Processing Information by Punctuated Spin Superradiance

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The possibility of realizing the regime of punctuated spin superradiance is advanced. In this regime, the number of superradiant pulses and the temporal intervals between them can be regulated. This makes it feasible to compose a kind of a Morse code alphabet and, hence, to develop a technique of processing information.

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Spin systems can exhibit a phenomenon that is analogous to atomic superradiance [1], because of which it is called spin superradiance. To realize this phenomenon, spin systems, similarly to atomic ones, are to be prepared in an inverted state. This is achieved by placing a polarized spin sample in an external magnetic field directed opposite to spin polarization. Contrary to atomic systems, coherent spin motion develops not owing to direct spin correlations but due to the interaction of spins with a resonator feedback field, for which purpose the spin sample has to be coupled with a resonant electric circuit, whose natural frequency is tuned to the Zeeman frequency of spins [2]. More details on similarities and differences between atomic superradiance and spin superradiance can be found in the review [3]. Spin superradiance is the process of *coherent spontaneous emission* by moving spins. As in the case of atomic systems, one may distinguish two main types of this phenomenon: transient superradiance and pulsing superradiance. *Transient spin superradiance* occurs when the spin sample is prepared in the inverted state, after which no following pumping is involved. In this case, a single superradiant burst arises, peaked at the delay time. *Pulsing spin superradiance* is radically different from the transient regime by the occurrence of a series of superradiant pulses, for which the spin sample is to be subject to a permanent pumping supporting the inverted spin polarization. Both regimes of spin superradiance, transient [4–6] as well as pulsing [7], were observed in experiments with different materials containing nuclear spins. A microscopic theory of these phenomena, based on realistic spin Hamiltonians, was developed [8–11], being in good agreement with experiment and with computer modeling [12]. It is worth stressing that, only by invoking microscopic Hamiltonians, it has become possible to give an accurate description of purely self-organized regimes which cannot be treated by the phenomenological Bloch equations [8–10].

In the present paper, we advance the possibility of realizing the third type of spin superradiance, which we call *punctuated spin superradiance,* and which is principally different from the transient and pulsing types. In this regime, unlike the transient case, not a single but many superradiant bursts can be produced. In distinction to the pulsing regime, where the number of pulses and the temporal distance between them are prescribed by a given setup and cannot be varied, in the process of punctuated superradiance both the number of superradiant bursts as well as time intervals between each pair of them can be regulated. The term *punctuation* here means this feasibility of changing interpulse intervals and of organizing various groups of superradiant bursts. In that way, a code, such as the Morse alphabet, can be composed, which may be employed in processing information.

The consideration below will be based on the Hamiltonian typical for spin systems employed in magnetic resonance [13]. The Hamiltonian reads

$$\hat{H} = \sum_i \hat{H}_i + \frac{1}{2}\sum_{i\neq j}\hat{H}_{ij}\,, \qquad (1)$$

where $\hat{H}_i$ corresponds to individual spins, while $\hat{H}_{ij}$, to spin interactions, with the indices $i, j = 1, 2, \ldots, N$ enumerating spins. The individual terms are given by the Zeeman energy $\hat{H}_i = -\mu_0 \mathbf{B}\cdot\mathbf{S}_i$, where $\mu_0 \equiv \hbar\gamma_S$, with γ_S being the gyromagnetic ratio of spin S, represented by the spin operator $\mathbf{S}_i$, and $\mathbf{B}$ is the total magnetic field acting on each spin. The spin interactions are described by the dipolar terms $\hat{H}_{ij} = \sum_{\alpha\beta} C_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta$, with the dipolar tensor $C_{ij}^{\alpha\beta}$. The total magnetic field $\mathbf{B} = B_0\mathbf{e}_z + H\mathbf{e}_x$ consists of a constant longitudinal field B_0 and a transverse field H formed by the resonant electric circuit coupled to the spin sample. The resonator field $H = 4\pi nj/cl$ is created by the electric current j circulating over a coil of n turns and length l. The current j is determined by the Kirchhoff equation. The electric circuit is characterized by resistance R, inductance L, and capacity C. With the notation for the circuit natural frequency $\omega \equiv 1/\sqrt{LC}$ and circuit damping $\gamma \equiv R/2L$, the Kirchhoff equation can be presented as

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

for the resonator magnetic field H, where η is a filling factor and $M_x = (\mu_0/V)\sum_i \langle S_i^x\rangle$ is the x component of the magnetization density of a sample with volume V. Since the resonator field H, acting on spins, is itself due to the transverse spin motion, this field H is called the feedback field.

