the theory of magnetic resonance [14], are called nonsecular interactions, as compared to the secular interaction a_{ij}. The initial motion of spins, when $u(0) = 0$ and $h(0) = 0$, is due solely to the action of nonsecular interactions. This conclusion is in agreement with computer simulations [4,20] for small and mesoscopic spin systems with $N \sim 10-10^3$. Thus, we are in a position to give the final answer to the question posed in this paper: The pure spin superradiance in a nonequilibrium system of polarized nuclear spins can be originated only by local fields due to nonsecular dipole interactions. The thermal Nyquist noise of resonator plays no role in this process.

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