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COHERENT NUCLEAR RADIATION

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The main part of this review is devoted to the comprehensive description of coherent radiation by nuclear spins. The theory of nuclear spin superradiance is developed and the experimental observations of this phenomenon are considered. The intriguing problem of how coherence develops from initially incoherent quantum fluctuations is analyzed. All main types of coherent radiation by nuclear spins are discussed, which are: free nuclear induction, collective induction, maser generation, pure superradiance, triggered superradiance, pulsing superradiance, punctuated superradiance, and induced emission. The influence of electron-nuclear hyperfine interactions and the role of magnetic anisotropy are studied. Conditions for realizing spin superradiance by magnetic molecules are investigated. The possibility of nuclear matter lasing, accompanied by pion or dibaryon radiation, is briefly touched.

Основная часть этого обзора посвящена всестороннему описанию когерентного излучения ядерными спинами. Развита теория ядерного спинового сверхизлучения и рассмотрены экспериментальные наблюдения этого явления. Проанализирована увлекательная проблема того, как когерентность возникает из начально некогерентных квантовых флуктуаций. Исследованы все основные типы когерентного излучения ядерными спинами: свободная ядерная индукция, коллективная индукция, мазерная генерация, чистое сверхизлучение, тригтерное сверхизлучение, пульсирующее сверхизлучение, прерывистое сверхизлучение и индуцированная эмиссия. Изучено влияние электрон-ядерных сверхтонких взаимодействий и роль магнитной анизотропии. Описаны условия реализации спинового сверхизлучения магнитными молекулами. Кратко рассмотрена возможность когерентного излучения ядерной материей пионов и дибарионов.

INTRODUCTION

Nuclei can radiate in different ways. For example, they emit gamma radiation in the process of changing internal quantum states. An ensemble of nuclei, emitting such hard photons, could form a coherent source, called gamma-ray laser or just gamma laser. However, the problem of gamma lasers remains a challenging but frustrating field of research, with not a great progress in theory and practically no experimental achievements [1–3].

Contrary to this, there exists a type of coherent nuclear radiation that is well documented experimentally and for which a detailed microscopic theory has recently been developed. This is nuclear spin radiation. The main part

of the present review is just devoted to the phenomenon of coherent radiation by nuclear spins. We, first, explain in simple words the physics of this effect and survey the basic experiments where it was observed. Then we pass to developing a comprehensive microscopic theory of strong nonlinear dynamics of nuclear spins. A special attention is paid to a very intriguing problem of how coherence arises from an incoherent quantum motion of randomly fluctuating spins. We describe all principal regimes of spin superradiance and study the influence of the hyperfine electron-nuclear coupling. We analyse the role of the single-ion magnetic anisotropy that is so important in the spin radiation of magnetic molecules. Finally, we note that not only photons can be emitted coherently by nuclei. Thus, excited nuclear matter can produce coherent radiation of gluons, pions, and dibaryons. Different types of coherent nuclear radiation can find a variety of applications, some of which are discussed in the review. The content of the latter is as follows: Introduction, 1. Nuclear Spin Superradiance, 2. Ensemble of Nuclear Spins, 3. Nuclear Spin Waves, 4. Scale Separation Approach, 5. Incoherent Quantum Stage, 5.1. Radiation by Magnetic Dipoles, 5.2. Resonator Nyquist Noise, 5.3. Local Spin Fluctuations, 6. Regimes of Coherent Radiation, 6.1. Transient Spin Superradiance, 6.2. Pulsing Spin Superradiance, 6.3. Induced Coherent Emission, 7. Electron-Nuclear Hyperfine Coupling, 8. Enhanced Nuclear Radiation, 9. Superradiance by Magnetic Molecules, 10. Pion and Dibaryon Radiation, Conclusion

1. NUCLEAR SPIN SUPERRADIANCE

Atomic systems, radiating at optical frequencies, exhibit a number of coherent effects [4, 5]. The majority of the latter have their counterparts in spin systems, generating radiation at radio-frequencies. One of the most interesting coherent effects is superradiance that may occur in both atomic and spin systems. The possibility of superradiance in atomic systems was predicted by Dicke [6], and the feasibility of organizing coherent motion of spins was discussed by Bloembergen and Pound [7]. The modern status of nuclear spin superradiance is presented in review [8].

By definition, superradiance is the process of *coherent spontaneous radiation*. Being spontaneous, it is assumed to be self-organized. In general, a coherent motion of spins can be realized by means of a strong external magnetic field. But this would result in nuclear induction, which, though beeing a coherent process, however is not superradiance. To realize the latter, one, first, needs to prepare the spin ensemble in a nonequilibrium state. For this, it is possible to polarize spins in one direction and after that to place them in an external magnetic field of opposite direction. Such a system of spins would be analogous to an ensemble of inverted atoms. Several other similarities between atomic and spin systems are

discussed in reviews [8, 9]. Two important problems related to spin assemblies are: What is the cause provoking spins to start their initial motion, after they have been inverted? And what is the mechanism collectivizing the following spin motion, making the latter coherent? The answer to the second question was proposed by Bloembergen and Pound [7] who suggested to place the spin sample into an electric coil being a part of an electric circuit with a natural frequency tuned close to the Zeeman transition frequency of spins. This coupling with a resonant electric circuit would produce a feedback field collectivizing the spin motion. The answer to the first question, what is the cause starting the spin motion, has not been understood for many years. This puzzle was solved recently [10] and will be considered in detail in the following sections. In brief, there are quantum fluctuations of spins that trigger the initial spin motion.

One more question would be how to measure the radiation generated by moving spins? The magnetodipole radiation, resulting from this motion, even being completely coherent, would have the intensity of order

$$I(t) \sim \frac{2}{3c^3}(\mu_0 I)^2 \omega_0^4 N^2,$$

where c is the light velocity; $\mu_0 \equiv \hbar\gamma_n$, with γ_n being the nucleus gyromagnetic ratio; I is the nuclear spin; ω_0 is the Zeeman transition frequency; N is the total number of spins taking part in the coherent radiation. Accepting the typical values $\mu_0 \sim 10^{-23}$ erg/G, $\omega_0 \sim 10^8$ Hz, and $N \sim 10^{23}$, we would have $I(t) \sim 10^{-6}$ W, where 1 W$= 10^7$ erg/s is a watt. This is a rather small quantity of the intensity $I(t)$, which would be difficult to notice. But this radiation induces electric current in the coil, with the power of current $P(t)$ being essentially larger than the intensity $I(t)$. Roughly speaking,

$$\frac{P(t)}{I(t)} \sim \frac{\lambda^3}{V_c},$$

where λ is the radiation wavelength and V_c is the coil volume. This ratio, for $\lambda \sim 10^2 - 10^3$ cm and $V_c \sim 10$ cm^3, is of order $10^5 - 10^8$, which makes $P(t)$ an easily measurable quantity. Thus, the magnetodipole radiation of a spin sample is quite weak and practically does not propagate into free space, but mainly is taken up by a resonant coil surrounding the sample [11].

One should not confuse superradiance with other types of coherent radiation. By definition, superradiance is *spontaneous collective emission*. The term *spontaneous* implies that the process is *self-organized* but not induced by external fields. And the word *collective* means that the radiation characteristics are essentially influenced by collective effects. For instance, the radiation intensity of N radiators is proportional to N^α, with $\alpha > 1$, while the radiated pulse is short, with the radiation time proportional to $N^{1-\alpha}$. Depending on the relation between characteristic times, there can arise different kinds of coherent radiation. Among these

typical times, the most important are: the crossover time t_c, separating the quantum incoherent stage of spin motion from their coherent rotation; the pulse time τ_p of an emitted pulse; and the dephasing time T_2. In the case, when there are no external transverse fields, one can distinguish seven coherent radiation regimes:

1) *Free induction* ($t_c = 0,\ \tau_p \geqslant T_2$).

Coherence of radiation here is not self-organized but imposed upon the system by an external field. This is the standard nuclear free decay induced by an initial coherent pulse or, equivalently, by an essential transverse polarization [12].

2) *Collective induction* ($t_c = 0,\ \tau_p < T_2$).

The shortening of the radiation damping is due to the coupling with a resonator. However, the process is mainly governed not by a self-organization but by a strong external coherent field or by an initial transverse polarization [7].

3) *Maser generation* ($t_c > 0,\ \tau_p \geqslant T_2$).

There are no external coherent fields, though some incoherent nonresonant pumping may exist. Self-organization becomes crucial. But the emitted pulses are not sufficiently narrow, implying that self-organization is not yet high. Although this is a spontaneous coherent process, it is not yet a genuine superradiance. Such a type of coherent maser generation was observed in many experiments [13–16]. The physics of this process is analogous to the known laser generation [17–19].

4) *Pure superradiance* ($t_c > 0,\ \tau_p < T_2$).

This is a purely self-organized process, developing from an incoherent chaotic stage to a highly correlated spin motion. Though some of the features of spin superradiance are similar to those of atomic superradiance [4, 20–22], there are also several principal differences. Experiments on and theory of spin superradiance will be described in detail below.

5) *Triggered superradiance* ($t_c > 0,\ \tau_p < T_2$).

In this process, self-organization is crucially important, but there is as well a weak external resonant field triggering and influencing the behaviour of spins. There are similarities with triggered superradiance in optics [20].

6) *Pulsing superradiance* ($t_c > 0,\ \tau_p < T_2$).

This regime differs from pure superradiance by the existence of a series of superradiant pulses, instead of a single superradiant burst. To realize such a pulsing superradiance, it is necessary to maintain a sufficiently high level of spin polarization by means of an incoherent pumping. The latter can be done, e.g., by dynamic nuclear polarization [12, 23, 24].

7) *Punctuated superradiance* ($t_c > 0,\ \tau_p < T_2$).

Here, similar to pulsing superradiance, there is a series of superradiant pulses; however this is achieved not by supporting nuclear polarization but by means of resonant external fields or forces. All regimes of spin superradiance will be thoroughly considered in the following sections.

From the above classification it follows that superradiance, in addition to being a coherent and spontaneous emission, should correspond to short pulses

with a finite crossover time, so that

$$t_c > 0, \qquad \tau_p < T_2 .$$

Thus, the correct and full definition of superradiance would be:

Superradiance is a coherent spontaneous emission by an ensemble of radiators of short pulses peaked at a time that is larger than the crossover time, with the duration of each pulse shorter than the dephasing time.

This definition suits for any kind of superradiance, whether it is atomic or spin superradiance. In what follows, we concentrate on spin superradiance produced by an ensemble of nuclear spins. Initially, we shall give a brief survey of experiments observing nuclear spin superradiance.

Pure spin superradiance by nuclei was first observed in Dubna [25, 26] and confirmed in St. Petersburg [27, 28]. These experiments were accomplished with propanediol $C_3H_8O_2$. This is a material rich of protons, with the density $\rho_H \approx 4 \cdot 10^{22}$ cm^{-3}. The proton spins were polarized, by dynamic nuclear polarization, in an external magnetic field $B_0 \sim 1$ T, which corresponds to the Zeeman frequency $\omega_0 \sim 10^8$ Hz. The sample was refrigerated to low temperatures $T \sim 0.1$ K, at which the nuclear spin-lattice relaxation was strongly suppressed, with the longitudinal relaxation time $T_1 \sim 10^5$ s. The transverse dephasing time, due to dipole spin-spin interactions, was $T_2 \sim 10^{-5}$ s. The coupled resonant electric circuit had a quality factor $Q \sim 100$ and a ringing time $\tau \sim 10^{-6}$ s.

Similar experiments were accomplished in Bonn [29] with butanol C_4H_9OH and ammonia NH_3. In these materials, the proton density $\rho_H \sim 10^{23}$ cm^{-3}. The characteristic Zeeman frequency was $\omega_0 \sim 10^8$ Hz. The materials were kept at low temperature, when the spin-lattice relaxation is suppressed, with the longitudinal relaxation time $T_1 \sim 10^5$ s. The transverse dephasing time was $T_2 \sim 10^{-5}$ s. The quality factor of the resonator electric circuit was $Q \sim 30$, and the ringing time $\tau \sim 5 \cdot 10^{-7}$ s.

Such proton-rich materials are widely used as targets in studying the scattering of particle beams from accelerators. It would, of course, be possible to involve for experiments on spin superradiance other materials employed as targets in high-energy scattering studies. For instance, a good candidate would be pentanol $C_5H_{12}O$, whose proton density is $\rho_H \approx 6 \cdot 10^{22}$ cm $^{-3}$.

Pulsing spin superradiance was explored in Zürich [30–33] on the basis of nuclei of ^{27}Al inside the ruby crystal Al_2O_3, where the density $\rho_{Al} \approx 4 \cdot 10^{22}$ cm^{-3}. The nuclei ^{27}Al have spins $I = 5/2$. The crystal was oriented in an external magnetic field $B_0 \sim 1$ T so that the fully resolved structure of the five $\Delta m = \pm 1$ transition lines could be clearly seen. When a resonant circuit is tuned to a selected transition line, then ^{27}Al spins form an effective two-level system. In experiments [30–33], the circuit was usually tuned to the central $\{-1/2, 1/2\}$

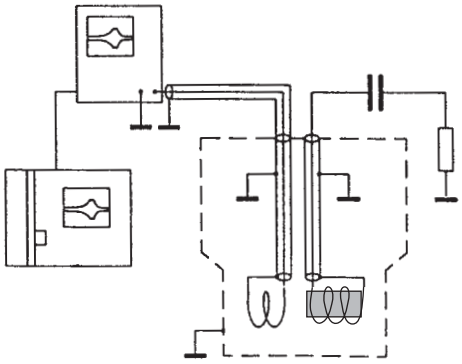

Fig. 1. Typical set-up in experiments on spin superradiance. The left coil plays the role of antenna. The right coil, surrounding the studied sample, is a part of the resonant electric circuit. The sample is shown as a dark bar inside the resonant coil. The upper left block is an oscilloscope, and the lower one is a plotter. The dashed line symbolizes refrigerator

line, with a transition frequency $\omega_0 \sim 10^8$ Hz. At low temperatures around $T \sim 1$ K, the spin-lattice relaxation time was $T_1 \sim 10^5$ s and the transverse dephasing time was $T_2 \sim 10^{-5}$ s. The resonant circuit had the quality factor $Q \sim 100$ and 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 Γ_1^* between 0.01 and 10 s^{-1}.

A typical experimental set-up employed for observing spin superradiance [25, 26] is shown in Fig. 1. The detected superradiance pulse has the form illustrated in Fig. 2 for two different initial spin polarizations. The higher is the prepared inversion, the stronger is the superradiant burst.

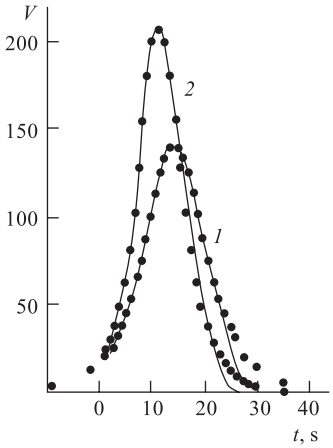

Fig. 2. Voltage signal of a superradiant pulse as a function of time, measured in units of 10^{-7} s, for two initial spin polarizations: $s_0 = 0.52$ (*1*) and $s_0 = 0.57$ (*2*)

The influence of the passive resonant circuit, coupled to a nuclear spin system, was also studied in nuclear spin echo experiments [34–36]. In the ferrite $Li_{0.5}Fe_{2.5}O_4$, the spins of nuclei ^{57}Fe were tuned to a resonant circuit with a frequency about $7 \cdot 10^7$ Hz. In the compound Co_2MnSi, the working substance was the nuclei ^{59}Co, whose spins were coupled to a circuit of frequency $1.4 \cdot 10^8$ Hz. The experiments [34–36] showed that the presence of the resonant circuit can enhance the intensity of the nuclear spin echo signal by up to 40%, as compared to the case without coupling to such a resonant circuit [37].

The theoretical description of spin dynamics in the presence of a resonant circuit has been commonly done on the basis of the Bloch plus Kirchhoff equations [27–33, 38–40]. Analytical solutions were usually obtained by involving

the adiabatic approximation. However, the latter approximation is only valid for dynamics close to stationary and it does not correctly describe transient effects, such as superradiant bursts. What is more, the Bloch equations presuppose the existence of coherent motion from the beginning, since these are classical equations. Such equations principally are not able to depict self-organized coherent phenomena, as pure superradiance, which develops from an initially incoherent quantum stage. This principal defect of Bloch equations cannot be overcome even if one employs more elaborate approximations [41, 42] based on the averaging technique [43]. As was first shown in Ref. 10, for the correct description of pure spin superradiance it is crucially important to take account of stochastic local spin fluctuations. This conclusion was also confirmed by numerical calculations [44–46].

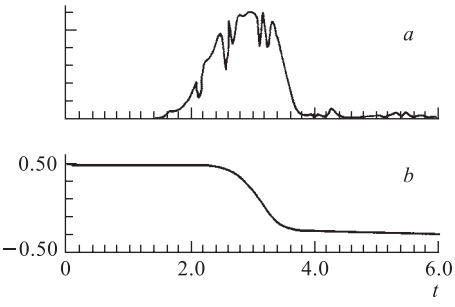

Fig. 3. Intensity of radiation (*a*) in arbitrary units and the longitudinal spin polarization (*b*) as functions of time, measured in units of T_2, obtained from computer simulations for 300 spins

Computer simulations of spin superradiance have been accomplished [47–51], investigating different regimes of coherent spin motion. A typical result of simulations is shown in Fig. 3. The weak points of such simulations are: First, one has to deal with a rather limited number of spins, say 10^2 or 10^3, because of which the obtained solutions give only a qualitative picture. Second, in such simulations, spins are treated as classical vectors, hence, quantum fluctuations, so important at the initial stage of motion, are not properly considered. Third, numerical results do not always present clear physical explanations of the studied processes. This is especially so, when there are tens of parameters to be varied, while in reality only some combinations of these parameters are important.

To give a thorough description of nuclear spin superradiance, it is necessary to develop a good microscopic theory of spin dynamics in a sample coupled to a resonant electric circuit. The feedback field created by the resonator is crucial for organizing the coherent motion of spins, moving from a strongly nonequilibrium state. Note that for spin motion close to equilibrium, as in nuclear magnetic resonance experiments, the feedback field is not of much significance [52]. For describing such a purely self-organized process as pure spin superradiance, it is vital to take into account local quantum spin fluctuations, which trigger the initial spin motion and define the delay time of the superradiant burst [10].

A comprehensive microscopic theory of spin superradiance, based on realistic spin Hamiltonians [12, 23, 53, 54] has been developed [10, 55–60]. This theory

for the first time has made it possible to discover the actual origin of pure spin superradiance [10, 55–57], to give an accurate description of various regimes of nonlinear spin dynamics [55–59], to present an explicit picture of how coherence develops from chaotic spin fluctuations [10, 60], to consider spin superradiance in different materials, such as polarized nuclear paramagnets [10, 56–59], ferromagnets, ferrimagnets [61–63], and molecular magnets [60], to analyze the superradiant operation of spin masers [60, 64, 65], and to advance the feasibility of punctuated spin superradiance [66]. Brief survey of this theory was done in reviews [8, 9]. In the following sections, we shall give a detailed account of the theory and of several its applications.

2. ENSEMBLE OF NUCLEAR SPINS

Consider a solid sample containing N nuclear spins I_i enumerated by the index $i = 1, 2, \ldots, N$. The Hamiltonian of the nuclear system can be written as

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

with $\hat{H}_i$ being related to individual spins and $\hat{H}_{ij}$ corresponding to spin interactions. The single-spin term

$$\hat{H}_i = -\mu_0 \mathbf{B} \cdot \mathbf{I}_i \tag{2}$$

is the Zeeman energy, where $\mu_0 \equiv g_I \mu_N = \hbar \gamma_n$; with g_I being the Landé factor for a nucleus of spin I; μ_N, nuclear magneton; γ_n, nuclear gyromagnetic ratio. Generally, for nuclei, μ_0 can be positive as well as negative. The total magnetic field

$$\mathbf{B} = B_0 \mathbf{e}_z + (B_1 + H) \mathbf{e}_x \tag{3}$$

contains external longitudinal, B_0, and transverse, B_1, magnetic fields, and also a feedback field H of the resonant electric circuit. The pair terms

$$\hat{H}_{ij} = \sum_{\alpha\beta} D_{ij}^{\alpha\beta} I_i^\alpha \, I_j^\beta \tag{4}$$

in the Hamiltonian (1) correspond to dipolar spin interactions through the dipolar tensor

$$D_{ij}^{\alpha\beta} = \frac{\mu_0^2}{r_{ij}^3} \left(\delta_{\alpha\beta} - 3 n_{ij}^\alpha \, n_{ij}^\beta \right) , \tag{5}$$

in which $\alpha, \beta = x, y, z$ and

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

The dipolar tensor enjoys the properties

$$\sum_{j(\neq i)} D_{ij}^{\alpha\beta} = 0, \qquad \sum_{\alpha} D_{ij}^{\alpha\alpha} = 0, \tag{6}$$

of which the second is exact and the first one is asymptotically exact for a macroscopic (in all directions) sample with a large number of spins $N \gg 1$.