To derive evolution equations, we follow the scale separation approach, described in full detail in Refs. [8–11]. For this purpose, we write the Heisenberg equations of motion for the lowering, S_i^-, raising, S_i^+, and polarization, S_i^z, operators. In these equations, it is possible to separate the combinations describing fast fluctuating local fields. Employing the method of random local fields [13], the latter are modeled by stochastic Gaussian variables, with zero mean and the width $2\gamma_3$, where γ_3 is the width of inhomogeneous dynamic broadening. Then the Heinsenberg equations are averaged over spin degrees of freedom, not touching the stochastic variables. Denoting the averaging over spins by single angle brackets $\langle \ldots \rangle$, we define the transition function x, coherence intensity y, and spin polarization z, respectively:

$$x \equiv \frac{1}{S}\langle S_i^-\rangle, \qquad y \equiv \frac{1}{S^2}\langle S_i^+\rangle\langle S_i^-\rangle, \qquad z \equiv \frac{1}{S}\langle S_i^z\rangle . \tag{3}$$

The wavelength of spin radiation is usually much larger than interparticle distance, because of which the uniform approximation for Eqs. (3) may be employed.

We direct the external magnetic field B_0 so that $\mu_0 B_0 < 0$ and define the Zeeman frequency $\omega_0 \equiv |\mu_0 B_0|/\hbar$. Also, introduce the notation $f \equiv -(i/\hbar)\mu_0 H + \xi$ for an effective force acting on spins. Finally, the evolution equations for the functions (3) can be cast [8–11] to the form

$$\begin{aligned} \frac{dx}{dt} &= -i(\omega_0 + \xi_0 - i\gamma_2)x + fz\,, \\ \frac{dy}{dt} &= -2\gamma_2 y(x^* f + f^* x)z\,, \\ \frac{dz}{dt} &= -\frac{1}{2}(x^* f + f^* x) - \gamma_1(z - \sigma)\,, \end{aligned} \tag{4}$$

where γ_1 and γ_2 are the longitudinal and transverse widths, respectively, and σ is an equilibrium polarization of a spin. When there is no external stationary pumping, $\sigma = -1$. Equations (4) are stochastic differential equations, since they contain random fields.

To solve Eqs. (4), we invoke a generalization [8–10] of the averaging technique [14] to the case of stochastic differential equations. This becomes possible owing to the existence of several small parameters: $\gamma_0/\omega_0 \ll 1$, $\gamma_1/\omega_0 \ll 1$, $\gamma_2/\omega_0 \ll 1$, and $\gamma_3/\omega_0 \ll 1$, where $\gamma_0 \equiv \pi\eta\rho\mu_0^2 S/\hbar$ is the natural width and $\rho \equiv N/V$ is the density of spins. In addition, the resonant circuit, coupled to the spin sample, is assumed to be of good quality and tuned close to the Zeeman frequency ω_0, so that $\gamma/\omega \ll 1$, $|\Delta|/\omega \ll 1$, with $\Delta \equiv \omega - \omega_0$.

First of all, using the occurrence of the small parameters, we can obtain an iterative solution of the Kirchhoff equation (2). For this purpose, invoking the method of Laplace transforms, we present Eq. (2) in the integral form

$$\begin{aligned} H &= -4\pi\eta \int_0^t G(t - t')\dot{M}_x(t')\,dt', \\ G(t) &= \left(\cos\omega' t - \frac{\gamma}{\omega'}\sin\omega' t\right)e^{-\gamma t}, \end{aligned} \tag{5}$$

where the overdot means time differentiation and $\omega' \equiv \sqrt{\omega^2 - \gamma^2}$. Since M_x is expressed through x, its time derivative $\dot{M}_x$ is directly related to the first of Eqs. (4). Using this, we find the solution of Eq. (5), in the first order with respect to small parameters, as $\mu_0 H/\hbar = i\alpha(x - x^*)$, where the function

$$\alpha = g\gamma_2(1 - e^{-\gamma t}), \qquad g \equiv \frac{\gamma\gamma_0\omega}{\gamma_2(\gamma^2 + \Delta^2)} \tag{6}$$

describes the intensity of coupling between the spin sample and the resonant circuit. Let us stress that the spin-resonator coupling (6) depends on time, taking into account retardation effects.