The single-spin term (2), with the total magnetic field (3), can be written as

$$\hat{H}_i = -\mu_0 B_0 I_i^z - \frac{1}{2}\mu_0 (B_1 + H)(I_i^+ + I_i^-), \tag{7}$$

where $I_j^{\pm} \equiv I_j^x \pm i I_j^y$ are the ladder spin operators. Employing the latter, the interaction term (4) can be presented in the form

$$\hat{H}_{ij} = a_{ij}\left(I_i^z I_j^z - \frac{1}{2} I_i^+ I_j^- \right) + b_{ij} I_i^+ I_j^+ + b_{ij}^* I_i^- I_j^- + 2c_{ij} I_i^+ I_j^z + 2c_{ij}^* I_i^- I_j^z, \tag{8}$$

in which the notation

$$a_{ij} \equiv D_{ij}^{zz}, \quad c_{ij} \equiv \frac{1}{2}\left(D_{ij}^{xz} - i D_{ij}^{yz} \right), \quad b_{ij} \equiv \frac{1}{4}\left(D_{ij}^{xx} - D_{ij}^{yy} - 2i D_{ij}^{xy} \right) \tag{9}$$

is introduced. From the first of Eqs. (6), it follows that

$$\sum_{j(\neq i)} a_{ij} = \sum_{j(\neq i)} b_{ij} = \sum_{j(\neq i)} c_{ij} = 0. \tag{10}$$

Note also that the quantities (9) are symmetric, so that $a_{ij} = a_{ji}$, $b_{ij} = b_{ji}$, and $c_{ij} = c_{ji}$.

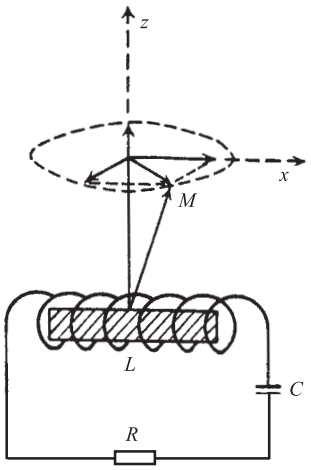

Fig. 4. Orientation of the coordinate axes with respect to the spin sample inserted into the coil of an electric circuit

In the total magnetic field (3), the external longitudinal and transverse fields B_0 and B_1, respectively, are supposed to be prescribed. The resonator feedback field H is created by the electric current of the coil surrounding the sample. The orientation of the coordinate axes with respect to the latter is illustrated in Fig. 4. The electric circuit is 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 described by the Kirchhoff equation

$$L\frac{dj}{dt} + Rj + \frac{1}{C}\int_0^t j(t')\, dt' = E_f - \frac{d\Phi}{dt}, \tag{11}$$

in which E_f is an electromotive force and

$$\Phi = \frac{4\pi}{c} n A_c \eta m_x \tag{12}$$

is a magnetic flux formed by the x component of the magnetization density

$$m_x = \frac{\mu_0}{V} \sum_i \langle I_i^x \rangle, \tag{13}$$

where $\eta \approx V/V_c$ is a filling factor; V is the sample volume, and the brackets $\langle \ldots \rangle$ imply statistical averaging.

The electric current, circulating over the coil, creates a magnetic field

$$H = \frac{4\pi n}{cl} j. \tag{14}$$

Let us employ the standard notation for the circuit natural frequency

$$\omega \equiv \frac{1}{\sqrt{LC}}, \qquad L \equiv 4\pi \frac{n^2 A_c}{c^2 l}, \tag{15}$$

and the circuit damping

$$\gamma \equiv \frac{R}{2L} = \frac{\omega}{2Q} \equiv \frac{1}{\tau}, \qquad Q \equiv \frac{\omega L}{R}, \tag{16}$$

where Q is the resonator quality factor and τ is called the circuit ringing time. Define the reduced electromotive force

$$h \equiv \frac{cE_f}{nA_c\gamma}. \tag{17}$$

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

$$\frac{dH}{dt} + 2\gamma H + \omega^2 \int_0^t H(t')\, dt' = \gamma h - 4\pi\eta \frac{dm_x}{dt} \tag{18}$$

for the feedback magnetic field created by the coil.

The feedback equation (18) can be presented in another equivalent form that proved to be very convenient for solving the evolution equations [55–60]. For this purpose, by involving the method of Laplace transforms and employing the transfer function

$$G(t) = \left(\cos \omega' t - \frac{\gamma}{\omega'} \sin \omega' t \right) \mathrm{e}^{-\gamma t} \qquad \left(\omega' \equiv \sqrt{\omega^2 - \gamma^2} \right), \tag{19}$$

we find the integral representation

$$H = \int_0^t G(t - t') \left[\gamma h(t') - 4\pi\eta \dot{m}_x(t') \right] dt', \tag{20}$$

where the overdot means, as usual, time derivative.

3. NUCLEAR SPIN WAVES

In order to understand the nature of the mechanisms triggering the spin motion, it is necessary to pay attention to the possibility of arising nuclear spin waves. The latter are known to exist in ferromagnets, where ferromagnetism is caused by electron spins [54]. To produce spin waves, nuclear spins should be somehow ordered, at least locally. The dipolar interactions (4), by themselves, usually are not able to order nuclear spins. However, in the presence of a sufficiently strong external magnetic field, typical of nuclear magnetic resonance experiments, some kind of the intermediate range ordering may appear among nuclear spins. This midrange ordering can be observed by different NMR techniques [67].

The situation with nuclear spins, forming a paramagnetic matter, seems to be analogous to what happens in paramagnetic systems of electron spins. Therefore, it is useful to make a retrospective journey into the problem of spin waves in electron paramagnetic assemblies. This may better explain the physics of the similar effects in nuclear spin samples.

The theory of electron spin waves in nonmagnetic metals was developed by Silin [68, 69] on the basis of the Landau Fermi-liquid phenomenological picture [70, 71]. Good surveys of the Silin theory can be found in [72–74]. Spin waves in nonmagnetic metals can exist only in the presence of an external magnetic field, and do not exist when this field is absent. To be well defined, spin waves require that the external magnetic field be sufficiently strong, so that the spin resonance frequency be larger than the spin-wave attenuation. Spin waves, in the field of about 10^3 G, were observed in several paramagnetic metals [75–78], such as Na, K, Rb, Cs, and Al. Calculation of electro-magnetic fields transmitted through metallic films by spin waves involves the usage of boundary conditions defined by the type of spin scattering on the surface of metals. Several kinds of these conditions were employed in the theory of electron spin resonance [79–87]. The Dyson boundary condition [79] was used for studying the influence of the surface magnetic anisotropy on the amplitudes of excited spin waves and on the transition coefficient under spin-wave resonance [89–91]. Actually, experiments on spin resonance cannot distinguish between different boundary conditions [85]. Therefore, the usage of the simple Dyson condition for calculating the signals transmitted by spin waves [89–91] was justified. The surface impedance of a semibounded metal was expressed through the characteristics of infinite matter [90]. The possibility of exciting spin waves in a semibounded paramagnetic sample was advanced [92, 93]. Thus, electron spin waves do exist in paramagnetic metals, provided that a sufficiently strong external magnetic field is applied. All this sets us thinking that an external magnetic field could also ensure the appearance of nuclear spin waves in paramagnetic nuclei assemblies.

Let us consider a paramagnetic system of nuclear spins, described by the Hamiltonian (1). The spin operators satisfy the Heisenberg equations of motion

$$i\hbar\frac{dI_i^\alpha}{dt} = \left[I_i^\alpha, \hat{H}\right]$$

and the commutation relations

$$\left[I_i^+, I_j^-\right] = 2\delta_{ij}I_i^z, \qquad \left[I_i^z, I_j^\pm\right] = \pm\delta_{ij}I_i^\pm.$$

To write down the evolution equations in an explicit way, it is convenient to introduce the notation

$$\begin{aligned} \xi_0 &\equiv \frac{1}{\hbar}\sum_{j(\neq i)}\left(a_{ij}I_j^z + c_{ij}^* I_j^- + c_{ij}I_j^+\right), \\ \xi &\equiv \frac{i}{\hbar}\sum_{j(\neq i)}\left(2c_{ij}I_j^z - \frac{1}{2}a_{ij}I_j^- + 2b_{ij}I_j^+\right) \end{aligned} \tag{21}$$

for the *local fields* acting on a spin from the side of other spins. For short, we do not label the local fields (21) by an index i. Also, define the *effective force*

$$f \equiv -\frac{i}{\hbar}\mu_0(B_1 + H) + \xi. \tag{22}$$

Then the evolution equations for the spin operators can be presented in the form

$$\begin{aligned} \frac{dI_i^-}{dt} &= i\left(\frac{1}{\hbar}\mu_0 B_0 - \xi_0\right)I_i^- + fI_i^z, \\ \frac{dI_i^z}{dt} &= -\frac{1}{2}\left(f^+ I_i^- + I_i^+ f\right). \end{aligned} \tag{23}$$

Denoting the statistical average of an operator I_i^α as $\langle I_i^\alpha\rangle$, we may write the identity

$$I_i^\alpha \equiv \langle I_i^\alpha\rangle + \delta I_i^\alpha, \qquad \delta I_i^\alpha \equiv I_i^\alpha - \langle I_i^\alpha\rangle, \tag{24}$$

where δI_i^α describes an operator deviation from the average value $\langle I_i^\alpha\rangle$. Also, without the loss of generality, we may assume that the averages

$$\langle I_i^\alpha\rangle = \langle I_j^\alpha\rangle \tag{25}$$

do not depend on the indices i, j, thus, transferring this dependence to the deviation term δI_i^α. Then, because of Eqs. (10), the local fields (21) become

$$\xi_0 = \frac{1}{\hbar}\sum_{j(\neq i)}\left(a_{ij}\delta I_j^z + c_{ij}^*\delta I_j^- + c_{ij}\delta I_j^+\right),$$

$$\xi = \frac{i}{\hbar} \sum_{j(\neq i)} \left(2c_{ij} \delta I_j^z - \frac{1}{2}\, a_{ij} \delta I_j^- + 2b_{ij} \delta I_j^+ \right) . \tag{26}$$

This better clarifies the meaning of the local fields $\xi_0 = \delta\xi_0$, and $\xi = \delta\xi$, actually, corresponding to *local field fluctuations*.

To show that an external magnetic field B_0 can support the appearance of nuclear spin waves, let us consider a stationary situation, when $B_0 = \mathrm{const}$ and $B_1 = H = 0$, so that there is a nonzero polarization $\langle I_i^z \rangle \neq 0$, but $\langle I_i^{\pm} \rangle = 0$. In such a case, $I_i^{\pm} = \delta I_i^{\pm}$. Equating in Eqs. (23) the terms linear with respect to spin deviations, we have

$$\frac{d}{dt} I_i^- = \frac{i}{\hbar} \mu_0 B_0 I_i^- + \langle I_i^z \rangle \xi, \qquad \frac{d}{dt} \delta I_i^z = 0. \tag{27}$$

Since $\delta I_i^z = \mathrm{const}$, we may set, by accepting the zero initial condition, that $\delta I_i^z = 0$. For the raising and lowering spin operators, we employ the Fourier transforms

$$I_j^{\pm} = \sum_k I_k^{\pm} \, \exp\left(\mp i \mathbf{k} \cdot \mathbf{r}_j \right) , \qquad I_k^{\pm} = \frac{1}{N} \sum_j I_j^{\pm} \, \exp\left(\pm i \mathbf{k} \cdot \mathbf{r}_j \right) . \tag{28}$$

Strictly speaking, the latter are exactly valid only if all spins were located in sites of an ideal crystalline lattice. When the studied spin sample does not form such an ideal lattice, transformation (28) can be treated as approximate. In that case, the summation over spins should be limited by a finite number of them inside a volume, with an effective size L_{eff}^3, where the matter can be regarded as approximately arranged in a lattice. The length L_{eff} is an effective linear size of uniformity.

Introduce the transforms

$$a_k \equiv \sum_{j(\neq i)} a_{ij} \, \exp\left(-i \mathbf{k} \cdot \mathbf{r}_{ij} \right) , \qquad b_k \equiv \sum_{j(\neq i)} b_{ij} \, \exp\left(-i \mathbf{k} \cdot \mathbf{r}_{ij} \right) . \tag{29}$$

Since $a_{ij} = a_{ji}$ and it is real, a_k is also real. And, because of Eqs. (10), $a_0 = b_0 = 0$. Then, using Eqs. (28) and (29), we reduce the first of Eqs. (27) to the equation

$$\frac{d}{dt} I_k^- = -i \mu_k I_k^- + i \lambda_k I_k^+ , \tag{30}$$

in which

$$\mu_k \equiv \frac{a_k}{2\hbar} \langle I_i^z \rangle - \frac{1}{\hbar} \mu_0 \, B_0 , \qquad \lambda_k \equiv \frac{2b_k}{\hbar} \langle I_i^z \rangle . \tag{31}$$

The solution to Eq. (30) can be presented as

$$I_k^- = u_k \, \mathrm{e}^{-i\omega_k t} + v_k^* \, \mathrm{e}^{i\omega_k t} , \tag{32}$$

with the spectrum of excitations

$$\omega_k = \sqrt{\mu_k^2 - |\lambda_k|^2}. \tag{33}$$

The uniform excitation, corresponding to $k = 0$, yields

$$\omega_0 = \frac{1}{\hbar}|\mu_0 B_0|. \tag{34}$$

The procedure of defining the excitation spectrum, as is used above, is analogous to that employed for ferromagnets [94].

The spectrum (33) can describe nuclear spin waves if it is positive. The requirement $\omega_k > 0$ leads to the *stability condition*

$$|2\mu_0 B_0 - \langle I_i^z\rangle a_k| > 4\,|\langle I_i^z\rangle b_k|\,. \tag{35}$$

From here, it is evident that in a paramagnet without a polarization, when $\langle I_i^z\rangle = 0$, and without an external field, i.e., $B_0 = 0$, there are no nuclear spin waves. But if there exists a polarization and an external field, that is, $\langle I_i^z\rangle \neq 0$ and $B_0 \neq 0$, then condition (35) can be valid for a sufficiently strong external field B_0. Therefore, nuclear spin waves are possible in a paramagnet placed in a magnetic field.

In the long-wave limit, the spectrum (33) is given by the equation

$$\omega_k^2 \simeq \omega_0^2 + \langle I_i^z\rangle \frac{\mu_0 B_0}{2\hbar^2} \sum_{j(\neq i)} a_{ij}\,(\mathbf{k}\cdot\mathbf{r}_{ij})^2\,, \tag{36}$$

where $k \to 0$. In the interval

$$L_{\rm eff}^{-1} \ll k \ll a^{-1}, \tag{37}$$

where $L_{\rm eff}$ is an effective length of uniformity and a is the mean distance between spins, one can limit himself by the summation over the nearest neighbours only. As an illustration, let us assume cubic symmetry, when there are six nearest neighbours. Then the tensors, defined in Eq. (9), are

$$\{a_{ij}\} = \frac{\mu_0^2}{a^3}\{1,1,1,1,-2,-2\}, \qquad \{b_{ij}\} = \frac{3\mu_0^2}{4a^3}\{-1,-1,1,1,0,0\}, \tag{38}$$

while $c_{ij} = 0$. The Fourier transforms (29) become

$$a_k = \frac{2\mu_0^2}{a^3}(\cos\,k_x a + \cos\,k_y a - 2\cos\,k_z a), \qquad b_k = -\frac{3\mu_0^2}{2a^3}(\cos\,k_x a - \cos\,k_y a),$$

which, in the case of Eq. (37), gives

$$a_k \simeq -\frac{\mu_0^2}{a}\left(k_x^2 + k_y^2 - 2k_z^2\right), \qquad b_k \simeq \frac{3\mu_0^2}{4a}\left(k_x^2 - k_y^2\right). \tag{39}$$

In this manner, Eq. (36) results in the spectrum equation

$$\omega_k^2 \simeq \omega_0^2 + \langle I_i^z \rangle \frac{\mu_0^3 B_0}{a\hbar^2} \left(k_x^2 + k_y^2 - 2k_z^2 \right) . \tag{40}$$

The spectrum ω_k, to be positive at the maximal wave vector $k = a^{-1}$, requires that

$$\frac{2\mu_0^2 I}{\hbar a^3 \omega_0} < 1.$$

In the standard situation, when $\mu_0^2/\hbar a^3 \sim 10^5$ s^{-1} and $\omega_0 \sim 10^8$ Hz, this inequality holds true. The attenuation of spin waves is of order $\Gamma_2 \sim \mu_0^2/\hbar a^3 \sim 10^5$ s^{-1}, hence it is much less than $\omega_0 \sim 10^8$ Hz. Thus, nuclear spin waves are well defined.

It is necessary to stress the importance of an external magnetic field for the stability of nuclear spin waves. Really, setting $B_0 = 0$ in condition (35), we have $|a_k| > 4|b_k|$. For a cubic structure with the wave vectors in the interval (37), this yields

$$\left| k_x^2 + k_y^2 - 2k_z^2 \right| > 3 \left| k_x^2 - k_y^2 \right| .$$

This inequality should be valid for all $\mathbf{k} = \{k_x, k_y, k_z\}$ from the given interval. However, as is evident, the inequality does not hold if either $k_x = k \equiv |\mathbf{k}|$ or $k_y = k$. Hence, these nuclear spin waves are not stable without an external magnetic field.

One could remember that at very low temperatures, of order $T \sim 10^{-7}$ K, nuclear spins interacting through dipolar forces can become magnetically ordered [23]. Then in a spin system with long-range magnetic order, there should appear spin waves, even without external fields. This, really, may happen, but only for those structures that are able to support nuclear spin waves. Generally, a stable structure is characterized by two types of stability, thermodynamic and dynamic [95]. The structure is thermodynamically stable, when its thermodynamic potential is extremal. For instance, when free energy is minimal. And the structure is dynamically stable if its spectrum of collective excitations is non-negative. This is also true for magnetic structures formed by dipolar spin systems. Only those magnetically ordered structures will survive, which possess the minimal free energy and are stable against arising spin waves.

As has been demonstrated above, nuclear spin waves may exist even when there is no long-range magnetic order caused by internal forces, but provided that there is a sufficiently strong external magnetic field. The latter can stabilize nuclear spin waves for any kind of crystalline symmetry. Moreover, the spin sample can be polycrystalline, being composed of many small crystals, or even it can be amorphous. Then not all space of the sample will be filled by a coherent system of spin waves. But the whole sample will be separated into regions, inside each of which spin waves are coherent, though are not coherent between different

regions. The effective linear size of such regions, where spins can form coherent spin waves, $L_{\rm eff}$, can be called the uniformity length. If spin waves arise inside the spatial regions of volume $L_{\rm eff}^3$ which is much smaller than the total sample volume, then such waves may be termed *local spin waves*. The coherence length $L_{\rm eff}$ must be much larger than the mean interspin distance in order that spin waves could arise, though this length may be smaller than the linear size of the total sample.

4. SCALE SEPARATION APPROACH

To study nonstationary spin dynamics, we have to deal with the general equations of motion (23). In order to analyze these equations, we invoke the scale separation approach [9, 42, 56, 57] consisting of three main steps: randomization of local fields; classification of relative quasi-invariants; and method of stochastic averaging.

Let us recall that there exist the operator constructions ξ_0 and ξ, playing the role of local fields (21) or, according to Eq. (26), of local field fluctuations. We may consider these local fields as a separate kind of operators. Since these fields describe local fluctuations, they can be modelled by random variables [12, 53]. In this way, we have two types of variables characterizing the system, spin operators $I_i^-,\ I_i^+,\ I_i^z$, and random fields $\xi_0,\ \xi,\ \xi^*$. The former are responsible for long-range global phenomena; while the latter, for short-range local fluctuations. Having defined two types of variables, we may introduce two sorts of averaging with respect to the corresponding variables. Thus, the statistical averaging over spin operators will be denoted by the single angle brackets $\langle \ldots \rangle$ and the averaging over random local fields will be denoted by means of the double angle brackets $\langle\langle \ldots \rangle\rangle$. This modelling of local fluctuating fields by random variables is the *randomization of local fields*. To make the method self-consistent, it is necessary to define the stochastic averages over the random fields. Treating the latter as white noise, we define

$$\begin{aligned} \langle\langle \xi_0(t) \rangle\rangle = \langle\langle \xi(t) \rangle\rangle = 0, \qquad \langle\langle \xi_0(t) \xi_0(t') \rangle\rangle = 2\Gamma_3 \delta(t-t'), \\ \langle\langle \xi_0(t) \xi(t') \rangle\rangle = \langle\langle \xi(t) \xi(t') \rangle\rangle = 0, \quad \langle\langle \xi^*(t) \xi(t') \rangle\rangle = 2\Gamma_3 \delta(t-t'), \end{aligned} \tag{41}$$

where Γ_3 is the width of *dynamic broadening*, which is the inhomogeneous broadening caused by the local field fluctuations. This broadening is similar to that arising in optical resonant systems because of dipole-dipole interactions through the exchange of transversely polarized photons [96].

Averaging over spin operators, we could employ the decoupling

$$\langle I_i^\alpha\ I_j^\beta \rangle = \langle I_i^\alpha \rangle \langle I_j^\beta \rangle \qquad (i \neq j). \tag{42}$$

This looks as the mean-field approximation. However, we should remember that the averaging, denoted by the single brackets $\langle \ldots \rangle$, by definition does not involve the stochastic variables. Therefore the quantum correlations are not lost in the decoupling (42) but are preserved because of the dependence of the averages $\langle I_i^\alpha \rangle$ on the random variables ξ_0 and ξ. This decoupling (42) may be termed the *stochastic mean-field approximation* [9]. Moreover, after having agreed to treat the spin operators and the local fields (21) as different variables, we do not often need the explicit usage of the decoupling (42). This will be used only once in the definition of coherence intensity.

Let us average the operator equations (23) over the spin operators, without touching the local random fields (21). And let us introduce the following definitions. The *transition function*

$$u \equiv \frac{1}{IN} \sum_{i=1}^{N} \langle I_i^- \rangle \tag{43}$$

describes the average rotation of transverse spin components. The *coherence intensity*

$$w \equiv \frac{1}{I^2 N(N-1)} \sum_{i \neq j}^{N} \langle I_i^+ I_j^- \rangle \tag{44}$$

shows the degree of coherence in the spin motion. And the *spin polarization*

$$s \equiv \frac{1}{IN} \sum_{i=1}^{N} \langle I_i^z \rangle \tag{45}$$

defines the average polarization in the system. Note that, under the validity of the decoupling (42), and for $N \gg 1$, we have $w = |u|^2$, with u given in Eq. (43).

Assume that the longitudinal magnetic field B_0 is constant and directed so that

$$\mu_0 B_0 < 0. \tag{46}$$

For nuclei with $\mu_0 > 0$, this means that $B_0 < 0$. Conversely, if $\mu_0 < 0$, then $B_0 > 0$. To allow for the influence of lattice, account must be taken of spin-lattice relaxation with a related relaxation parameter $\Gamma_1 \equiv T_1^{-1}$. And to take into account the spin-spin attenuation, we include the corresponding transverse width $\Gamma_2 \equiv T_2^{-1}$.

In this way, for the variables (43) to (45), we obtain the evolution equations

$$\begin{gathered} \frac{du}{dt} = -i\,(\omega_0 + \xi_0 - i\Gamma_2)\,u + fs, \qquad \frac{dw}{dt} = -2\Gamma_2 w + (u^* f + f^* u)\,s, \\ \frac{ds}{dt} = -\frac{1}{2}\,(u^* f + f^* u) - \Gamma_1 (s - \zeta), \end{gathered} \tag{47}$$

where ω_0 is the Zeeman frequency (34) and $\zeta \in [-1,1]$ is an average spin polarization. In the absence of pumping, $\zeta = -1$. But if there is a kind of pumping, for instance, by means of dynamic nuclear polarization, then $\zeta > -1$.

Equations (47) are stochastic differential equations, containing the stochastic variables ξ_0 and ξ. The presence of the latter will make it possible to consider quantum effects. Also, Eqs. (47) are nonlinear due to the resonator feedback field entering through the effective force (22). The resonator field is defined by the integral presentation (20), in which the magnetization density (13) writes

$$m_x = \frac{1}{2}\rho\mu_0 I(u^* + u) \qquad \left(\rho \equiv \frac{N}{V}\right). \tag{48}$$

Thus, the spin evolution is characterized by the system of stochastic nonlinear integro-differential equations (20), (22), (47), and (48). These equations are assumed to be complimented by the initial conditions $u(0) = u_0$, $w(0) = w_0$, and $s(0) = s_0$.

To proceed further, we notice that there exist several small parameters. Thus, the spin-lattice and spin-spin attenuations are usually small as compared to the Zeeman frequency, similarly to the dynamic broadening,

$$\frac{\Gamma_1}{\omega_0} \ll 1, \qquad \frac{\Gamma_2}{\omega_0} \ll 1, \qquad \frac{\Gamma_3}{\omega_0} \ll 1. \tag{49}$$

The last inequality means that the influence of local random fields can be treated as weak, since the stochastic averages (41) are proportional to Γ_3. The interaction energy of magnetic moments $\mu_n \equiv \hbar\gamma_n I$ with the resonator field is proportional to the *natural width*

$$\Gamma_0 \equiv \frac{\pi}{\hbar}\eta\rho\mu_0^2 I = \pi\eta\rho\gamma_n\mu_n, \tag{50}$$

which, being of order Γ_2, is also small with respect to ω_0. And the resonant circuit is assumed to be of good quality, i.e., $Q \gg 1$, because of which the resonator ringing width γ is much smaller than the circuit natural frequency ω. Hence, there are two other small parameters

$$\frac{\Gamma_0}{\omega_0} \ll 1, \qquad \frac{\gamma}{\omega} \ll 1. \tag{51}$$

Specifying the external transverse magnetic field, entering the total field (3), we take

$$B_1 = h_0 + h_1 \cos \omega t, \tag{52}$$

where $h_0 = \mathrm{const}$. And let the resonant part of the reduced electromotive force (17) be

$$h = h_2 \cos \omega t. \tag{53}$$

Defining the quantities

$$\nu_0 \equiv \frac{\mu_0 h_0}{\hbar}, \qquad \nu_1 \equiv \frac{\mu_0 h_1}{2\hbar}, \qquad \nu_2 \equiv \frac{\mu_0 h_2}{2\hbar}, \tag{54}$$

we keep in mind that the amplitudes of all transverse fields (52) as well as (53) are small, in the sense that

$$\frac{|\nu_0|}{\omega_0} \ll 1, \qquad \frac{|\nu_1|}{\omega_0} \ll 1, \qquad \frac{|\nu_2|}{\omega_0} \ll 1. \tag{55}$$

The existence of the small parameters (49), (51), and (55) allows us to realize the *classification of relative quasi-invariants*. From Eqs. (47) we infer that the transition function (43) has to be considered as fastly varying in time, as compared to the slow functions (44) and (45). This implies that w and s are *temporal quasi-invariants* with respect to u. Fast variation means that u varies with a frequency of order ω_0 or ω. These two frequencies are supposed to be close to each other, satisfying the *resonance condition*

$$\frac{\Delta}{\omega} \ll 1, \qquad \Delta \equiv \omega - \omega_0, \tag{56}$$

which can be always done by tuning the natural resonator frequency to the Zeeman frequency.

The occurrence of the small parameters, listed above, allows us to solve the integral resonator equation (20) by an iteration procedure, starting with the solution of the first of Eqs. (47) taken in zero order with respect to small parameters, that is, substituting in the right-hand integral of Eq. (20) the form $u \simeq u_0 \exp(-i\omega_0 t)$. Accomplishing the integration gives the first-order approximant

$$\frac{\mu_0 H}{\hbar} = i(\alpha u - \alpha^* u^*) + 2\beta \cos \omega t, \tag{57}$$

in which $\alpha = \alpha(t)$ is the coupling function of spins with the resonator feedback field,

$$\alpha = \frac{\Gamma_0 \omega_0}{\gamma + i\Delta} \left(1 - \mathrm{e}^{-i\Delta t - \gamma t}\right), \tag{58}$$

and $\beta = \beta(t)$ is the coupling function of spins with the electromotive force,

$$\beta = \frac{\nu_2}{2} \left(1 - \mathrm{e}^{-\gamma t}\right). \tag{59}$$

The resonance condition (56) is used in deriving Eqs. (57) with (58).

An important quantity that appears is the *spin-resonator coupling*

$$g \equiv \frac{\gamma \Gamma_0 \omega_0}{\Gamma_2(\gamma^2 + \Delta^2)}. \tag{60}$$

This parameter enters the real and imaginary parts of the coupling function (58), so that

$$\mathrm{Re}\,\alpha = g\Gamma_2\left[1 - \left(\cos\,\Delta t - \frac{\Delta}{\gamma}\,\sin\,\Delta t\right)\mathrm{e}^{-\gamma t}\right],$$

$$\mathrm{Im}\,\alpha = -g\Gamma_2\left[\frac{\Delta}{\gamma} - \left(\sin\,\Delta t + \frac{\Delta}{\gamma}\,\cos\,\Delta t\right)\mathrm{e}^{-\gamma t}\right].$$

These formulas can be simplified if the resonance is sharp, such that

$$\frac{|\Delta|}{\gamma} = 2\frac{|\Delta|}{\omega}Q \ll 1. \tag{61}$$

Then the coupling function (58) becomes

$$\alpha = g\Gamma_2\left(1 - \mathrm{e}^{-\gamma t}\right). \tag{62}$$

This simplification is not principal but just helps us to avoid cumbersome expressions.

Taking into account in the effective force (22) the expression (52) for the transverse magnetic field and Eq. (57) for the resonator field, we define

$$f_1 \equiv -i\nu_0 - 2i(\nu_1 + \beta)\,\cos\,\omega t + \xi. \tag{63}$$

Then Eqs. (47) can be transformed to the system of equations

$$\begin{aligned}
\frac{du}{dt} &= -i(\omega_0 + \xi_0)u - (\Gamma_2 - \alpha s)u + f_1 s - \alpha s u^*,\\
\frac{dw}{dt} &= -2(\Gamma_2 - \alpha s)w + (u^* f_1 + f_1^* u)s - 2\alpha s\,\mathrm{Re}\,u^2,\\
\frac{ds}{dt} &= -\alpha w - \frac{1}{2}\,(u^* f_1 + f_1^* u) - \Gamma_1(s - \zeta) + \alpha\,\mathrm{Re}\,u^2.
\end{aligned} \tag{64}$$

Recall that, according to the classification of relative quasi-invariants, the function u is considered as fast, being compared with the functions w and s. The latter are temporal quasi-invariants with respect to u. The time derivatives of the coupling functions (59) and (62) are proportional to γ, because of which α and β are also quasi-invariants with respect to u.

Following further the scale separation approach [9, 42, 56, 57], we may solve the system of equations (64) by the *method of stochastic averaging*, which is a generalization of the averaging technique [43] to stochastic and partial differential equations. First, we solve the equation for the fast variable u from Eqs.(64), with all quasi-invariants kept fixed. This yields

$$\begin{aligned}
u = u_0 \exp\left\{-(i\omega_0 + \Gamma_2 - \alpha s)t - i\int_0^t \xi_0(t')\,dt'\right\} +\\
+ s\int_0^t f_1(t)\exp\left\{-(i\omega_0 + \Gamma_2 - \alpha s)(t - t') - i\int_{t'}^t \xi_0(t'')\,dt''\right\}dt'.
\end{aligned} \tag{65}$$

Then, we substitute the solution (65) into the equations for the slow functions w and s, which are the second and third of Eqs. (64). After this, we average the resulting equations over time in the infinite interval, keeping the quasi-invariant fixed, and over the stochastic fields, employing the averages (41). To slightly simplify the resulting equations, we take the initial condition for the transition function u in the real form

$$u_0 = \frac{1}{I}\langle I_i^x(0)\rangle. \tag{66}$$

Note that the counter-rotating term, containing u^*, in the first of Eqs. (64), gives a small correction to the solution (65), of order Γ_2/ω_0. Hence, this negligible correction is omitted. In accomplishing the averaging of the second and third of Eqs. (64), the nonresonant terms, proportional to $\mathrm{Re}\, u^2$, also give small contributions to the right-hand sides of these averaged equations. Taking all this into account, we could omit in Eqs. (64) the last terms and also could take the force (63) in the form

$$f_1 = -i\nu_0 - i(\nu_1 + \beta)\, \mathrm{e}^{-i\omega t} + \xi.$$

This is a kind of the rotating-wave approximation or, in other words, the resonance approximation.

Recall that, when averaging the right-hand sides of Eqs. (64), we keep all quasi-invariants fixed. The function $\exp(-\Gamma t)$ is also considered as a quasi-invariant. The stochastic average of Eq. (65) is

$$\langle\langle u\rangle\rangle = -\frac{\nu_0 s}{\omega_0 - i\Gamma} + \frac{(\nu_1+\beta)s}{\Delta + i\Gamma}\, \mathrm{e}^{-i\omega t} + \\ + \left[u_0 + \frac{\nu_0 s}{\omega_0 - i\Gamma} - \frac{(\nu_1+\beta)s}{\Delta + i\Gamma}\right] \mathrm{e}^{-(i\omega_0+\Gamma)t}, \tag{67}$$

where the *collective width*

$$\Gamma \equiv \Gamma_2 + \Gamma_3 - \alpha s \tag{68}$$

is introduced. Let us also define the *effective attenuation*

$$\tilde{\Gamma} \equiv \Gamma_3 + \frac{\nu_0^2\Gamma}{\omega_0^2 + \Gamma^2} - \frac{\nu_0(\nu_1+\beta)\Gamma}{\omega_0^2+\Gamma^2}\, \mathrm{e}^{-\Gamma t} + \frac{(\nu_1+\beta)^2\Gamma}{\Delta^2+\Gamma^2}\left(1 - \mathrm{e}^{-\Gamma t}\right). \tag{69}$$

Following the described procedure of averaging the second and third of Eqs. (64), we come to the equations for the *guiding centres*,

$$\frac{dw}{dt} = -2(\Gamma_2 - \alpha s)w + 2\tilde{\Gamma}s^2, \qquad \frac{ds}{dt} = -\alpha w - \tilde{\Gamma}s - \Gamma_1(s-\zeta). \tag{70}$$

These are the main evolution equations describing the nonlinear dynamics of nuclear spin motion. The exponential factors entering the effective attenuation

(69) characterize retardation effects that are always present in real processes and, hence, are to be taken into account. This retardation may essentially influence the spin dynamics.

5. INCOHERENT QUANTUM STAGE

One of the most interesting questions in the theory of spin superradiance is: What initiates the motion of spins when no transition coherence is imposed on the system at $t = 0$ and no external fields push spins in the transverse direction? That is, what is the origin of pure spin superradiance? In other words, how does the coherence in spin motion develop from initially uncorrelated spin fluctuations?

Let us note that here we are interested in the transition coherence that is related to the arising coherent radiation. In general, one may distinguish two types of coherence, state coherence and transition coherence [60]. A spin ensemble possesses *state coherence*, when it is prepared in a spin polarized state, with a nonzero spin polarization (45). And *transition coherence* develops when the transition function (43) is nonzero, or the coherence intensity (44) becomes noticeable. These functions (43) to (45) may be considered as *dynamical order parameters* [97] characterizing the level of state coherence or transition coherence. If the spin sample is initially polarized, it possesses state coherence. But if no external fields are thrust upon the system, there is no transition coherence. An intriguing question is: How does the transition coherence develop from the state coherence in a self-organized way?

It is well known how transition coherence appears in a system of inverted atoms [4, 5]. The relaxation begins with atomic spontaneous radiation, which is a quantum incoherent process. After the appearance of the seed radiation field, atomic correlations start arising through the interatomic photon exchange. Transition coherence develops as soon as the interatomic correlations become sufficiently intensive. Then the quantum stage of spontaneous emission changes for the coherent stage, resulting in atomic superradiance. Since moving spins produce magnetodipole radiation, one may ask if the latter can be the cause of collective spin motion, by analogy with what happens in atomic systems.

5.1. Radiation of Magnetic Dipoles. The vector potential created by radiating objects is

$$\mathbf{A}_{\rm rad}(\mathbf{r},t) = \frac{1}{c}\int \mathbf{j}\left(\mathbf{r}', t - \frac{1}{c}|\mathbf{r}-\mathbf{r}'|\right)\frac{d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}, \tag{71}$$

where the density of current, formed by the magnetization density, writes

$$\mathbf{j}(\mathbf{r},t) \equiv c\boldsymbol{\nabla}\times\mathbf{m}(\mathbf{r},t). \tag{72}$$

Let this current be produced by $N_{\rm eff}$ spins in the volume $L_{\rm eff}^3$. These spins can act as an altogether only if the radiation wavelength is much larger than $L_{\rm eff}$, so

that

$$k_0 L_{\rm eff} \ll 1 \qquad \left(k_0 \equiv \frac{\omega_0}{c} \right) . \tag{73}$$

The self-action of a radiating spin corresponds to $N_{\rm eff} = 1$, with $L_{\rm eff}$ being the nucleus radius.

Under condition (73), the vector potential (71) can be presented as an expansion

$$\mathbf{A}_{\rm rad}(\mathbf{r},t) \simeq \frac{1}{c} \int \left(\frac{1}{x} - \frac{1}{c}\frac{\partial}{\partial t} + \frac{x}{2c^2}\frac{\partial^2}{\partial t^2} - \frac{x^2}{6c^3}\frac{\partial^3}{\partial t^3} \right) \mathbf{j}(\mathbf{r}+\mathbf{x},t)\, d\mathbf{x} \tag{74}$$

in which $\mathbf{x} \equiv \mathbf{r}' - \mathbf{r}$ and $x \equiv |\mathbf{x}|$. Assuming that there is no current through the surface of the volume $L^3_{\rm eff}$, for the magnetic field $\mathbf{H}_{\rm rad} \equiv \boldsymbol{\nabla} \times \mathbf{A}_{\rm rad}$, we have

$$\mathbf{H}_{\rm rad}(\mathbf{r},t) = \frac{1}{c} \int \left(\frac{1}{x^3} - \frac{1}{2xc^2}\frac{\partial^2}{\partial t^2} + \frac{1}{2c^3}\frac{\partial^3}{\partial t^3} \right) \left[\mathbf{x} \times \mathbf{j}(\mathbf{r}+\mathbf{x},t) \right] d\mathbf{x}. \tag{75}$$

The latter, involving Eq. (72) and defining the total magnetization

$$\mathbf{M} \equiv \frac{1}{2c} \int \mathbf{r} \times \mathbf{j} d\mathbf{r} = \int \mathbf{m} d\mathbf{r}, \tag{76}$$

can be transformed to

$$\mathbf{H}_{\rm rad}(\mathbf{r},t) = \int \left[\frac{3(\mathbf{m}\cdot\mathbf{x})\mathbf{x}}{x^5} - \frac{\mathbf{m}}{x^3} \right] d\mathbf{x} - \frac{1}{2c^2} \int \left[\frac{(\ddot{\mathbf{m}}\cdot\mathbf{x})\mathbf{x}}{x^3} + \frac{\ddot{\mathbf{m}}}{x} \right] d\mathbf{x} + \frac{2}{3c^3}\frac{d^3\mathbf{M}}{dt^3}, \tag{77}$$

where the overdot implies the time differentiation and $\mathbf{m} = \mathbf{m}(\mathbf{r}+\mathbf{x},t)$.

Note that this way of presenting the magnetic field created by a system of radiating magnetic dipoles goes back to Ginzburg [98] and since then has been considered by many authors (see, e.g., [99, 100]). The first term in Eq. (77) describes a dipolar demagnetizing field, depending on the shape of the sample. For the spherical shape, or for a sufficiently large volume, this term is zero, similarly to the first of Eqs. (6). The second integral in Eq. (77) can be approximately presented [100] as

$$\int \left[\frac{(\ddot{\mathbf{m}}\cdot\mathbf{x})\mathbf{x}}{x^3} + \frac{\ddot{\mathbf{m}}}{x} \right] d\mathbf{x} \cong \frac{8}{3\pi L_{\rm eff}} \frac{d^2\mathbf{M}}{dt^2}.$$

Then one has

$$\mathbf{H}_{\rm rad} = -\frac{4}{3\pi c^2 L_{\rm eff}} \ddot{\mathbf{M}} + \frac{2}{3c^3}\frac{d^3\mathbf{M}}{dt^3}. \tag{78}$$

Here the term with three time derivatives corresponds to the so-called electromagnetic friction. Generally, such terms with odd number of time derivatives

lead to some kind of relaxation in the spin motion. For example, the term $\dot{\mathbf{M}}$ in the Landau–Lifshitz equation [101] can be connected [102] with spin relaxation due to spin-phonon interactions.