From Eqs. (4), in the presence of the small parameters, it follows that the variables y and z are temporal quasi-invariants with respect to x. Then, we solve the first of Eqs. (4), with these quasi-invariants fixed, substitute the found solution x into the second and third of Eqs. (4), and average the right-hand sides of these equations over time and over the stochastic fields. As a result, we obtain the guiding center equations,

$$\begin{aligned} \frac{dy}{dt} &= -2(\gamma_2 - \alpha z)y + 2\gamma_3 z^2, \\ \frac{dz}{dt} &= -\alpha y - \gamma_3 z - \gamma_1(z - \sigma)\,. \end{aligned} \tag{7}$$

In the dynamics of solutions to Eqs. (7), one may distinguish two stages, quantum and coherent. At the *quantum stage,* when $\gamma t \ll 1$, the coupling function is close to zero and no noticeable coherence in the motion of transverse spins is yet developed. The dynamics is governed by quantum spin interactions. At this stage, when $\alpha \approx 0$, Eqs. (7) are linear, and their solution is easy. With increasing time, the spin-resonator coupling (6) grows, and coherent effects, caused by the resonator feedback field, gradually come into play. The crossover time between the quantum and the coherent regimes can be defined as the time t_c, at which the first term in the first of Eqs. (7) changes its sign. This is because the quantity $\Gamma \equiv \gamma_2 - \alpha z$ plays the role of an effective attenuation. When the latter is positive, transverse coherence decays, while a negative attenuation implies the generation of coherence. Hence, the moment of time, when $\Gamma(t_c)$ changes its sign, separates qualitatively different regimes of spin motion. The *crossover time* t_c, defined by the equality $\alpha(t_c) \times z(t_c) = \gamma_2$, is $t_c = \tau\ln[gz_0/(gz_0 - 1)]$, with $\gamma\tau = 1$. The solutions y and z at this boundary of the quantum stage are $y(t_c) \simeq y_0 + 2\gamma_3 t_c z_0^2$ and $z(t_c) \simeq z_0 + \gamma_1 t_c \sigma$, where $y_0 = y(0)$ and $z_0 = z(0)$. The *coherent stage* of motion comes after the crossover time t_c, when the spin-resonator coupling α fastly grows to $g\gamma_2$. Since $\gamma_3 \leq \gamma_2$, and if $g \gg 1$, then $g\gamma_2 \gg \gamma_3$, and the term with γ_3 can be omitted. In the transient regime, when $t \ll T_1 \equiv \gamma_1^{-1}$, the term containing γ_1 can also be neglected. Then Eqs. (7) possess the exact solution

$$y = \left(\frac{\gamma_p}{g\gamma_2}\right)^2 \operatorname{sech}^2\left(\frac{t - t_0}{\tau_p}\right), \qquad z = -\frac{\gamma_p}{g\gamma_2} \tanh\left(\frac{t - t_0}{\tau_p}\right) + \frac{1}{g}, \tag{8}$$

in which the pulse time τ_p and the pulse width γ_p, with $\gamma_p \tau_p \equiv 1$, are defined by the expressions $\gamma_p^2 = \gamma_g^2 + (g\gamma_2)^2(y_0 + 2\gamma_3 t_c z_0^2)$, $\gamma_g \equiv \gamma_2(1 - g z_0)$, and the delay time is

$$t_0 = t_c + \frac{\tau_p}{2} \ln\left|\frac{\gamma_p - \gamma_g}{\gamma_p + \gamma_g}\right| . \tag{9}$$

Solution (8) describes a transient superradiant burst, with the maximal intensity at the delay time t_0, when $y(t_0) = (z_0 - 1/g)^2(1 + 2\gamma_3 t_c)$, $z(t_0) = 1/g$. After this, for $t \gg t_0$, the coherence intensity exponentially diminishes and the spin polarization becomes inverted,

$$y \simeq 4y(t_0)e^{-2\gamma_p t}, \qquad z \simeq -z_0 + 2/g . \tag{10}$$

For sufficiently large coupling parameter g, the reversal of spin polarization is practically complete.