The radiation field (78), in which

$$\mathbf{M} = \mu_0 \sum_{i=1}^{N_{\rm eff}} \langle \mathbf{I}_i \rangle, \tag{79}$$

has to be added to the total magnetic field (3). Then in the Zeeman term (2) there appears the additional interaction $-\mu_0 \mathbf{H}_{\rm rad} \cdot \mathbf{I}_i$. Wishing to concentrate the consideration on the role of the radiation field (78), we assume that there are no transverse fields, that is, $B_1 = 0$ and $H = 0$. Then the Zeeman interaction (7) can be written as

$$\hat{H}_i = -\mu_0 \left(B_0 + H^z_{\rm rad} \right) I^z_i - \frac{1}{2} \mu_0 \left(H^+_{\rm rad} I^-_i + I^+_i H^-_{\rm rad} \right), \tag{80}$$

where $H^{\pm}_{\rm rad} \equiv H^x_{\rm rad} \pm i H^y_{\rm rad}$ is expressed through Eq. (78) and the magnetization components

$$M^- = \mu_n N_{\rm eff} u, \qquad M^z = \mu_n N_{\rm eff} s,$$

in which M^+ is the complex conjugate of M^- and $\mu_n \equiv \mu_0 I = \hbar \gamma_n I$. Introducing the notation for the *radiation width*

$$\Gamma_{\rm rad} \equiv \frac{2}{3} k_0^3 \gamma_n \mu_n N_{\rm eff}, \tag{81}$$

we may write

$$\frac{1}{\hbar} \mu_0 H^-_{\rm rad} = \frac{\Gamma_{\rm rad}}{\omega_0^3} \left(\frac{d^3 u}{dt^3} - \frac{2c}{\pi L_{\rm eff}} \ddot{u} \right), \quad \frac{1}{\hbar} \mu_0 H^z_{\rm rad} = \frac{\Gamma_{\rm rad}}{\omega_0^3} \left(\frac{d^3 s}{dt^3} - \frac{2c}{\pi L_{\rm eff}} \ddot{s} \right).$$

Now, the evolution equations for the functions (43) to (45) can again be presented in the form (47), in which ω_0 is to be replaced by $\omega_0 + \mu_0 H^z_{\rm rad}/\hbar$ and

$$f = -\frac{i}{\hbar} \mu_0 H^-_{\rm rad}.$$

Keeping in mind the existence of the small parameter

$$\frac{\Gamma_{\rm rad}}{\omega_0} \ll 1, \tag{82}$$

we can find the radiation field (78) by iterating its right-hand side with the zero-order approximation $u \simeq u_0 \exp(-i\omega_0 t)$ and $s \simeq s_0$. Then

$$f = \Gamma_{\rm rad} \left(1 - \frac{2i}{\pi k_0 L_{\rm eff}} \right) u$$

and $H^z_{\rm rad} = 0$. As a result, instead of Eqs. (47), we obtain

$$\begin{aligned} \frac{du}{dt} &= -i(\omega_0 + \delta\omega_0 + \xi_0 - i\Gamma_2)u + \Gamma_{\rm rad} s u, \\ \frac{dw}{dt} = -2(\Gamma_2 - \Gamma_{\rm rad} s)w, &\qquad \frac{ds}{dt} = -\Gamma_{\rm rad} w - \Gamma_1(s - \zeta), \end{aligned} \tag{83}$$

with the frequency shift

$$\delta\omega_0 \equiv \frac{2\Gamma_{\rm rad} s}{\pi k_0 L_{\rm eff}}.$$

Equations (83) show that the magnetodipole radiation can lead to the arising collective effects only if the radiation width (81) is much larger than the transverse dephasing width Γ_2 due to dipole-dipole interactions. For the latter one has

$$\Gamma_2 = n_0 \rho \gamma_n \mu_n, \tag{84}$$

where n_0 is of the order of the nearest-neighbour number [12, 23, 53]. Comparing Eqs. (81) and (84), with taking account of $\rho = N_{\rm eff}/L^3_{\rm eff}$, we find

$$\frac{\Gamma_{\rm rad}}{\Gamma_2} = \frac{2}{3n_0} \left(k_0 L_{\rm eff}\right)^3 \ll 1, \tag{85}$$

since $n_0 \sim 10$ and $k_0 L_{\rm eff} \ll 1$ according to condition (73). Hence $\Gamma_{\rm rad} \ll \Gamma_2$ and, respectively, the radiation time $T_{\rm rad} \equiv \Gamma^{-1}_{\rm rad}$ is much larger than the dephasing time $T_2 \equiv \Gamma_2^{-1}$.

To estimate the related parameters, we may take the typical value $\omega_0 \sim 10^8$ Hz, so that $k_0 \sim 10^{-2}$ cm^{-1}. For protons, the gyromagnetic ratio $\gamma_n = 2.675 \cdot 10^4$ G$^{-1}\cdot$s^{-1} and spin $I = 1/2$. The proton magnetic moment $\mu_n = 2.793\mu_N$ can be expressed through the nuclear magneton $\mu_N = 5.051\times \times 10^{-24}$ erg$\cdot$G^{-1}. We have $\mu_n = 1.411 \cdot 10^{-23}$ erg$\cdot$G^{-1}. The dimension of Gauss is such that G^2 = erg$\cdot$cm^{-3}. In this way, $\Gamma_{\rm rad} \sim 10^{-25} N_{\rm eff}$ s^{-1}, while $\Gamma_2 \sim 10^5$ s^{-1} for $\rho \sim 10^{23}$ cm^{-3}. The relaxation caused by the self-action through magnetodipole radiation, when $N_{\rm eff} = 1$, yields the radiation time $T_{\rm rad} \sim 10^{25}$ s. This, with 1 year $\sim 10^7$ s, gives $T_{\rm rad} \sim 10^{18}$ y. Such an enormous time is not only much larger than $T_2 \sim 10^{-5}$ s, but also surpasses the Earth lifetime of $5\cdot 10^9$ y and is even longer than the Universe lifetime of 10^{10} y. Clearly, the self-action, caused by the magnetodipole radiation, is completely negligible. Even with the account of collective effects, when $N_{\rm eff} \sim 10^{23}$, we get $\Gamma_{\rm rad} \sim 10^{-2}$ s^{-1}, hence $T_{\rm rad} \sim 10^2$ s, which is much larger than T_2, their ratio being $T_{\rm rad}/T_2 \sim 10^7$. These estimates show that the ratio (85) is extremely small, $\Gamma_{\rm rad}/\Gamma_2 \leqslant 10^{-7}$.

Thus, we come to the conclusion that magnetodipole radiation is absolutely unable to organize the coherent motion of spins. This conclusion is based on the inequality (85), which is always valid, independently of the nature of the spins involved, whether these are nuclear, electron, atomic, or molecular spins.

5.2. Resonator Nyquist Noise. For many years, practically all researchers have used to write that the major cause producing spin motion, resulting in pure superradiance is the thermal Nyquist noise of the resonant electric circuit. This belief has been especially surprising because Bloembergen and Pound [7] mentioned that this noise cannot be a noticeable cause of spin relaxation. The common delusion about the principal role of the Nyquist noise was clarified in the papers [10, 55–57].

The role of this noise can be studied by analyzing the effective attenuation (69). Assume that there are no transverse external fields, so that $\nu_0 = \nu_1 = 0$, but there is only the electromotive force corresponding to the resonator Nyquist noise. Then the effective attenuation (69) is

$$\tilde{\Gamma} = \Gamma_3 + \Gamma_{\rm res}, \tag{86}$$

where, in accordance with Eq. (59),

$$\Gamma_{\rm res} = \frac{\nu_2^2 \Gamma}{4(\Delta^2 + \Gamma^2)} \left(1 - {\rm e}^{-\Gamma t}\right) \left(1 - {\rm e}^{-\gamma t}\right)^2 . \tag{87}$$

The latter attenuation is caused by the Nyquist noise of the resonant electric circuit. For concreteness, we shall keep in mind the standard situation, when the resonator width γ is larger or of the order of Γ_2 and Γ_3. And, for simplicity, we consider the case of good resonance, when condition (61) is valid. Then at short time, the attenuation (87) reads

$$\Gamma_{\rm res} \simeq \frac{1}{4} \nu_2^2 \gamma_2 t^3 \qquad (\gamma t \ll 1). \tag{88}$$

Notice that $\Gamma_{\rm res} = 0$ at $t = 0$.

The resonator electromotive force, entering the Kirchhoff equation (11), can be written as

$$E_f = E_{\rm res} \cos \omega t. \tag{89}$$

The amplitude of the reduced electromotive force (17) or (53) is h_2, for which we have

$$h_2 = \frac{cE_{\rm res}}{nA_c\gamma}, \qquad h_2^2 = \frac{8\pi\eta E_{\rm res}^2}{\gamma RV}.$$

The amplitude squared of the electromotive force (89) caused by the Nyquist noise, can be presented [18] as

$$E_{\rm res}^2 = \frac{\hbar\omega}{2\pi} \gamma R \coth \frac{\omega}{2\omega_T} \qquad \left(\omega_T \equiv \frac{k_B T}{\hbar}\right), \tag{90}$$

with ω_T being the thermal frequency. At radiofrequencies, one has $\omega \ll \omega_T$. Then Eq. (90) can be slightly simplified to

$$E_{\rm res}^2 \simeq \frac{\hbar}{\pi} \gamma R \omega_T \qquad \left(\frac{\omega}{\omega_T} \ll 1\right). \tag{91}$$

For the quantity ν_2, defined in Eq. (54), we get

$$\nu_2^2 = \frac{2\Gamma_0 E_{\rm res}^2}{\hbar\gamma IRN} \simeq \frac{2\Gamma_0\omega_T}{\pi IN}. \tag{92}$$

For the attenuation (88) at $t \approx 1/\gamma$, we find

$$\Gamma_{\rm res} \simeq \frac{\nu_2^2}{4\gamma} \simeq \frac{\Gamma_0\omega_T}{2\pi I\gamma N}. \tag{93}$$

This is, actually, the maximal value of the relaxation that can be reached by the attenuation (87).

To estimate Eq. (93), we again accept the typical values of $\Gamma_2 \sim \Gamma_0 \sim 10^5$ s^{-1} and $\gamma \sim 10^6$ s^{-1}. At temperature $T \sim 0.1$ K, one has $\omega_T \sim 10^{11}$ Hz. Thus, we obtain $\Gamma_{\rm res} \sim (10^{10}/N)$ s^{-1}. If $N \sim 10^{23}$, then $\Gamma_{\rm res} \sim 10^{-13}$ s^{-1}, which is much smaller than $\Gamma_3 \sim \Gamma_2$. The related time, during which the Nyquist noise could produce spin relaxation, $T_{\rm res} \equiv \Gamma_{\rm res}^{-1}$, is $T_{\rm res} \sim 10^{13}$ s $\sim 10^6$ y. This is many orders longer than $T_2 \sim T_3 \equiv \Gamma_3^{-1}$. Therefore, the resonator Nyquist noise plays no role in spin relaxation of macroscopic samples and can never serve as a triggering cause starting the spin motion, unless the number of spins is much smaller than 10^5.

5.3. Local Spin Fluctuations. We continue considering the case when there are no transverse external fields pushing spins, so that $\nu_0 = \nu_1 = 0$. As we have found out, neither the magnetodipole radiation, nor the resonator Nyquist noise are able to start the spin motion. So, we set $\nu_2 = 0$, hence $\beta = 0$. Then the effective attenuation (69) becomes $\tilde{\Gamma} = \Gamma_3$. Recall that the width Γ_3 is due to the dynamic broadening caused by local spin fluctuations, which are a kind of local spin waves. These spin fluctuations are, thus, the sole possible origin that could trigger the spin motion, when there are no external fields.

To understand how the spin motion starts, consider short times, when $\gamma t \ll 1$ and the resonator coupling function (62) is yet small, $\alpha \approx 0$. Then Eqs. (70), under $\tilde{\Gamma} = \Gamma_3$, are

$$\frac{dw}{dt} = -2\Gamma_2 w + 2\Gamma_3 s^2, \qquad \frac{ds}{dt} = -(\Gamma_1 + \Gamma_3)s + \Gamma_1\zeta. \tag{94}$$

Their solution at short times reads

$$\begin{aligned} w &\simeq \left(w_0 - \frac{\Gamma_3}{\Gamma_2} s_0^2 \right) \exp\left(-2\Gamma_2 t\right) + \frac{\Gamma_3}{\Gamma_2} s_0^2, \\ s &\simeq \left(s_0 - \frac{\Gamma_1\zeta}{\Gamma_1 + \Gamma_3} \right) \exp\left\{-(\Gamma_1 + \Gamma_3)t\right\} + \frac{\Gamma_1\zeta}{\Gamma_1 + \Gamma_3}. \end{aligned} \tag{95}$$

This shows that, even if no transverse coherence is imposed on the spin system at the initial time, so that $w_0 = 0$, the coherence, anyway, begins to arise triggered by the local spin fluctuations in the presence of a nonzero initial polarization.

The resonator coupling function (62) increases with time. The incoherent quantum stage of spin motion lasts till that time when the coupling function $\alpha(t)$ reaches a value such that $\alpha s = \Gamma_2$. Then, as is seen from Eqs. (70), fast generation of transverse coherence starts in the system. The crossover time t_c, separating the incoherent quantum stage and the coherent regime of motion, is defined by the equality

$$\alpha(t_c)s(t_c) = \Gamma_2. \tag{96}$$

Assuming that the crossover time t_c is the smallest among other characteristic relaxation times, such as τ, T_1, and T_2, we can simplify the solution (95) to the form

$$w(t_c) \simeq w_0 + 2\Gamma_3 t_c s_0^2, \qquad s(t_c) \simeq s_0 + 2\Gamma_1 t_c \zeta, \tag{97}$$

taken at $t = t_c$. This assumption about the time t_c being the shortest is necessary for the existence of superradiance. From definition (96), we find the *crossover time*

$$t_c = \tau \ln\left(\frac{gs_0}{gs_0 - 1}\right), \tag{98}$$

where τ is the resonator ringing time. In order that the quantum stage could certainly change for the coherent regime, the crossover time (98) must be positive and finite, which requires that

$$gs_0 > 1 \qquad (t_c > 0), \tag{99}$$

which is, actually, the condition of maser generation. In the case of sufficiently strong coupling, Eq. (98) reduces to

$$t_c \simeq \frac{\tau}{gs_0} \qquad (gs_0 \gg 1), \tag{100}$$

which shows that the crossover time can really be made shorter than other characteristic times. Since the spin-resonator coupling (60) is positive, condition (99) also tells us that the initial spin polarization s_0 is to be positive, which implies that the spin system is to be prepared in a nonequilibrium state, being in an external field satisfying condition (46).

6. REGIMES OF COHERENT RADIATION

After the crossover time (98), the coupling function (62) quickly increases and reaches the value $\alpha \approx g\Gamma_2$. At this stage, the motion of spins becomes coherent. The main regimes of coherent spin radiation are described below.

6.1. Transient Spin Superradiance. The transient stage corresponds to times larger than the crossover time t_c but essentially shorter than the longitudinal relaxation time T_1. Then in Eqs. (70) one may neglect the longitudinal relaxation parameter Γ_1. Consider the case, when there are no external transverse fields, so that the effective attenuation (69) is $\tilde{\Gamma} = \Gamma_3$. Here we take into account that the resonator noise plays no role in spin motion. Being interested in the situation of sufficiently strong spin-resonator coupling $g \gg 1$, we see that $g\Gamma_2 \gg \Gamma_3$, since $\Gamma_2 \sim \Gamma_3$. Thence, we may omit in Eqs. (70) the terms containing Γ_3. Thus, at this transient stage, we consider the equations

$$\frac{dw}{dt} = -2\Gamma_2(1 - gs)w, \qquad \frac{ds}{dt} = -g\Gamma_2 w. \tag{101}$$

These equations possess an exact solution [10, 55–57] that reads

$$w = \left(\frac{\Gamma_p}{g\Gamma_2}\right)^2 \operatorname{sech}^2\left(\frac{t - t_0}{\tau_p}\right), \qquad s = -\frac{\Gamma_p}{g\Gamma_2}\tanh\left(\frac{t - t_0}{\tau_p}\right) + \frac{1}{g}. \tag{102}$$

The integration parameters in this solution are obtained by considering as the initial condition the values (97) at $t = t_c$, which gives the *delay time*

$$t_0 = t_c + \frac{\tau_p}{2}\ \ln\left|\frac{\Gamma_p + \Gamma_g}{\Gamma_p - \Gamma_g}\right|, \tag{103}$$

where

$$\Gamma_p^2 = \Gamma_g^2 + (g\Gamma_2)^2\left(w_0 + 2\Gamma_3 t_c s_0^2\right), \qquad \Gamma_g \equiv \Gamma_2(gs_0 - 1), \qquad \Gamma_p\tau_p = 1. \tag{104}$$

In order that the delay time be larger than the crossover time, but finite, that is, $t_c < t_0 < \infty$, it should be that $\Gamma_p > \Gamma_g > 0$, which yields

$$w_0 + 2\Gamma_3 t_c s_0^2 > 0, \qquad gs_0 > 1. \tag{105}$$

At the delay time (103), the coherence intensity is maximal,

$$w(t_0) = \left(s_0 - \frac{1}{g}\right)^2 (1 + 2\Gamma_3 t_c), \qquad s(t_0) = \frac{1}{g}, \tag{106}$$

after which the transition coherence fastly decays,

$$w \simeq 4w(t_0)\exp\left(-2\Gamma_p t\right), \qquad s \simeq -s_0 + \frac{2}{g} \qquad (t \gg t_0). \tag{107}$$

The effective time of the coherent pulse, keeping in mind that $\Gamma_3 t_c \ll 1$, writes

$$\tau_p = \frac{T_2}{\sqrt{(gs_0 - 1)^2 + g^2 w_0}}\left[1 - \frac{\Gamma_3 t_c (gs_0)^2}{(gs_0 - 1)^2 + g^2 w_0}\right]. \tag{108}$$

The necessary condition for superradiance is $\tau_p < T_2$, from which one has the inequality

$$(gs_0 - 1)^2 + g^2 w_0 > 1. \tag{109}$$

However, condition (109) is not sufficient for superradiance and defines three possible regimes:

$$\begin{aligned} w_0 = 0, \qquad gs_0 > 2 \qquad &\text{(pure superradiance)}, \\ g^2 w_0 > 1 - (gs_0 - 1)^2, \qquad gs_0 > 1 \qquad &\text{(triggered superradiance)}, \\ g^2 w_0 > 1, \qquad gs_0 \leqslant 1 \qquad &\text{(collective induction)}. \end{aligned} \tag{110}$$

Recall that superradiance is a self-organized process, which requires that the crossover time t_c be positive, hence, according to Eq. (99), it should be that $gs_0 > 1$. Collective induction is not a self-organized process, but it is induced by an initially imposed coherence intensity w_0.

Thus, there exist two types of transient spin superradiance, pure superradiance that is developing as a completely self-organized process and triggered superradiance when the process is mainly self-organized but at the beginning it is slightly pushed by an externally imposed coherence. As follows from Eqs. (107), it is only in the regime when $gs_0 > 2$ that the spin polarization becomes inverted after the superradiant pulse. This may happen in either the regime of pure or triggered superradiance. When $g \gg 2$, the polarization s after t_0 is almost completely inverted to the value $-s_0$. This effect of polarization reversal can be used for fast repolarization of solid-state targets employed in scattering experiments [29, 103].

6.2. Pulsing Spin Superradiance. If there are no external fields acting on the spin system, then, after the transient superradiant burst, occurring at the delay time t_0, the transition coherence dies out. In order to produce a series of superradiant pulses, it is necessary to involve an external action. There are two ways of organizing a regime with a series of superradiant bursts, which can be called pulsing superradiance and punctuated superradiance.

To realize the regime of pulsing spin superradiance, the inversion of spin polarization has to be supported by a permanent pumping, which can be accomplished by means of dynamic nuclear polarization. In that case, the longitudinal relaxation parameter Γ_1 plays the role of the pump rate Γ_1^* that can be much larger than the spin-lattice attenuation of 10^{-5} s^{-1}. The pump rate Γ_1^* can be 10 s^{-1} or larger. In what follows, we shall continue writing Γ_1 instead of Γ_1^*, keeping in mind that the value of Γ_1 corresponds to the pump rate. This is done in order to avoid cumbersome notations. In the presence of pumping, the stationary polarization ζ is the pump polarization, which can reach the value of $\zeta = 1$.

We consider the case when there are no external transverse fields, so that the effective attenuation (69) reduces to $\tilde{\Gamma} = \Gamma_3$. At larger time, when $\gamma t \gg 1$, the coupling function (62) acquires its maximal value $\alpha \cong g\Gamma_2$. Then Eqs. (70) write

$$\frac{dw}{dt} = -2\Gamma_2(1 - gs)w + 2\Gamma_3 s^2, \qquad \frac{ds}{dt} = -g\Gamma_2 w - \Gamma_3 s - \Gamma_1(s - \zeta). \quad (111)$$

To find the conditions when the pulsing regime could arise, we need to consider the long-time behaviour of the solutions to Eqs. (111). For this purpose, we define the stationary solutions to these equations and accomplish the Lyapunov stability analysis [60]. The appearance of complex characteristic exponents, related to particular fixed points, would mean the existence of oscillations around the corresponding stationary solutions.

If the pumping is not very strong, so that $g\zeta \ll -1$, then the stable fixed points are

$$w_1^* \simeq \frac{\zeta^2\Gamma_3}{|g\zeta|\Gamma_2}, \qquad s_1^* \simeq \zeta\left(1 - \frac{\Gamma_3}{|g\zeta|\Gamma_1}\right). \quad (112)$$

For the associated characteristic exponents, we find

$$J_1^+ \simeq -\Gamma_1\left(1 - \frac{\Gamma_3}{2|g\zeta|\Gamma_2}\right), \qquad J_1^- \simeq -2\Gamma_2(1 + |g\zeta|) - \frac{2\Gamma_2\Gamma_3}{\Gamma_1}\left(1 - \frac{\Gamma_1}{2\Gamma_2}\right).$$

These exponents, being real and negative, show that the fixed point (112) is a stable node.

When the spin-resonator coupling is weak or the pumping is not strong, so that $|g\zeta| \ll 1$, then the stable stationary solution is

$$\begin{aligned} w_1^* &\simeq \left(\frac{\zeta\Gamma_1}{\Gamma_1 + \Gamma_3}\right)^2 \frac{\Gamma_3}{\Gamma_2}\left[1 + \frac{\Gamma_1(\Gamma_1 - \Gamma_3)}{(\Gamma_1 + \Gamma_3)^2} g\zeta\right], \\ s_1^* &\simeq \frac{\zeta\Gamma_1}{\Gamma_1 + \Gamma_3}\left(1 - \frac{\Gamma_1\Gamma_3}{\Gamma_1 + \Gamma_3}\, g\zeta\right). \end{aligned} \quad (113)$$

The characteristic exponents, if $\Gamma_1 + \Gamma_3 \neq 2\Gamma_2$, are

$$J_1^+ \simeq -2\Gamma_2 + \frac{2\Gamma_1\Gamma_2(\Gamma_1 - 2\Gamma_2 - \Gamma_3)}{(\Gamma_1 + \Gamma_3)(\Gamma_1 - 2\Gamma_2 + \Gamma_3)} g\zeta,$$

$$J_1^- \simeq -\Gamma_1 - \Gamma_3 + \frac{4\Gamma_1\Gamma_2\Gamma_3}{(\Gamma_1 + \Gamma_3)(\Gamma_1 - 2\Gamma_2 + \Gamma_3)} g\zeta,$$

which classifies the stationary solution (113) as a stable node. But in the special case, when

$$\Gamma_1 + \Gamma_3 = 2\Gamma_2, \quad (114)$$

the characteristic exponents become

$$J_1^{\pm} \simeq -2\Gamma_2 \pm i\,(2\Gamma_1\Gamma_3 g\zeta)^{1/2} + \frac{1}{2}\Gamma_1 g\zeta.$$

Then solutions (113) describe a stable focus, if $g\zeta > 0$. This means that there exists a series of pulses approximately separated from each other by the *separation time*

$$T_{\rm sep} = \pi\sqrt{\frac{2T_1T_3}{g\zeta}} \qquad (0 < g\zeta \ll 1), \tag{115}$$

where $T_3 \equiv \Gamma_3^{-1}$ and condition (114) is valid.

For strong spin-resonator coupling and sufficient pumping, such that $g\zeta \gg 1$, the stationary solution to Eqs. (111) is

$$w_2^* \simeq \frac{\zeta\Gamma_1}{g\Gamma_2}, \qquad s_2^* \simeq \frac{1}{g}. \tag{116}$$

The characteristic exponents

$$J_2^{\pm} \simeq -\frac{1}{2}(\Gamma_1 + \Gamma_3) - \frac{\Gamma_2\Gamma_3}{g\zeta\Gamma_1}$$

show that solution (116) is a stable node.

Now let us analyse the behaviour of stationary solutions to Eqs. (111) for arbitrary values of $g\zeta$, but for different relations between the pumping rate Γ_1 and the dynamic broadening Γ_3. Thus, when $\Gamma_1 \ll \Gamma_3$, then

$$w_1^* \simeq \frac{\zeta^2\Gamma_1^2}{\Gamma_2\Gamma_3}, \qquad s_1^* \simeq \zeta\,\frac{\Gamma_1}{\Gamma_3}\left[1 - (1 + g\zeta)\frac{\Gamma_1}{\Gamma_3}\right]. \tag{117}$$

The corresponding characteristic exponents, if $\Gamma_3 \neq 2\Gamma_2$, are

$$J_1^+ \simeq -2\Gamma_2 - \frac{2(2\Gamma_2 + \Gamma_3)\Gamma_1\Gamma_2}{(2\Gamma_2 - \Gamma_3)\Gamma_3} g\zeta, \qquad J_1^- \simeq -\Gamma_3 - \frac{(4g\zeta + 2\Gamma_2 - \Gamma_3)\Gamma_1}{2\Gamma_2 - \Gamma_3},$$

which defines the fixed point (117) as a stable node. And for the special case, when

$$\Gamma_3 = 2\Gamma_2, \tag{118}$$

one gets

$$J_1^{\pm} \simeq -\Gamma_3 \pm i\sqrt{2g\zeta\Gamma_1\Gamma_3} + \frac{1}{2}(g\zeta - 1)\Gamma_1.$$

In such a case, the fixed point (117) becomes a stable focus, provided that $g\zeta > 0$. This implies the existence of a series of pulses, with the separation intervals

$$T_{\rm sep} = \pi\,\sqrt{\frac{T_1T_2}{g\zeta}} \qquad (\Gamma_1 \ll \Gamma_3 = 2\Gamma_2). \tag{119}$$

Finally, for a large pump rate, such that $\Gamma_1 \gg \Gamma_3$, we have the stationary solutions, which acquire different forms for different values of $g\zeta$. In the case of weak coupling, when $g\zeta < 1$, we get

$$w_1^* \simeq \frac{\zeta^2\Gamma_3}{(1-g\zeta)\Gamma_2}, \qquad s_1^* \simeq \zeta\left[1 - \frac{\Gamma_3}{(1-g\zeta)\Gamma_1}\right], \tag{120}$$

with the characteristic exponents

$$J_1^+ \simeq -\Gamma_1, \qquad J_1^- \simeq -2(1-g\zeta)\Gamma_2$$

telling that the fixed point (120) is a stable node. For the intermediate case, when $g\zeta = 1$, we find

$$w_1^* \simeq \frac{\zeta^2\sqrt{\Gamma_1\Gamma_3}}{\Gamma_2}\left(1 - \frac{3}{2}\sqrt{\frac{\Gamma_3}{\Gamma_1}}\right), \qquad s_1^* \simeq \zeta\left(1 - \sqrt{\frac{\Gamma_3}{\Gamma_1}} + \frac{\Gamma_3}{2\Gamma_1}\right). \tag{121}$$

This point is again a stable node since

$$J_1^+ \simeq -\Gamma_1 + 2\Gamma_2\sqrt{\Gamma_3/\Gamma_1}, \qquad J_1^- \simeq -4\Gamma_2\sqrt{\Gamma_3/\Gamma_1}.$$

And for strong coupling and effective pumping, when $g\zeta > 1$, we obtain

$$w_2^* \simeq \frac{(g\zeta-1)\Gamma_1}{g^2\Gamma_2}\left[1 - \frac{(g\zeta-2)\Gamma_3}{(g\zeta-1)^2\Gamma_1}\right], \qquad s_2^* \simeq \frac{1}{g}\left[1 - \frac{\Gamma_3}{(g\zeta-1)\Gamma_1}\right]. \tag{122}$$

The related characteristic exponents

$$J_2^{\pm} \simeq -\Gamma_1/2\left[1 \pm \sqrt{1 - 8\Gamma_2/\Gamma_1(g\zeta-1)}\right]$$

are real in the interval $1 < g\zeta < 1 + \Gamma_1/8\Gamma_2$, but become complex if

$$g\zeta > 1 + \Gamma_1/(8\Gamma_2). \tag{123}$$

Under this condition, the frequency

$$\omega_\infty \equiv \sqrt{2\Gamma_1\Gamma_2(g\zeta - 1 - \Gamma_1/(8\Gamma_2))} \tag{124}$$

is real. Then we may write

$$J_2^{\pm} \simeq -\Gamma_1/2 \pm i\omega_\infty.$$

This means that the fixed point (122) is a stable focus. Here again there arises a series of pulses separated by the time

$$T_{\rm sep} \equiv (2\pi)/\omega_\infty \cong \pi\sqrt{(2T_1T_2)/(g\zeta-1)}, \tag{125}$$

where, to simplify the expression, we take into account that $\Gamma_1 < \Gamma_2$.

In this way, the regime of pulsing superradiance can appear under the conditions resulting in the formation of a series of pulses, which at long times, are separated from each other by the corresponding separation time (115), or (119), or (125), depending on the accepted conditions. This conclusion has been confirmed by direct numerical solution of Eqs. (111), displaying the regime of pulsing superradiance [9, 63, 65]. Experimental observation of this regime was done in Refs. 30–33. Pulsing spin superradiance can be used for organizing superradiant operation of spin masers [60, 65].

There exists another possibility of producing a series of superradiant pulses, which was named *punctuated spin superradiance* [66]. This can be done in the following way. Suppose that the transient spin superradiance has been realized, resulting in a superradiant burst at the delay time t_0. Then, according to Eqs. (107), at some time after $t_0 + \tau_p$ the spin polarization is reversed. For large coupling g, this reversal is practically complete. Now assume that we again inverse the spin polarization from $-s_0$ to s_0, thus obtaining an inverted nonequilibrium system. Such an inversion can be realized in three ways: reversing the external magnetic field B_0, acting on spins by a resonant π-pulse, or turning the sample 180° about the x axis. After the time t_0, counted from the moment when the newly nonequilibrium state is prepared, another superradiant burst will arise. When the second burst dies out, one can again invert the spin polarization by one of the described methods. Then one more superradiant pulse will occur. This procedure can be repeated as many times as required for creating a desired number of sharp superradiant pulses. Contrary to the regime of pulsing superradiance, analyzed above, when the intervals between superradiant bursts are defined by the system parameters, the time intervals between superradiant pulses in punctuated superradiance can be regulated. It is admissible to form various groups of pulses, with different intervals between separated groups. Hence, it is feasible to compose a code, like the Morse alphabet, which could be employed for processing information [66].

6.3. Induced Coherent Emission. In the investigation of radiation regimes above, we have assumed that there are no external transverse fields that would permanently act on spins. The possible existence of such external fields has been reduced only to the preparation of initial conditions for w_0 and s_0, or to supporting the value of the pumping parameter ζ. But the external transverse field (52) has been switched off. This was done for studying the process of self-organization, which would not be perturbed by external fields pushing spins in the transverse direction and, thus, helping to develop the transition coherence. The temporal arising of self-organized coherent motion of spins, from an initially incoherent state, is the most interesting and the least studied problem. Actually, the gradual appearance of the transition spin coherence has never been described before Refs. 8, 60, 66. This is why we, first, paid the main attention to this problem.

Switching on the external transverse field (52) of course influences spin dynamics. The presence of this field makes the classification of possible regimes of coherent emission less evident. If the transverse field (52) is very weak, so that the leading part in the effective attenuation (69) is due to the dynamic dipolar broadening Γ_3, then the analyzed above regimes do not change much. But if the transverse field is sufficiently strong, so that in the effective attenuation (69) the term Γ_3 is not the largest, but other terms are either also essential or even prevail over Γ_3, then spin dynamics may be noticeably disturbed. However, in general, one may distinguish two principally different regimes related to either weak or strong spin-resonator coupling. If this coupling is weak, such that $gs_0 \leqslant 1$, then even very strong transverse fields can induce coherent radiation with only $\tau_p \leqslant T_2$. But when both the spin-resonator coupling is strong, so that $gs_0 > 1$, and the external transverse field is strong, then the pulse of induced coherent emission can become short, with $\tau_p < T_2$. In any case, when the spin system is subject to the action of the transverse coherent field (52), but there is no pumping supporting the value of the pumping parameter $\zeta > -1$, then there can arise the sole main coherent burst, which may be accompanied by a series of small fastly attenuating oscillations. That is, the transverse field (52) induces only transient coherence radiation. This type of coherent radiation, essentially caused and influenced by an external transverse field, can be called *induced coherent emission*. This should not be confused with superradiance that is a self-organized process occurring without the action of any transverse field of type (52).

As an example, let us consider how a strong constant transverse field would influence spin radiation. When in the transverse field (52) only the constant part h_0 is present, but $h_1 = 0$, then the effective attenuation (69) takes the form

$$\tilde{\Gamma} = \Gamma_3 + \frac{\nu_0^2 \Gamma}{\omega_0^2 + \Gamma^2},$$

with ν_0 defined in Eq. (54) and Γ in Eqs. (68) and (62). From here, we see, first, that the applied constant field has to be rather strong in order to lead to a noticeable, as compared to Γ_3, term. Generally, the second term here is smaller than Γ_3, if condition (55) holds true. If so, then the following process is not much different from what has been analyzed above. Just in all formulas, we need to shift Γ_3 replacing it by

$$\Gamma_3 + \frac{\nu_0^2(\Gamma_2 + \Gamma_3)}{\omega_0^2 + (\Gamma_2 + \Gamma_3)^2} \cong \Gamma_3 + \frac{\nu_0^2}{\omega_0^2}(\Gamma_2 + \Gamma_3).$$

Such a shift would mainly influence the value of the delay time (103). In particular, the natural magnetic anisotropy of spin samples is often modelled by an external constant field. Hence, the existence of such an anisotropy would also play its role in triggering the initial spin motion [61, 62]. However, the term Γ_3 plays the major role.

In the case, when the transverse field (52) consists of only a resonant alternating field, with the amplitude h_1, then the effective attenuation (69) becomes

$$\tilde{\Gamma} = \Gamma_3 + \frac{\nu_1^2 \Gamma}{\Delta^2 + \Gamma^2} \left(1 - \mathrm{e}^{-\Gamma t} \right) ,$$

where ν_1 is given in Eq. (54), $\Gamma = \Gamma_2 + \Gamma_3 - \alpha s$, and the coupling function α being defined in Eq. (62). We have thoroughly analysed the behaviour of solutions to Eqs. (70) for various system parameters, when the resonant transverse field is present. This has been accomplished by numerically solving Eqs. (70) for different initial conditions w_0 and s_0. Note that the term in $\tilde{\Gamma}$, due to the resonant field, is of the order of ν_1^2/Γ_2, which can be comparable with or even larger than Γ_3. Thence the resonant field influences spin motion essentially stronger than a constant transverse field, which is, actually, rather understandable. Numerically solving Eqs. (70), we found that, varying the system parameters and the initial conditions, it is possible to realize a variety of transient coherent pulses. Increasing the amplitude of the resonator field makes the duration of the coherent pulse longer. In all the cases the solutions $w(t)$ and $s(t)$, independently of their initial conditions, die out to almost zero on the scale not longer than T_2. The attenuation of the solutions slows down when increasing ν_1. The latter, under conditions (55), is limited by the relation $\nu_1/\Gamma_2 \ll 10^3$. If the external resonant field is so strong that conditions (55) are no longer valid, then, instead of Eqs. (70), one should go back to Eqs. (47). In the latter case of a very strong resonant field, the solutions to Eqs. (47) may display chaotic behaviour [33, 104, 105].

In conclusion to this section, discussing different regimes of coherent spin radiation, it is useful to recall how the latter could be measured. An ensemble of coherently moving nuclear spins generates magnetodipole radiation with the total intensity

$$I(t) = \frac{2}{3c^3} \left| \ddot{\mathbf{M}}(t) \right|^2 , \tag{126}$$

in which

$$\mathbf{M} = \hbar \gamma_n \sum_{i=1}^{N} \langle \mathbf{I}_i(t) \rangle$$

is the total magnetization of N nuclei. The standardly considered quantity is the radiation intensity averaged over fast oscillations. For the intensity (126), this gives

$$\overline{I}(t) = \frac{2}{3c^3} \mu_n^2 \omega_0^4 N^2 w(t), \tag{127}$$

where $\mu_n \equiv \hbar \gamma_n I$ and $w(t)$ is the function of coherence intensity (44). The proportionality of the radiation intensity (127) to the number of spins squared is characteristic of any coherent radiation, which is not necessarily superradiance.

The level of radiation intensity (127) is rather weak. Thus, the maximal intensity, when $w(t) \approx 1$, for proton spins, with $\mu_n \sim 10^{-23}$ erg·G^{-1}, $\omega_0 \sim 10^8$ Hz, and $N \sim 10^{23}$, is only $\overline{I} \sim 10^{-5}$ W.

However, despite such weak radiation intensity, it can be easily measured. This is because one can measure the power of the current

$$P(t) = Rj^2(t), \tag{128}$$

which is generated in a coil by the radiating spins. Employing the relation (14) between the electric current and the induced magnetic field, we get

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

The resonator field $H(t)$ can be found from Eq. (57). Neglecting the thermal Nyquist noise implies $\beta = 0$. Averaging over fast oscillations yields

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

Using this, for the averaged current power (128), we have

$$\overline{P}(t) = g\Gamma_2 I \hbar\omega N \left(1 - \mathrm{e}^{-\gamma t}\right)^2 w(t). \tag{129}$$

Comparing Eqs. (129) and (127) for $\gamma t \gg 1$, we find

$$\frac{\overline{P}(t)}{\overline{I}(t)} = \frac{3Q\lambda^3}{8\pi^2 V_c}, \tag{130}$$

where $\lambda = 2\pi c/\omega$. The ratio (130) can be quite large. For instance, under $\omega \sim 10^8$ Hz, hence $\lambda \sim 10^3$ cm, $Q \sim 100$, and $V_c \sim 10$ cm^3, one gets the order of 10^8. This explains why even a very low radiation intensity can be easily measured, as is mentioned in Sec. 1.

7. ELECTRON-NUCLEAR HYPERFINE COUPLING

Since real condensed matter contains both nuclei as well as electrons, it is important to understand how the latter could influence the nuclear spin dynamics. Electron and nuclear spins interact with each other through hyperfine forces. A combined system of nuclear and electronic spins is described [53] by the Hamiltonian

$$\hat{H} = \hat{H}_{\rm nucl} + \hat{H}_{\rm el} + \hat{H}_{\rm hyp}, \tag{131}$$

consisting of the parts related to nuclei, $\hat{H}_{\rm nucl}$, electrons, $\hat{H}_{\rm el}$, and their hyperfine interactions, $\hat{H}_{\rm hyp}$.

The nuclear Hamiltonian

$$\hat{H}_{\rm nucl} = \sum_i \hat{H}_i + \frac{1}{2}\sum_{i\neq j}\hat{H}_{ij} \tag{132}$$

is the same as in Eq. (1), with the Zeeman term (2) and the dipolar term (4). The electronic spin Hamiltonian is

$$\hat{H}_{\rm el} = -\frac{1}{2}\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + \hbar\gamma_e\sum_i \mathbf{B}\cdot\mathbf{S}_i, \tag{133}$$

in which J_{ij} is an exchange interaction potential; $\mathbf{S}_i$ is an electron spin, and γ_e is the electron gyromagnetic ratio defining the electron magnetic moment $\mu_e = \hbar\gamma_e S = 0.928\cdot 10^{-20}$ erg $\cdot$ G^{-1}, which approximately equals the Bohr magneton $\mu_B = 0.927\cdot 10^{-20}$ erg $\cdot$ G^{-1}. The total magnetic field acting on electron spins is the same field (3) as that acting on nuclear spins. The dipolar interactions between electrons are much weaker than their exchange interactions.