Now imagine that at some time after $t_0 + \tau_p$ we again invert the spin polarization from that in Eq. (10) to the symmetric positive value. For large g, this inversion is practically from $-z_0$ to z_0. Such an inversion can be realized in three possible ways: inverting the external magnetic field B_0, acting on spins by a resonant π-pulse, or just turning the sample 180° about an axis perpendicular to B_0. As a result, we get again a strongly nonequilibrium state of inverted spins. After the time t_0, counted from the moment when the newly nonequilibrium state is prepared, another superradiant burst will arise. After the second burst dies out, one can again invert the spin polarization by one of the mentioned methods. Then one more superradiant burst will appear. This procedure can be repeated as many times as necessary for creating a required number of sharp superradiant pulses. The time intervals between bursts can be regulated, allowing the formation of different groups of pulses, with varying intervals between separate groups. Thus, it is feasible to compose a code, similar to the Morse alphabet, which can be used in processing information. It is this possibility of regulating temporal intervals between superradiant bursts which permits us to call the described phenomenon *punctuated spin superradiance*.

Spin superradiance can be realized in different materials under various experimental setups. Thus, it was observed on proton spins in propanediol $C_3H_8O_2$, butanol C_4H_9OH, ammonia NH_3 [4–6], and on ^{27}Al nuclear

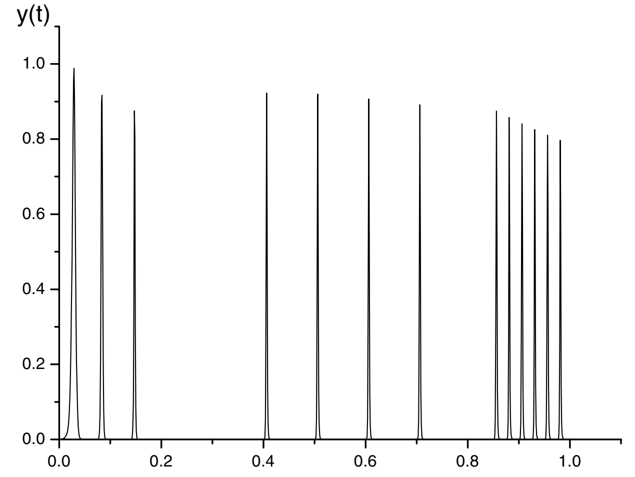

FIG. 1. Punctuated spin superradiance with three different groups of superradiant bursts. All parameters are explained in the text. Time is measured in dimensionless units. The time intervals are $t_0 = 0.029$, $\tau_1 = 0.065$, $\tau_{12} = 0.26$, $\tau_2 = 0.1$, $\tau_{23} = 0.15$, and $\tau_3 = 0.025$.

spins in ruby Al_2O_3 [7]. The characteristic parameters for these experiments with nuclear spins are the density of spins $\rho \sim 10^{22}$–10^{23} cm^{-3}, the Zeeman frequency $\omega_0 \sim 10^8$ Hz, the spin-lattice relaxation $\gamma_1 \sim 10^{-5}$ s^{-1}, the spin-spin dephasing parameter $\gamma_2 \sim 10^5$ s^{-1}, the dynamic broadening width $\gamma_3 \sim 10^4$–10^5 s^{-1}, and the resonator ringing time $\tau \sim 10^{-6}$ s, that is, the resonator damping $\gamma \sim 10^6$ s^{-1}. For these values, the spin-resonator coupling parameter g in Eq. (6) varies between 10 and 100. As has been shown [15], if nuclear spins are inside a ferromagnet or ferrimagnet, possessing long-range magnetic order, then the coupling parameter g can be increased by a factor of $\mu_B/\mu_N \sim 10^3$, where μ_B is the Bohr magneton and μ_N is the nuclear magneton. Therefore, the coupling parameter g can be made as large as $g \sim 10^5$. Among other materials, where

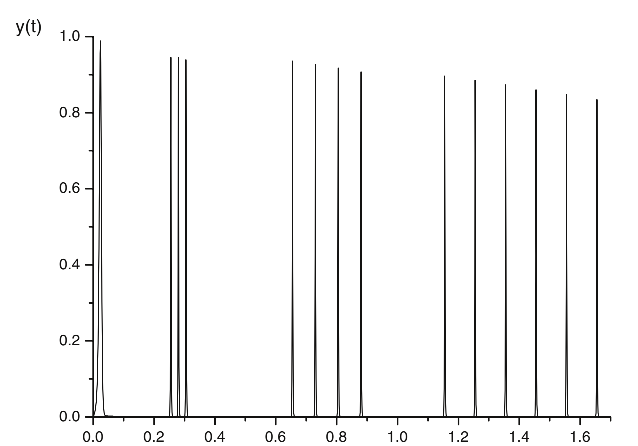

FIG. 2. Punctuated spin superradiance with four groups of superradiant pulses, the first group containing a single burst, the time intervals being $t_0 = 0.024$, $\tau_{12} = 0.226$, $\tau_2 = 0.025$, $\tau_{23} = 0.35$, $\tau_3 = 0.075$, $\tau_{34} = 0.275$, and $\tau_4 = 0.1$.