The Hamiltonian of hyperfine interactions is

$$\hat{H}_{\rm hyp} = A\sum_i \mathbf{I}_i\cdot\mathbf{S}_i + \frac{1}{2}\sum_{i\neq j}\sum_{\alpha\beta} A_{ij}^{\alpha\beta} I_i^\alpha S_j^\beta. \tag{134}$$

The first term here is the Fermi contact hyperfine interaction between nuclei and s electrons, with the energy

$$A = \frac{8\pi}{3}\hbar^2\gamma_n\gamma_e|\psi(0)|^2, \tag{135}$$

where $\psi(\mathbf{r})$ is the electron wave function. Using the estimate $|\psi(0)|^2 \sim 3/4\pi r_B^3$, where $r_B = 5.292\cdot 10^{-9}$ cm is the Bohr radius, we have

$$A \sim \frac{2\hbar^2\gamma_n\gamma_e}{r_B^3}.$$

Since $\mu_0 = \hbar\gamma_n$, for protons, with $\gamma_n = 2.675\cdot 10^4$ G$^{-1}\cdot$s^{-1} we get $A/\hbar \sim 10^9$ s^{-1}. The second term in the hyperfine Hamiltonian (134) describes the dipolar interactions, with the dipolar tensor

$$A_{ij}^{\alpha\beta} = -\hbar^2\,\frac{\gamma_n\gamma_e}{r_{ij}^3}\left(\delta_{\alpha\beta} - 3n_{ij}^\alpha n_{ij}^\beta\right). \tag{136}$$

Similarly to Eqs. (6), one has

$$\sum_{j(\neq i)} A_{ij}^{\alpha\beta} = 0, \qquad \sum_\alpha A_{ij}^{\alpha\alpha} = 0. \tag{137}$$

It is again convenient to pass to the ladder spin operators $S_j^{\pm} \equiv S_j^x \pm iS_j^y$. Then the electron spin Hamiltonian (133) can be written as

$$\hat{H}_{\rm el} = \sum_i \hat{H}_i^{\rm el} + \frac{1}{2}\sum_{i\neq j} \hat{H}_{ij}^{\rm el}, \tag{138}$$

where in the Zeeman term, one has

$$\hat{H}_i^{\rm el} = \hbar\gamma_e B_0 S_i^z + \frac{1}{2}\hbar\gamma_e (B_1 + H)\left(S_i^+ + S_i^-\right), \tag{139}$$

and the interaction term contains

$$\hat{H}_{ij}^{\rm el} = -J_{ij}\left(S_i^+ S_i^- + S_i^z S_j^z\right). \tag{140}$$

The hyperfine Hamiltonian (134) can be presented in the form

$$\hat{H}_{\rm hyp} = \sum_i \hat{H}_i^{\rm hyp} + \frac{1}{2}\sum_{i\neq j} \hat{H}_{ij}^{\rm hyp}. \tag{141}$$

Here the first term is due to contact interactions, with

$$\hat{H}_i^{\rm hyp} = \frac{1}{2}A\left(I_i^+ S_i^- + I_i^- S_i^+ + 2I_i^z S_i^z\right). \tag{142}$$

And the second term corresponds to dipolar hyperfine interactions, with

$$\hat{H}_{ij}^{\rm hyp} = \alpha_{ij}\left(I_i^z S_j^z - \frac{1}{4}I_i^+ S_j^- - \frac{1}{4}I_i^- S_j^+\right) + \beta_{ij} I_i^+ S_j^+ + \beta_{ij}^* I_i^- S_j^- + \\ + \sigma_{ij}\left(I_i^+ S_j^z + I_i^z S_j^+\right) + \sigma_{ij}^*\left(I_i^z S_j^- + I_i^- S_j^z\right), \tag{143}$$

where we use the notation

$$\alpha_{ij} \equiv A_{ij}^{zz}, \quad \sigma_{ij} \equiv \frac{1}{2}\left(A_{ij}^{xz} - iA_{ij}^{yz}\right), \quad \beta_{ij} \equiv \frac{1}{4}\left(A_{ij}^{xx} - A_{ij}^{yy} - 2iA_{ij}^{xy}\right). \tag{144}$$

From the first of Eqs. (137), we have

$$\sum_{j(\neq i)} \alpha_{ij} = \sum_{j(\neq i)} \beta_{ij} = \sum_{j(\neq i)} \sigma_{ij} = 0.$$

The resonator feedback field is, as early, defined by the Kirchhoff equation (18), but with

$$m_x = \frac{\hbar}{V}\sum_i \left(\gamma_n \langle I_i^x\rangle - \gamma_e \langle S_i^x\rangle\right). \tag{145}$$

With this magnetization density, the integral presentation (20) is again valid.

Writing down the Heisenberg equations of motion, we employ the commutation relations for the nuclear spin operators I_i^α and also for electron spins,

$$[S_i^+, S_j^-] = 2\delta_{ij} S_i^z, \qquad [S_i^z, S_j^\pm] = \pm\delta_{ij} S_i^\pm, \qquad [I_i^\alpha, S_j^\beta] = 0.$$

Generalizing Eqs. (21), we define the local fields acting on nuclear spins

$$\begin{aligned} \xi_0 &\equiv \frac{1}{\hbar} \sum_{j(\neq i)} \left(c_{ij} I_j^+ + c_{ij}^* I_j^- + a_{ij} I_j^z + \frac{1}{2}\sigma_{ij} S_j^+ + \frac{1}{2}\sigma_{ij}^* S_j^- + \frac{1}{2}\alpha_{ij} S_j^z \right), \\ \xi &\equiv \frac{i}{\hbar} \sum_{j(\neq i)} \left(2b_{ij} I_j^+ - \frac{1}{2} a_{ij} I_j^- + 2c_{ij} I_j^z + \beta_{ij} S_j^+ - \frac{1}{4}\alpha_{ij} S_j^- + \sigma_{ij} S_j^z \right), \end{aligned} \tag{146}$$

and local fields acting on electron spins,

$$\begin{aligned} \varphi_0 &\equiv \frac{1}{2\hbar} \sum_{j(\neq i)} \left(\sigma_{ij} I_j^+ + \sigma_{ij}^* I_j^- + \alpha_{ij} I_j^z \right) + \frac{1}{\hbar} \left(J_0 S_i^z - \sum_{j(\neq i)} J_{ij} S_j^z \right), \\ \varphi &\equiv \frac{i}{\hbar} \sum_{j(\neq i)} \left(\beta_{ij} I_j^+ - \frac{1}{4}\alpha_{ij} I_j^- + \sigma_{ij} I_j^z \right) + \frac{1}{\hbar} \left(J_0 S_i^- - \sum_{j(\neq i)} J_{ij} S_j^- \right), \end{aligned} \tag{147}$$

where $J_0 \equiv \sum_{j(\neq i)} J_{ij}$. Introduce the frequency operators

$$\hat{\Omega}_n \equiv -\gamma_n B_0 + \frac{A}{\hbar} S_i^z + \xi_0, \qquad \hat{\Omega}_e \equiv \gamma_e B_0 + \frac{A}{\hbar} I_i^z + \varphi_0, \tag{148}$$

and the force operators

$$\hat{f}_n \equiv -i\gamma_n (B_1 + H) + i\, \frac{A}{\hbar} S_i^- + \xi, \qquad \hat{f}_e \equiv i\gamma_e (B_1 + H) + i\, \frac{A}{\hbar} I_i^- + \varphi_0. \tag{149}$$

Then the Heisenberg equations of motion can be presented in the form

$$\begin{aligned} \frac{dI_i^-}{dt} &= -i\hat{\Omega}_n I_i^- + I_i^z \hat{f}_n, \quad \frac{dI_i^z}{dt} = -\frac{1}{2}\left(\hat{f}_n^+ I_i^- + I_i^+ \hat{f}_n \right), \\ \frac{dS_i^-}{dt} &= -i\hat{\Omega}_e S_i^- + S_i^z \hat{f}_e, \quad \frac{dS_i^z}{dt} = -\frac{1}{2}\left(\hat{f}_e^+ S_i^- + S_i^+ \hat{f}_e \right). \end{aligned} \tag{150}$$

In the stationary regime, Eqs. (150) define coupled nuclear-electron spin waves, whose description can be done similarly to Sec. 3. To accomplish this, we may define, by analogy with (24),

$$S_i^\alpha \equiv \langle S_i^\alpha \rangle + \delta S_i^\alpha. \tag{151}$$

Here, as in Eq. (25), we assume an ideal lattice, for which $\langle S_i^\alpha \rangle = \langle S_j^\alpha \rangle$. Considering the case without transverse magnetic fields, when $B_1 = H = 0$, we set $\langle S_i^+ \rangle = 0$, while $\langle S_i^z \rangle \neq 0$. This is analogous to setting $\langle I_i^\pm \rangle = 0$, while $\langle I_i^z \rangle \neq 0$. Hence, $I_i^\pm = \delta I_i^\pm$ and $S_i^\pm = \delta S_i^\pm$, because of which the local fields (146) and (147) correspond to random local spin fluctuations, with $\xi_0 = \delta\xi_0$, $\xi = \delta\xi$, $\varphi_0 = \delta\varphi_0$, and $\varphi = \delta\varphi$. Then the second and fourth of Eqs. (150) give $\delta I_i^z = 0$ and $\delta S_i^z = 0$. To the remaining equations, we substitute the Fourier transformed nuclear spin operators (28) and

$$S_j^\pm = \sum_k S_k^\pm \, \exp\left(\mp i \mathbf{k} \cdot \mathbf{r}_j\right) . \tag{152}$$

We invoke the Fourier transforms (29) and also introduce

$$\begin{gathered} \alpha_k \equiv \textstyle\sum_{j(\neq i)} \alpha_{ij} \, \exp\left(-i\mathbf{k} \cdot \mathbf{r}_{ij}\right) , \quad \beta_k \equiv \textstyle\sum_{j(\neq i)} \beta_{ij} \, \exp\left(-i\mathbf{k} \cdot \mathbf{r}_{ij}\right) , \\ J_k \equiv \textstyle\sum_{j(\neq i)} J_{ij} \, \exp\left(-i\mathbf{k} \cdot \mathbf{r}_{ij}\right) . \end{gathered} \tag{153}$$

Generalizing Eqs. (31), we use the notation

$$\begin{gathered} \mu_k \equiv -\gamma_n B_0 + \frac{a_k}{2\hbar} \langle I_i^z \rangle + \frac{A}{\hbar} \langle S_i^z \rangle , \qquad \lambda_k \equiv \frac{2b_k}{\hbar} \langle I_i^z \rangle , \\ \varepsilon_k \equiv \gamma_e B_0 + \frac{1}{\hbar} (J_k - J_0) \langle S_j^z \rangle + \frac{A}{\hbar} \langle I_j^z \rangle . \end{gathered} \tag{154}$$

Then Eqs. (150) can be reduced to the equations

$$\begin{aligned} i \frac{dI_k^-}{dt} &= \mu_k I_k^- - \lambda_k I_k^+ - \frac{1}{\hbar} \left(A - \frac{\alpha_k}{4} \right) \langle I_i^z \rangle S_k^- - \frac{\beta_k}{\hbar} \langle I_i^z \rangle S_k^+ , \\ i \frac{dS_k^-}{dt} &= \varepsilon_k S_k^- - \frac{1}{\hbar} \left(A - \frac{\alpha_k}{4} \right) \langle S_i^z \rangle I_k^- - \frac{\beta_k}{\hbar} \langle S_i^z \rangle I_k^+ , \end{aligned} \tag{155}$$

which describe coupled nuclear-electron spin waves. The spectrum of the latter follows from Eqs. (155) yielding the fourth-order algebraic equation defining two positive branches of coupled spin-wave collective excitations. The coupling between the branches comes from the terms of Eqs. (155) containing the hyperfine parameters A, α_k, and β_k. Without the latter, there would exist two separate types of collective excitations corresponding to nuclear spin waves with the spectrum (33) and to electron spin waves with the spectrum ε_k. In the presence of the hyperfine coupling, one may conditionally distinguish one of the branches of the coupled collective excitations as the branch related to nuclear spin waves, while another branch can be ascribed to electron spin waves. The hyperfine coupling, of course, essentially modifies the spectra of the coupled nuclear-electron spin waves. In particular, nuclear spin waves become noticeably influenced by the

indirect exchange through electron spins, when the latter possess a long-range magnetic order. The related exchange force is called the Suhl–Nakamura force [54], which can stabilize the nuclear spin waves. When nuclear spins are initially in a nonequilibrium state, nuclear-electron spin waves play the role of a trigger starting the motion of nuclear spins from the nonequilibrium to their equilibrium state.

8. ENHANCED NUCLEAR RADIATION

To describe the nonequilibrium dynamics of coupled nuclear and electron spins, we again employ the scale separation approach [9, 42, 56, 57, 106]. To this end, the variables (146) and (147), characterizing local field fluctuations, are treated as random fields. The stochastic averages for the random variables (146) are defined as in Eq. (41). However, the dynamic broadening Γ_3, in the presence of hyperfine interactions, is renormalized according to the relation

$$\Gamma_3^2 = \Gamma_{nn}^2 + \Gamma_{ne}^2, \tag{156}$$

where $\Gamma_{nn} \approx \rho_n \gamma_n \mu_n$ is the broadening due to dipolar nuclear interactions, with ρ_n being the density of nuclear spins, and $\Gamma_{ne} \approx \sqrt{\rho_n \rho_e}\gamma_n \mu_e$ is the broadening caused by the hyperfine nuclear-electron interactions, with ρ_e being the density of electron spins.

For the random fields (147), the stochastic averages can be defined as

$$\begin{aligned} \langle\langle \varphi_0(t)\rangle\rangle = \langle\langle \varphi(t)\rangle\rangle = 0, \quad \langle\langle \varphi_0(t)\varphi_0(t')\rangle\rangle = 2\gamma_3 \delta(t-t'), \\ \langle\langle \varphi_0(t)\varphi(t')\rangle\rangle = \langle\langle \varphi(t)\varphi(t')\rangle\rangle = 0, \quad \langle\langle \varphi^*(t)\varphi(t')\rangle\rangle = 2\gamma_3 \delta(t-t'), \end{aligned} \tag{157}$$

with the dynamic broadening γ_3 given by the relation

$$\gamma_3^2 = \Gamma_{ee}^2 + \Gamma_{en}^2. \tag{158}$$

Here, $\Gamma_{ee} \approx \rho_e \gamma_e \mu_e$ is the broadening caused by electron spin fluctuations, while $\Gamma_{en} \approx \sqrt{\rho_e \rho_n}\gamma_e \mu_n$ is the broadening due to electron-nuclear hyperfine interactions. Recall that $\mu_n \equiv \hbar \gamma_n I$ and $\mu_e \equiv \hbar \gamma_e S$ are the nuclear and electron magnetic moments, respectively.

To estimate the related broadening widths, we may assume that $\rho_n \sim \rho_e \sim 10^{23}$ cm^{-3}. Then, with $\gamma_n = 2.675 \cdot 10^4$ G$^{-1} \cdot$ s^{-1}, $\gamma_e = 1.759 \cdot 10^7$ G$^{-1} \cdot$ s^{-1}, $\mu_n = 1.411 \cdot 10^{-23}$ erg $\cdot$ G^{-1}, and $\mu_e = 0.928 \cdot 10^{-20}$ erg $\cdot$ G^{-1}, we find $\Gamma_{nn} \sim 10^4$ s^{-1}, $\Gamma_{ne} \sim 10^7$ s^{-1}, $\Gamma_{ee} \sim 10^{10}$ s^{-1}, and $\Gamma_{en} \sim 10^7$ s^{-1}. As is seen, hyperfine interactions may strongly influence local nuclear spin fluctuations but are not important for electron spin motion.

By analogy with Eqs. (43) to (45), we introduce the functional variables for electron spins: the transition function

$$x \equiv \frac{1}{SN_e} \sum_{j=1}^{N_e} \langle S_j^- \rangle , \tag{159}$$

where N_e is the number of electrons, the coherence intensity

$$y \equiv \frac{1}{S^2 N_e (N_e - 1)} \sum_{i \neq j}^{N_e} \langle S_i^+ S_j^- \rangle , \tag{160}$$

and the electron spin polarization

$$z \equiv \frac{1}{SN_e} \sum_{j=1}^{N_e} \langle S_j^z \rangle . \tag{161}$$

The averaging in Eqs. (159) to (161) is accomplished over the spin degrees of freedom, not involving the random variables (146) and (147). The stochastic averages for the latter are defined in Eqs. (41) and (157). The random variables ascribed to nuclear and electron fluctuations are assumed to be uncorrelated with each other.

Employing expressions (159) to (161), we may present the averages of the frequency operators (148) as

$$\langle \hat{\Omega}_n \rangle = \omega_n + \xi_0 , \qquad \langle \hat{\Omega}_e \rangle = \omega_e + \varphi_0 , \tag{162}$$

where the effective nuclear and electron frequencies are

$$\omega_n \equiv -\gamma_n B_0 + \frac{1}{\hbar} ASz , \qquad \omega_e \equiv \gamma_e B_0 + \frac{1}{\hbar} AIs . \tag{163}$$

For an external field $B_0 \sim 1$ T $= 10^4$ G, since $\gamma_n \sim 10^4$ G$^{-1} \cdot$s^{-1}, $\gamma_e \sim 10^7$ G$^{-1} \cdot$s^{-1}, and $A/\hbar \sim 10^9$ s^{-1}, we see that the hyperfine term in the nuclear frequency ω_n can become larger than the nuclear Zeeman frequency $\omega_0 \equiv |\gamma_n B_0| \sim 10^8$ Hz, provided that there exists a nonzero average electron magnetization, with $z \neq 0$. Hence, it is necessary to accurately take into account the possible hyperfine shift of the nuclear frequency. At the same time, the electron Zeeman frequency $\gamma_e B_0 \sim 10^{11}$ Hz is much larger than the hyperfine frequency shift, so that $\omega_e \approx \gamma_e B_0$.

For the averages

$$f_n \equiv \langle \hat{f}_n \rangle , \qquad f_e \equiv \langle \hat{f}_e \rangle \tag{164}$$

of the force operators (149), we have

$$f_n = -i\gamma_n(B_1 + H) + \frac{i}{\hbar}ASx + \xi, \qquad f_e = i\gamma_e(B_1 + H) + \frac{i}{\hbar}AIu + \varphi. \quad (165)$$

Here, the hyperfine terms play the role of additional fields acting on nuclear and electron spins.

It is worth mentioning that the hyperfine shifts in the effective frequencies (163) are caused by the first-order hyperfine effect. Generally, there exists the second-order effect due to the Suhl–Nakamura forces. The second-order frequency shift appears as follows. Let us consider the evolution equations (150) for the electron spin operators, with the explicit form of the variables (147), without treating the latter as random. Then, the third of Eqs. (150) yields

$$S_i^-(t) \approx \left[S_i^-(0) + i\frac{\hat{F}_{en}}{\omega_e} S_i^z \right] \mathrm{e}^{-i\omega_e t} - i\frac{\hat{F}_{en}}{\omega_e} S_i^z,$$

where

$$\hat{F}_{en} \equiv \frac{i}{\hbar} \sum_{j(\neq i)} \left(\beta_{ij} I_j^+ - \frac{1}{4}\alpha_{ij} I_j^- + \sigma_{ij} I_j^z \right).$$

Substituting this to the first of equations (150), we come to the conclusion that the effective nuclear frequency should have the form

$$\omega_n = \omega_0 + \frac{A}{\hbar}\langle S_i^z \rangle + \Delta_{\mathrm{SN}}, \quad (166)$$

in which the Suhl–Nakamura frequency shift is

$$\Delta_{\mathrm{SN}} = \sum_{j(\neq i)} \frac{|\sigma_{ij}|^2}{\hbar^2 \omega_e} \langle S_i^z \rangle s. \quad (167)$$

This is often called the dynamic frequency shift because of its dependence on $s = s(t)$.

However, the second-order frequency shift (167) is much smaller than the first-order hyperfine shift in Eq. (166),

$$\left| \frac{\hbar \Delta_{\mathrm{SN}}}{A} \right| \ll 1.$$

Thus, Eq. (167) shows that $\Delta_{\mathrm{SN}} \sim n_0 S \rho_n \rho_e (\hbar \gamma_n \gamma_e)^2 / \omega_e$, provided that there is a long-range magnetic order of electron spins, i.e., $\langle S_i^z \rangle \neq 0$. Here $n_0 \sim 10$ is the number of the nearest neighbours. For the typical values of parameters, this gives $\Delta_{\mathrm{SN}} \sim 10^4$ Hz, which is much smaller than $A/\hbar \sim 10^9$ Hz, hence

$\hbar\Delta_{\rm SN}/A \sim 10^{-5}$ is really a very small value. This makes it admissible to take account of only first-order hyperfine frequency shifts.

Averaging Eqs. (150) over the spin degrees of freedom, we shall use the decoupling of nuclear and spin operators

$$\langle I_i^\alpha S_j^\beta \rangle = \langle I_i^\alpha \rangle \langle S_j^\beta \rangle , \tag{168}$$

valid for all i and j. And, similarly to Eq. (42), we employ the decoupling

$$\langle S_i^\alpha S_j^\beta \rangle = \langle S_i^\alpha \rangle \langle S_j^\beta \rangle \qquad (i \neq j) \tag{169}$$

for different indices i and j. Recall that this type of decoupling has been called the stochastic mean-field approximation, since the averages $\langle \ldots \rangle$ do not include the averaging over the stochastic degrees of freedom, thus, allowing for the consideration of quantum effects [8, 9].

Finally, from Eqs. (150), we come to the stochastic differential equations describing the evolution of nuclear spins,

$$\begin{aligned} \frac{du}{dt} = -i(\omega_n + \xi_0 - i\Gamma_2)u + f_n s, \quad \frac{dw}{dt} = -2\Gamma_2 w + (u^* f_n + f_n^* u)\, s, \\ \frac{ds}{dt} = -\frac{1}{2}\,(u^* f_n + f_n^* u) - \Gamma_1 (s - \zeta), \end{aligned} \tag{170}$$

as well as the motion of electron spins,

$$\begin{aligned} \frac{dx}{dt} = -i(\omega_e + \varphi_0 - i\gamma_2)x + f_e z, \qquad \frac{dy}{dt} = -2\gamma_2 y + (x^* f_e + f_e^* x) z, \\ \frac{dz}{dt} = -\frac{1}{2}\,(x^* f_e + f_e^* x) - \gamma_1 (z - \sigma), \end{aligned} \tag{171}$$

where γ_1 and γ_2 are the spin-lattice and spin-spin relaxation parameters for electronic spins, while σ is an equilibrium value of $\langle S_i^z \rangle / S$. Equations (170) and (171) are to be complimented by the Kirchhoff equation (18), with m_x given by Eq. (145), which reads

$$m_x = \frac{1}{2}\rho_n \mu_m (u^* + u) - \frac{1}{2}\rho_e \mu_e (x^* + x). \tag{172}$$

The evolution equations for nuclear and electron spins are very similar to each other. This means that we could consider electron spin superradiance on the absolutely same footing as the nuclear spin superradiance. The major difference is that hyperfine interactions can essentially influence the motion of nuclear spins, while these interactions do not disturb much the electron spin motion. Therefore, we concentrate now our attention on the evolution of nuclear spins.

The system of equations (170) plus (171) can again be solved involving the scale separation approach [8, 9, 42, 56, 57]. For this purpose, we remember the existence of several small parameters defined in Eqs. (49), (51), (55), and also assume the validity of the resonance condition (56). In addition, we have as well the inequalities

$$\frac{\gamma_1}{|\omega_e|} \ll 1, \qquad \frac{\gamma_2}{|\omega_e|} \ll 1, \qquad \frac{\gamma_3}{|\omega_e|} \ll 1. \tag{173}$$

Also, we have to keep in mind that

$$\frac{\mu_n}{\mu_e} = \frac{\gamma_n I}{\gamma_e S} \ll 1. \tag{174}$$

Being based on these inequalities, we may realize the following *classification of relative quasi-invariants*: The functions w and s are temporal quasi-invariants with respect to the functional variables u, x, y, and z; the functions y and z are quasi-invariants with respect to x; and u is also a quasi-invariant with respect to x.

To derive the expression for the resonator feedback field, we again use the integral form (20), iterating it with the approximate solutions for x and u, keeping in mind that, because of the inequalities (174), we have $|\omega_n/\omega_e| \ll 1$. We substitute in integral (20) the solutions

$$x \cong \left(x_0 - \frac{AIz}{\hbar\omega_e} u \right) \mathrm{e}^{-i\omega_e t} + \frac{AIz}{\hbar\omega_e} u$$

and $u \cong u_0 \exp(-i\omega_n t)$. This shows that electron spins oscillate with the frequencies ω_e, ω_n, and $\omega_e + \omega_n$. Calculating the transverse magnetization (172), we insert it into integral (20), replacing z by its average σ. In that way, we come to the expression $\gamma_n H = \mu_0 H/\hbar$ having the same form (57), but with the coupling function

$$\tilde{\alpha} = \tilde{g}\Gamma_2 \left(1 - \mathrm{e}^{-\gamma t} \right),$$

instead of α, where the renormalized spin-resonator coupling is

$$\tilde{g} \equiv g \left(1 - \frac{\rho_e \mu_e AI\sigma}{\rho_n \mu_n \hbar \omega_e} \right).$$

Here g is defined in Eq. (60) and $\omega_e \cong \gamma_e B_0$. Taking into account condition (46), according to which $\gamma_n B_0 < 0$, the renormalized spin-resonator coupling can be presented as

$$\tilde{g} \equiv g \left(1 + \frac{\rho_e AS\sigma}{\rho_n \hbar \omega_0} \right), \tag{175}$$

where $\omega_0 \equiv |\gamma_n B_0|$. Analogously to the previous procedures, we may study different regimes of coherent nuclear radiation, just replacing everywhere the spin

resonator coupling (60) by its renormalized value (175) and by using expression (156) for the dynamic broadening $\Gamma_3 = \sqrt{\Gamma_{nn}^2 + \Gamma_{ne}^2}$.

The noticeable renormalization of the coupling (175) occurs when there exists a long-range magnetic order in electron spins, so that $\sigma \neq 0$. Then the coupling (175) can become an order larger than g, that is, $\tilde{g}/g \sim 10$. This, respectively, leads to the *enhancement of nuclear spin radiation*, with the current power (129) becoming an order stronger. Simultaneously, the crossover time (98), or (100), and the pulse time (108) become an order shorter. Thus, in the case of pure spin superradiance, when $w_0 = 0$, and for $\tilde{g}s_0 \gg 1$, we have

$$t_c \simeq \frac{\tau}{\tilde{g}s_0}, \qquad \tau_p \simeq \frac{T_2}{\tilde{g}s_0}.$$

Then the delay time (103) changes to

$$t_0 \simeq \frac{\tau}{\tilde{g}s_0} + \frac{T_2}{2\tilde{g}s_0} \ln \left| \frac{2}{\tilde{\Gamma}_3 \tau} \right| ,$$

which shows that t_0 also becomes about an order shorter. It is important to stress that the enhancement effect can exist not only in ferromagnets or ferrimagnets, but also in paramagnets, provided that the external field is sufficiently large and the temperature is low. Thus, for a paramagnetic system of electrons, the average magnetization per spin, due to the external magnetic field B_0, is

$$\sigma = \tanh \left(\frac{\mu_e B_0}{k_B T} \right) .$$

For the typical values of parameters, with $B_0 \sim 10^4$ G, we have $\mu_e B_0 \sim 10^{-16}$ erg, hence $\mu_e B_0/\hbar \sim 10^{11}$ s^{-1}. The thermal frequency $\omega_T \equiv k_B T/\hbar$, at $T \sim 0.1$ K, is $\omega_T \sim 10^{11}$ s^{-1}, which is of the same order as $\mu_e B_0/\hbar \sim \omega_e$. Then $\sigma \sim \tanh(1) \approx 0.76$. Therefore, the enhancement effect, under such conditions, exists for a paramagnet as well.

9. SUPERRADIANCE BY MAGNETIC MOLECULES

There exists an interesting class of composite objects behaving as giant nuclei with total spins ranging from $1/2$ to rather large values. These are magnetic molecules [60, 107–109], which can form crystalline materials termed molecular magnets. In these materials, all magnetic clusters possess the same shape, size, and orientation, because of which the inhomogeneous broadening caused by the system nonuniformity is very low. As examples of such magnetic molecules, we may mention the following, where in brackets the total ground-state spin is shown:

$$\mathrm{K}_6 \left[\mathrm{V}_{15}^{4+} \mathrm{As}_6 \mathrm{O}_{42} (\mathrm{H}_2\mathrm{O}) \right] \cdot 8\mathrm{H}_2\mathrm{O} \qquad (S = 1/2),$$

$$[(PhSiO_2)_6Cu_6(O_2SiPh)_6] \qquad (S = 3),$$

$$[Mn_{12}O_{12}(CH_3COO)_{16}(H_2O)_4] \cdot 2CH_3COOH \cdot 4H_2O \qquad (S = 10),$$

$$[Fe_8O_2(OH)_{12}T_6]^{8+} \qquad (S = 10),$$

$$Mn_6O_4Br_4(Et_2dbm)_6 \qquad (S = 12),$$

$$[Cr(CNMnL)_6]\,(ClO_4)_9 \qquad (S = 27/2).$$

The first of the above molecules are briefly denoted as V_{15}. Molecular magnets formed by these molecules have no magnetic anisotropy. The short-hand notation for the third molecule is Mn_{12}. The corresponding molecular magnets possess a rather strong single-site anisotropy characterized by the parameter $D \approx 0.967 \cdot 10^{-16}$ erg, which gives $D/k_B \approx 0.7$ K and $D/\hbar \approx 0.917 \cdot 10^{11}$ s^{-1}. At temperatures below the blocking temperature $T_B \approx 3$ K, the magnetization of a molecular crystal formed of Mn_{12} is preserved during the relaxation time $T_1 \sim 10^7$ s. The size of each molecule Mn_{12} is about 10 Å and the distance between the nearest neighbours is around 14 Å. Hence the average density is $\rho \approx 0.364 \cdot 10^{21}$ cm^{-3}. The spin-spin relaxation parameter is $\Gamma_2 = n_0 \rho \gamma_S |\mu_0| S$, where

$$\mu_0 = -g_S \mu_B = -\hbar \gamma_S, \tag{176}$$

g_S is the Landé factor; μ_B, Bohr magneton, and γ_S is the gyromagnetic ratio of a molecule with spin S. This parameter $\Gamma_2 \sim 10^{10}$ s^{-1} is due to dipolar spin interactions.

The fourth molecule has the abbreviation Fe_8. In its chemical formula, the letter T stands for the organic ligand triazacyclononane. The corresponding molecular magnet possesses the magnetic anisotropy described by the parameter $D \approx 0.414 \cdot 10^{-16}$ erg. Then, $D/k_B \approx 0.3$ K and $D/\hbar \approx 0.392 \cdot 10^{11}$ s^{-1}. Below the blocking temperature $T_B \approx 1$ K, the relaxation of magnetization to zero in a molecular magnet occurs during the relaxation time $T_1 \sim 10^5$ s. The average density is $\rho \approx 0.4 \cdot 10^{21}$ cm^{-3}. The interaction between molecules is also through dipolar forces, resulting in the spin-spin attenuation $\Gamma_2 \sim 10^{10}$ s^{-1}.

In the last molecule from the list above, the letter L in its chemical formula means a neutral pentadentate ligand. Its properties are close to those of the molecules Mn_{12} and Fe_8 described above.

In principle, at very low temperatures, of the order of $\hbar\Gamma_2/k_B \sim 0.1$ K, a purely dipolarly interacting molecular magnet can become magnetically ordered. The paramagnet-ferromagnet transition at $T_c \approx 0.16$ K was observed [110] in the dipolar magnet formed of the molecules of the fifth type from the list above, this kind of a molecule being abbreviated as Mn_6. The molecular magnet composed of the latter molecules has a week anisotropy, with $D \approx 1.795 \cdot 10^{-18}$ erg. Thus, $D/k_B \approx 0.013$ K and $D/\hbar \approx 1.7 \cdot 10^9$ s^{-1}. The spin-spin dipolar relaxation parameter is $\Gamma_2 \sim 10^{10}$ s^{-1}. The spin-lattice relaxation time, at $B_0 \sim 10^4$ G and

$T \sim 0.1$ K, is $T_1 \sim 10^{-3}$ s, so that $\Gamma_1 \sim 10^3$ s^{-1}. Generally, T_1 can be estimated from the formula

$$T_1 \approx \tau_0 \exp\left(\frac{\hbar\gamma_S B_0}{k_B T} \right) .$$

For the molecule Mn_6, one has $\tau_0 \approx 3 \cdot 10^{-4}$ s.

The Hamiltonian of a molecular magnet, consisting of N magnetic molecules, each having spin S, has the form

$$\hat{H} = \sum_i \hat{H}_i + \frac{1}{2} \sum_{i \neq j} \hat{H}_{ij} \; , \tag{177}$$

in which $\hat{H}_i$ is related to individual spins and $\hat{H}_{ij}$, to pair spin interactions. The individual term

$$\hat{H}_i = -\mu_0 \mathbf{B} \cdot \mathbf{S}_i - D(S_i^z)^2 \tag{178}$$

includes the part characterizing the single-site magnetic anisotropy with the anisotropy parameter D. Positive $D > 0$ implies an easy-axis anisotropy, while $D < 0$ means an easy-plane anisotropy. The pair term in the Hamiltonian (177) corresponds to dipolar spin interactions

$$\hat{H}_{ij} = \sum_{\alpha\beta} C_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta \tag{179}$$

with the dipolar tensor

$$C_{ij}^{\alpha\beta} = \frac{\mu_0^2}{r_{ij}^3} \left(\delta_{\alpha\beta} - 3 n_{ij}^\alpha n_{ij}^\beta \right) .$$

All notations in this sections are close to those of Sec. 2. The difference is that now we are considering the molecular spins S_i^α formed by electrons. The ground-state molecular spin S can be quite large. The principal distinction, as compared to Sec. 2, is the necessity of taking into account the magnetic anisotropy that can be rather strong. As early, the dipolar tensor enjoys the properties

$$\sum_\alpha C_{ij}^{\alpha\beta} = 0, \qquad \sum_{j(\neq i)} C_{ij}^{\alpha\beta} = 0.$$

Similar to Eq. (9), we use the notation

$$a_{ij} \equiv C_{ij}^{zz}, \qquad c_{ij} \equiv \frac{1}{2} \left(C_{ij}^{xz} - i C_{ij}^{yz} \right) , \qquad b_{ij} \equiv \frac{1}{4} \left(C_{ij}^{xx} - C_{ij}^{yy} - 2i C_{ij}^{xy} \right) ,$$

with Eq. (10) being valid. Employing the ladder spin operators $S_i^\pm \equiv S_i^x \pm i S_i^y$, we may present the term (178) as

$$\hat{H}_i = -\mu_0 B_0 S_i^z - D(S_i^z)^2 - \frac{1}{2} \mu_0 (B_1 + H)(S_i^+ + S_i^-), \tag{180}$$

where the total magnetic field is assumed to be the same as in Eq. (3). And the pair term (179) takes the form

$$\hat{H}_{ij} = a_{ij}\left(S_i^z S_j^z - \frac{1}{2}S_i^+ S_j^-\right) + \\ + b_{ij}S_i^+ S_j^+ + b_{ij}^* S_i^- S_j^- + 2c_{ij}S_i^+ S_j^z + 2c_{ij}^* S_i^- S_j^z. \quad (181)$$

In the Kirchhoff equation (18), the magnetization density is

$$m_x = \frac{\mu_0}{V}\sum_i \langle S_i^x\rangle. \quad (182)$$

The fluctuating local fields are denoted as

$$\xi_0 \equiv \frac{1}{\hbar}\sum_{j(\neq i)}\left(a_{ij}S_j^z + c_{ij}^* S_j^- + c_{ij}S_j^+\right), \\ \xi \equiv \frac{i}{\hbar}\sum_{j(\neq i)}\left(2c_{ij}S_j^z - \frac{1}{2}a_{ij}S_j^- + 2b_{ij}S_j^+\right). \quad (183)$$

Writing down the Heinsenberg equations for the spin operators, we shall need, in addition to the standard commutation relations, the relation

$$\left[S_i^-, (S_j^z)^2\right] = \delta_{ij}\left(S_i^- S_i^z + S_i^z S_i^-\right).$$

Thus, the evolution equations for the spin operators, with the notation

$$f \equiv i\gamma_S(B_1 + H) + \xi, \quad (184)$$

become

$$\frac{dS_i^-}{dt} = -i(\gamma_S B_0 + \xi_0)S_i^- + fS_i^z + i\frac{D}{\hbar}\left(S_i^- S_i^z + S_i^z S_i^-\right), \\ \frac{dS_i^z}{dt} = -\frac{1}{2}\left(f^* S_i^- + S_i^+ f\right). \quad (185)$$

Analogously to Sec. 3, we can show that there exist *molecular spin waves*. A subtle point here is the linearization of the last term in the first of Eqs. (185), which has to be done so that to take into account the absence of this term for $S = 1/2$. This can be achieved by invoking the linearization

$$S_i^- S_i^z + S_i^z S_i^- = \left(2 - \frac{1}{S}\right)\langle S_i^z\rangle S_i^- \quad (186)$$

possessing the correct asymptotic behaviour for $S = 1/2$ as well as for $S \to \infty$ (see discussion in Refs. 60, 94). Introduce the effective frequency

$$\omega_D \equiv \gamma_S B_0 - \frac{D}{\hbar}\left(2 - \frac{1}{S}\right)\langle S_i^z\rangle. \tag{187}$$

From Eqs. (185), we obtain

$$\frac{dS_k^-}{dt} = -i\mu_k S_k^- + i\lambda_k S_k^+, \tag{188}$$

where

$$\mu_k \equiv \frac{a_k}{2\hbar}\langle S_i^z\rangle + \omega_D, \qquad \lambda_k \equiv \frac{2b_k}{\hbar}\langle S_i^z\rangle. \tag{189}$$

Then the spectrum of molecular spin waves is described by Eq. (33). The condition for the spectrum to be positive, in the case of a cubic symmetry reads

$$\frac{2\rho\mu_0^2 S\omega_0}{\hbar\omega_D^2} < 1 \qquad \left(\omega_0 \equiv \frac{1}{\hbar}|\mu_0 B_0|\right).$$

The external longitudinal magnetic field is directed so that, similarly to the inequality (46), we have

$$\mu_0 B_0 < 0, \qquad \gamma_S B_0 > 0. \tag{190}$$

Following Sec. 4, we define the transition function

$$u \equiv \frac{1}{SN}\sum_{i=1}^{N}\langle S_i^-\rangle, \tag{191}$$

the coherence intensity

$$w \equiv \frac{1}{S^2 N(N-1)}\sum_{i\neq j}^{N}\langle S_i^+ S_j^-\rangle, \tag{192}$$

and the spin polarization

$$s \equiv \frac{1}{SN}\sum_{i=1}^{N}\langle S_i^z\rangle, \tag{193}$$

where the angle brackets denote the averaging over the spin degrees of freedom, interpreting the local fields (183) as random variables with the stochastic averages (41). For the products of spin operators with different indices, we employ the

stochastic mean-field approximation (42), while for the operators with coinciding indices, we use the decoupling

$$\langle S_i^- S_i^z + S_i^z S_i \rangle = \left(2 - \frac{1}{S} \right) \langle S_i^- \rangle \langle S_i^z \rangle$$

resulting from Eq. (186).

The uniform molecular spin-wave frequency (187) becomes

$$\omega_D = \omega_0 - (2S-1)\frac{D}{\hbar} s, \tag{194}$$

with $\omega_0 \equiv \gamma_S B_0$. Then for the functions (191) to (193) we derive the same equations (47), except that the first of these equations appears as

$$\frac{du}{dt} = -i(\omega_D + \xi_0 - i\Gamma_2)u + fs, \tag{195}$$

with ω_0 replaced by ω_D. Note that ω_0 is always positive, while ω_D, given by Eq. (194), can be both positive and negative.

To deduce the expression for the resonator feedback field, we resort to the integral Eq. (20). In analogy with Eq. (57), we find

$$\frac{\mu_0 H}{\hbar} = i(\alpha_D u - \alpha_D^* u^*) + 2\beta \cos \omega t,$$

where, instead of Eq. (58), we have the coupling function

$$\alpha_D = \Gamma_0 \omega_D \left[\frac{1 - \exp\{-i(\omega - \omega_D)t - \gamma t\}}{\gamma + i(\omega - \omega_D)} + \frac{1 - \exp\{i(\omega + \omega_D)t - \gamma t\}}{\gamma - i(\omega + \omega_D)} \right], \tag{196}$$

in which the natural width is

$$\Gamma_0 \equiv \pi \eta \rho \gamma_S \mu_S \qquad (\mu_S \equiv \hbar \gamma_S S). \tag{197}$$

An efficient coupling with the resonator occurs only when $\omega \approx \omega_D$, if $\omega_D > 0$, or when $\omega \approx -\omega_D$, if $\omega_D < 0$. This implies the resonance condition

$$\frac{|\Delta_D|}{\omega} \ll 1, \qquad \Delta_D \equiv \omega - |\omega_D|.$$

If the resonance is sharp, such that $|\Delta_D| < \gamma$, then the coupling function (196) reduces to

$$\alpha_D = g_D \Gamma_2 \left(1 - \mathrm{e}^{-\gamma t} \right), \tag{198}$$

where the spin-resonator coupling is

$$g_D \equiv \frac{\gamma \Gamma_0 \omega_D}{\Gamma_2(\gamma^2 + \Delta_D^2)}. \tag{199}$$

In this way, introducing the effective force

$$f_1 \equiv -i\nu_0 - i(\nu_1 + \beta)\, \mathrm{e}^{-i\omega t} + \xi,$$

we come to the evolution equations

$$\begin{aligned} \frac{du}{dt} &= -i(\omega_D + \xi_0)u - (\Gamma_2 - \alpha_D s)u + f_1 s, \\ \frac{dw}{dt} &= -2(\Gamma_2 - \alpha_D s)w + (u^* f_1 + f_1^* u)\, s, \\ \frac{ds}{dt} &= -\alpha_D w - \frac{1}{2}\,(u^* f_1 + f_1^* u) - \Gamma_1(s - \zeta). \end{aligned} \tag{200}$$

Here we have omitted the nonresonant terms, corresponding to the last terms of Eqs. (64), which do not contribute to the equations for the guiding centres in the averaging technique. Using this technique, we obtain the guiding-centre equations of the same form as Eqs. (70), but with α, ω_0, and Δ replaced by α_D, ω_D, and Δ_D, respectively. The following description of possible regimes of coherent radiation by magnetic molecules is the same as that for nuclear spins.

First of all, it is necessary to stress that the electromagnetic interaction of radiating magnetic dipoles through the common radiation field can never produce coherent radiation. This was explained in detail in Subsec. 5.1, whose results are valid for arbitrary spins, whether these are nuclear or molecular spins. The impossibility of collectivizing the spin motion by means of the magnetodipole radiation is caused by the fact that the corresponding radiation time $T_{\rm rad} \gg T_2$ is many orders larger than the spin-spin dephasing time due to dipolar spin interactions. This is expressed by inequality (85). Therefore, the assumption [111] that molecular nanomagnets could exhibit superradiance caused by magnetodipole radiation is wrong. In addition, the regime considered in Ref. 111 requires a very strong transverse magnetic field of about 6 T$\sim 10^5$ G. Such a strong transverse field suppresses the single-site anisotropy, whose effective field is $B_D \equiv (2S-1)D/\hbar\gamma_S$. The latter, e.g., for Mn_{12}, with $\gamma_S \sim 10^7$ $\mathrm{G}^{-1}\cdot\mathrm{s}^{-1}$ and $D/\hbar \sim 10^{11}$ s^{-1}, is of the order of $B_D \sim 10^5$ G. Any magnetic relaxation in the presence of such a strong transverse magnetic field has nothing to do with superradiance, which is a self-organized spontaneous radiation. When a transverse field induces spin relaxation, this is called spin induction. Even if a kind of spin induction could be realized for magnetic molecules, the related relaxation time would be of the order of T_2. So, it would be just a simple spin induction, without any collective effects.