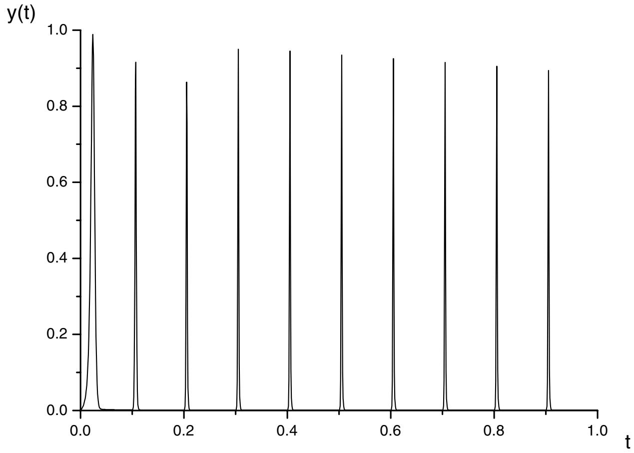

FIG. 3. Punctuated spin superradiance with a comb of equidistant pulses, starting at $t_0 = 0.024$, having time intervals $\tau_1 = 0.1$.

spin superradiance could, in principle, be observed, are granular magnets and molecular magnets. In this way, there exists a large variety of materials with different characteristics allowing for the optimal choice of parameters for realizing punctuated spin superradiance. Note that the intervals between superradiant pulses can be varied in a very wide diapason of the order of $T_1 = \gamma_1^{-1}$. For nuclear magnets, with $\gamma_1 \sim 10^{-5}\ s^{-1}$, this time T_1 may be as much as several days; and, for molecular magnets at low temperatures, it could range up to months.

To illustrate the feasibility of creating different groups of superradiant pulses, with varying time intervals, we solved Eqs. (7) numerically. Different time intervals are obtained by changing the moments of polarization inversion. For the characteristic parameters, those are taken that are typical of nuclear magnets [4–7]. In particular, we take $\gamma_1 = 10^{-5}\ s^{-1}$, $\gamma_2 = 10^5\ s^{-1}$, and γ_3 is varied between 10^4 and $10^5\ s^{-1}$. The variation of γ_3 even in a wider diapason does not essentially change the picture. The coupling parameter g has also been varied between 10 and 10^3; the whole picture being qualitatively the same, with the main difference that for larger g the spin inversion, according to Eq. (10), is better. For the presentation in Figs. 1to 3, we set $g = 10^3$. In the absence of dynamic nuclear polarization, $\sigma = -1$. The resonator attenuation $\gamma = 10^6\ s^{-1}$. Finally, as initial conditions we take $y(0) = 0$ and $z(0) = 1$. The first of the latter tells that at the initial time the transverse coherence is absent. That is, we consider a purely self-organized process when radiation coherence arises in a spontaneous manner. Figures 1to 3 give some examples of how it is possible to create different bunches of superradiant bursts. The shown function $y(t)$ is proportional to radiation intensity. The meaning of this function, according to its definition (3), is to describe the level of coherence in the system. As seen from the figures, the maxima of superradiant bursts display a high level of coherence, almost reaching 100%. The time variable in the figures is measured in units of $T_2 = \gamma_2^{-1}$. The short time scale is chosen here just for the convenience of presentation. As explained above, the same picture can be stretched to the time scale characterized by T_1, which, for nuclear magnets, would range up to several days. The first superradiant burst occurs at the delay time t_0. To simplify the figure captions, we accept the notation for the time intervals between the pulses of the i group as τ_i and for the intervals between the i and j groups as τ_{ij}. Figure 3 demonstrates that a regime of equidistant superradiant pulses can also be realized. Such a regime can be used for producing spin masers [9] operating in pulsing superradiant mode.

In conclusion, we have demonstrated, both analytically and numerically, the feasibility of realizing the regime of punctuated spin superradiance. In this regime, one may form various groups of superradiant bursts, with different spacing between the pulses inside each group as well as with different time intervals between the groups. The possibility of so punctuating spin superradiance can be employed for processing information.

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