In order to realize the real spin superradiance by magnetic molecules, it is necessary to couple them to a resonant electric circuit, whose feedback field could collectivize the spin motion. When there are no transverse external fields, as it should be for the regime of pure superradiance, the process is triggered by

molecular spin waves. Then the arising feedback field can make the spin motion coherent, in the same way as it has been considered above for nuclear spins. The difference with the latter is the presence of magnetic anisotropy that may complicate the experimental realization of molecular spin superradiance.

From the evolution equations (200) it is seen that the increase of the transverse coherence occurs when the effective attenuation $\Gamma_2 - g_D s$ becomes negative, which requires that the spin-resonator coupling (199) be positive and sufficiently large. To have positive g_D, one needs that the effective frequency (194) be also positive, that is

$$\omega_0 > (2S-1)\frac{D}{\hbar}s.$$

This happens, assuming that $s > 0$, when D is negative, so that the molecules form an easy-plane nanomagnet, or, if $D > 0$, when the applied longitudinal field B_0 is sufficiently strong. For instance, in the case of Fe_8, the anisotropy field is $B_D \sim 10^4$ G. Hence, the external field has to be stronger than 1 T.

Another complication is that the frequency (194) is, actually, a function of time through $s = s(t)$. Therefore, to organize the resonance condition $\omega \approx \omega_D$, one has, in general, to vary either the resonator natural frequency ω or the external field B_0, so that to achieve the approximate equality

$$\gamma_S B_0 \cong \omega + (2S-1)\frac{D}{\hbar}s \qquad (\Delta_D = 0).$$

In principle, such a temporal varying of the system parameters, for achieving the resonance conditions, is feasible and is known for optical systems [5], where it is called *chirping*. The chirping technique can also be used for realizing superradiance from magnetic molecules [60].

The situation becomes simple when $S = 1/2$, so that the anisotropy disappears, or when $\omega_0 \gg (2S-1)D/\hbar$. Then $\omega_D \approx \omega_0$, and all the consideration reduces to the same as has been done above for nuclear spins. Another possibility is 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 the permanent pumping, as it was done for the nuclear spin $I = 5/2$ of ^{27}Al in experiments [30–33]. In the latter case, solely the regime of pulsing spin superradiance can be achieved.

In this way, though the presence of magnetic anisotropy complicates the experimental realization of coherent spin radiation by magnetic molecules, nevertheless there are several possibilities for reaching the required conditions and producing such a coherent radiation. This could be used for superradiant operation of spin masers [60]. Radiation from magnetic molecules would be essentially stronger than that from nuclear spins. And not only the current power (129) could be easily measured, but radiation intensity (127) would also be very high. To estimate the radiation intensity (127), let us take $\omega_D \sim \omega_0 \sim 10^{12}$ Hz, which

corresponds to microwaves with the wavelength $\lambda \sim 0.1$ cm. For the parameters typical for Mn_{12} or Fe_8, we have $\Gamma_2 = n_0 \rho \gamma_S \mu_S \sim 10^{10}$ s^{-1} and $\Gamma_0 = 0.1\Gamma_2$. The spin-resonator coupling (199) is quite large, $g_D \sim 10^5$. The radiation intensity $\overline{I} \sim \mu_S^2 \omega_0^4 N^2 / c^3$, for $N \sim 10^{20}$, reaches the value $\overline{I} \sim 10^{11}$ W. The superradiant pulse can be very short. The pulse time $\tau_p \approx T_2 / g s_0$ can be as short as 10^{-15} s. So, it is feasible to realize femtosecond superradiant pulses. The possibility of producing coherent radiation by magnetic molecules can find various applications.

10. PION AND DIBARYON RADIATION

Nuclear systems under extreme conditions can emit not only photons but also other particles. If the emitted particles are bosons, they can form coherent beams, similarly to the coherent photon radiation. The required extreme conditions can be experimentally achieved in the process of nuclear and heavy-ion collisions. Then, for instance, multiple pions can be produced. If the density of the latter is sufficiently high, they can form a coherent state, thus, providing the feasibility of getting a pion laser [112]. When the energy of colliding nuclei is high, they can form fireballs of nuclear matter, having temperature and density sufficient for realizing the deconfinement transition [113–118]. Then, except pions, other bosons can be emitted, such as dibaryons and gluons, presenting the possibility of obtaining different nuclear-matter lasers [119].

In order to find out the conditions under which the massive creation of bosons in nuclear matter could be achieved, it is necessary to analyze the equation of state of hot and dense nuclear matter, taking into account various channels of reactions where composite particles could be formed. To our mind, the most general approach for reaching this goal is presented by the *theory of clustering matter* [117, 120, 121]. This approach is based on three pivotal concepts: cluster representation, statistical correctness, and potential scaling.

The idea of *cluster representation* goes back to the works studying the abundances of chemical elements by treating each element as a quasiparticle characterized by its atomic weight and binding energy, with the related chemical potentials taking account of the allowed chemical reactions [122]. The problem of constructing an accurate quasiparticle representation for composite particles was initiated by Weinberg [123–125]. The most mathematically elaborated approach was formulated by Girardeau [126–128], whose method was applied to different systems containing bound clusters, including quark-hadron matter [129, 130].

The basic points in the cluster picture are as follows. Let us consider a multiparticle system with the total space of quantum states being a Fock space $\mathcal{F}$, on which the algebra of observables $\mathcal{A}$ is defined. Let among these quantum

states be free states as well as different bound states corresponding to clusters of several particles. Each type of bound clusters can be individualized by a set of characteristic parameters, such as the compositeness number z_i, showing the number of elementary particles bound into a cluster, effective mass of the cluster m_i, its baryon number B_i, strangeness S_i, and so on. And let us treat each type of these bound clusters as a separate sort of particles, with the associated Fock space $\mathcal{F}_i$ called the ideal cluster space. The *total cluster space* is defined as the tensor product

$$\mathcal{F}_T \equiv \otimes_i \mathcal{F}_i \otimes \mathcal{F} \tag{201}$$

termed the *Fock–Tani space* [126–128, 131]. The relation between the Fock space of elementary particles and the Fock–Tani cluster space (201) is presented by means of the unitary Tani mapping $\hat{U}_T^+ = \hat{U}_T^{-1}$, so that

$$\mathcal{F} = \hat{U}_T \mathcal{F}_T, \qquad \mathcal{F}_T = \hat{U}_T^+ \mathcal{F}. \tag{202}$$

The cluster algebra of observables is given by the Fock–Tani representation

$$\mathcal{A}_T \equiv \hat{U}_T^+ \mathcal{A} \hat{U}_T. \tag{203}$$

The latter definition guarantees that all matrix elements of the algebra $\mathcal{A}$ on $\mathcal{F}$ are the same as those of $\mathcal{A}_T$ on $\mathcal{F}_T$, since $\mathcal{F}_T^+ \mathcal{A}_T \mathcal{F}_T = \mathcal{F}^+ \mathcal{A} \mathcal{F}$. As far as the representation of $\mathcal{A}_T$ on $\mathcal{F}_T$ is isomorphic to that of $\mathcal{A}$ on $\mathcal{F}$, all observable quantities are the same in the standard picture of elementary particles and in the quasiparticle picture of the cluster system.

The Tani mapping is constructed in the following way. Let $a_i(x)$ be the field operator of elementary particles, defined on the Fock space $\mathcal{F}$, with x being a set of spatial variables. Suppose $\varphi_p(x_1, x_2, \ldots, x_p)$ is a Schrödinger wave function describing a bound state of p elementary particles. The field operator of the associated bound cluster, defined on the same space $\mathcal{F}$, can be presented as

$$\Psi_p(x) \equiv \int \varphi_p(x_1 - x, x_2 - x, \ldots, x_p - x) a_1(x_1) a_2(x_2) \cdots a_p(x_p) dx_1 dx_2 \cdots dx_p.$$

The image of this bound state on the ideal cluster space $\mathcal{F}_p$ is given by a p-particle cluster described by the field operator $\psi_p(x)$. The Tani mapping is defined by means of the unitary transformation

$$\hat{U}_T \equiv \exp\left(\frac{\pi}{2}\,\hat{F}\right), \quad \hat{F} = \sum_p \int \left[\psi_p^\dagger(x)\Psi_p(x) - \Psi_p^\dagger(x)\psi_p(x)\right] dx,$$

where the summation implies that over all admissible clusters. All operators $a_i(x)$ and $\psi_p(x)$ satisfy the canonical commutation relations, depending on whether the considered particle or cluster is a boson or fermion.

The algebra of observables (203) is defined on the Fock–Tani cluster space (201). In particular, one has the cluster Hamiltonian

$$H_T \equiv \hat{U}_T^+ H \hat{U}_T, \tag{204}$$

where H is the initial Hamiltonian in terms of elementary particles, given on $\mathcal{F}$. The cluster statistical state is $\langle \mathcal{A}_T \rangle$, with the angle brackets implying the statistical averaging related to the cluster Hamiltonian (204). The density of the j-type cluster is

$$\rho_j = \frac{1}{V} \langle \hat{N}_j \rangle, \qquad \hat{N}_j \equiv \int \psi_j^\dagger(x) \psi_j(x) dx,$$

where V is the system volume. We may introduce the *cluster probability*

$$w_j \equiv z_j \frac{\rho_j}{\rho} \qquad \left(\rho \equiv \sum_j z_j \rho_j \right), \tag{205}$$

characterizing the statistical weight of the j-type clusters. This probability satisfies the standard properties, being semipositive and normalized,

$$0 \leqslant w_j \leqslant 1, \qquad \sum_j w_j = 1.$$

The direct construction of the cluster Hamiltonian (204) is a rather tedious and complicated procedure. Moreover, the resulting Hamiltonian is presented by an infinite series. Because of this it is customary to define an effective Hamiltonian whose construction is based on physical reasoning. Generally, an effective Hamiltonian $H_{\rm eff} = H_{\rm eff}(\{\rho_j\}, T)$ includes an explicit dependence on thermodynamic variables, such as cluster densities ρ_j and temperature T. However, including thermodynamic variables into the Hamiltonian may break the validity of the general thermodynamic relations.

The correct cluster Hamiltonian has to be defined so that to preserve all thermodynamic relations. This is the meaning of the principle of *statistical correctness* [117, 120]. For this purpose, the effective Hamiltonian is complimented by an additional term, guaranteeing the statistical correctness of the resulting total cluster Hamiltonian

$$H_T = H_{\rm eff} + CV, \tag{206}$$

which assumes the validity of the equations

$$\left\langle \frac{\partial H_T}{\partial \rho_j} \right\rangle = 0, \qquad \left\langle \frac{\partial H_T}{\partial T} \right\rangle = 0. \tag{207}$$

Choosing C as a nonoperator quantity yields the equations

$$\frac{\partial C}{\partial \rho_j} = -\frac{1}{V}\left\langle \frac{\partial H_{\rm eff}}{\partial \rho_j}\right\rangle, \qquad \frac{\partial C}{\partial T} = -\frac{1}{V}\left\langle \frac{\partial H_{\rm eff}}{\partial T}\right\rangle, \tag{208}$$

defining $C = C(\{\rho_j\}, T)$. These conditions guarantee the correctness of all thermodynamic relations, such as

$$\begin{aligned} P = -\frac{\partial \Omega}{\partial V} = -\frac{\Omega}{V}, \quad \varepsilon = T\frac{\partial P}{\partial T} - P + \mu_B n_B + \mu_S n_S = \frac{1}{V}\langle H_T\rangle, \\ s = \frac{\partial P}{\partial T} = \frac{1}{T}\left(\varepsilon + P - \mu_B n_B - \mu_S n_s\right), \\ n_B = \frac{\partial P}{\partial \mu_B} = \sum_j B_j \rho_j, \qquad n_S = \frac{\partial P}{\partial \mu_S} = \sum_j S_j \rho_j, \end{aligned} \tag{209}$$

in which Ω is the grand potential; ε and s are the energy and entropy densities; P is pressure; μ_B is baryon potential; μ_S is strangeness potential; n_B and n_S are the baryon and strangeness densities, and

$$\Omega = -T \ln {\rm Tr}\, {\rm e}^{-\beta \tilde{H}}, \qquad \tilde{H} = H_T - \sum_j \mu_j N_j,$$

$$\rho_j = \frac{\partial P}{\partial \mu_j} = \frac{1}{V}\langle \hat{N}_j\rangle, \qquad \mu_j = \mu_B B_j + \mu_S S_j,$$

with μ_j being the chemical potential; and β, inverse temperature.

The cluster Hamiltonian (206) contains the terms describing effective interactions between different clusters. Defining the related interaction potentials is done by means of the principle of *potential scaling* [117, 120]. According to this, the interaction potentials from the same class of universality are connected by the scaling relation

$$\frac{\Phi_{ij}}{z_i z_j} = \frac{\Phi_{ab}}{z_a z_b}. \tag{210}$$

This principle allows the definition of all qualitatively equivalent interaction potentials through one known universal potential. Another admissible form of the potential scaling could be

$$\frac{\Phi_{ij}}{m_i m_j} = \frac{\Phi_{ab}}{m_a m_b},$$

since masses are usually proportional to the corresponding compositeness numbers, $m_j \sim z_j$.

The theory of clustering matter has been applied to clustering quark-hadron matter, with elementary particles being quarks, antiquarks, and gluons, while hadrons being various bound clusters of these elementary particles [117, 120].

The results of this consideration are presented in the following Figs. 5–16, where the energy is measured in units of $J = 225$ MeV, temperature $\Theta = k_B T$

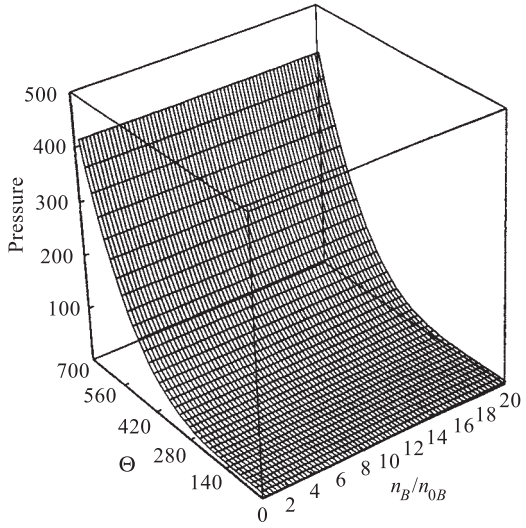

Fig. 5. Pressure (in units of J^4) of the clustering quark-hadron matter

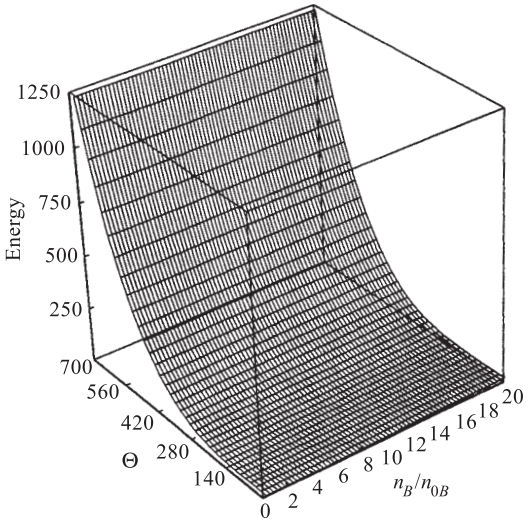

Fig. 6. Energy density (in units of J^4) on the temperature-baryon density plane

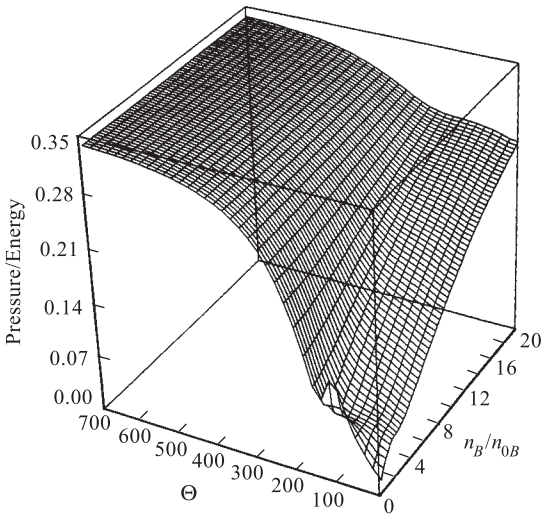

Fig. 7. Pressure-to-energy density ratio defining the effective sound velocity squared $c_{\text{eff}}^2 \equiv P/\varepsilon$, with a maximum at $\Theta_d = 160$ MeV related to the deconfinement

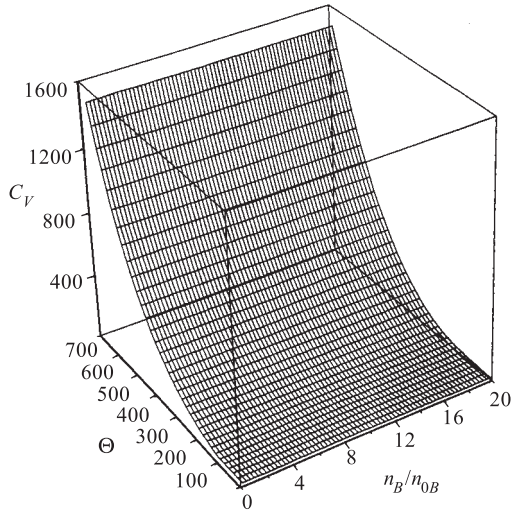

Fig. 8. Specific heat $C_V = \partial\varepsilon/\partial T$ (in units of J^3)

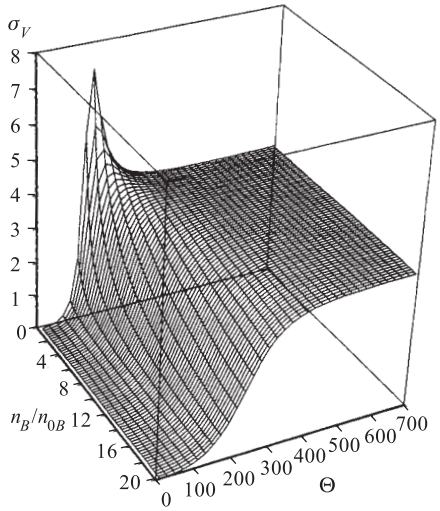

Fig. 9. Reduced specific heat $\sigma_V = TC_V/\varepsilon$ displaying a maximum at $\Theta_d = 160$ MeV associated with the deconfinement crossover

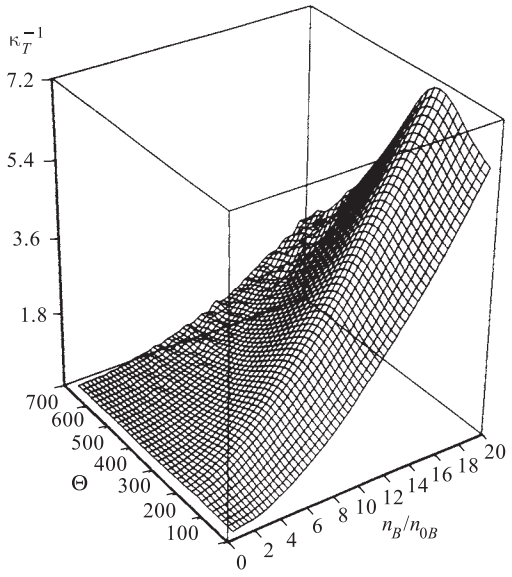

Fig. 10. Compression modulus $\kappa_T^{-1} = n_B \partial P/\partial n_B$ (in units of J^4) also having a maximum at the deconfinement crossover

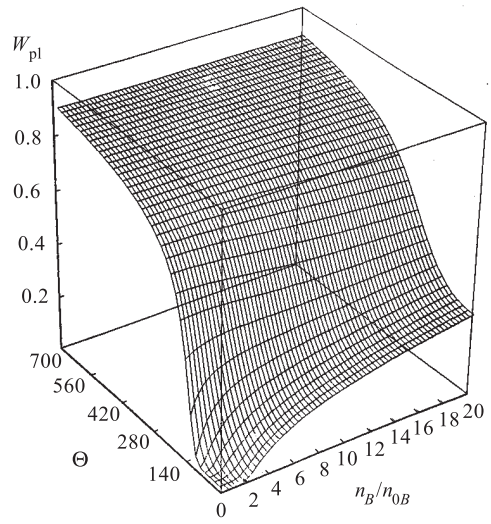

Fig. 11. Cluster probability of the quark-gluon plasma, being the sum of the probabilities of quarks, antiquarks, and gluons

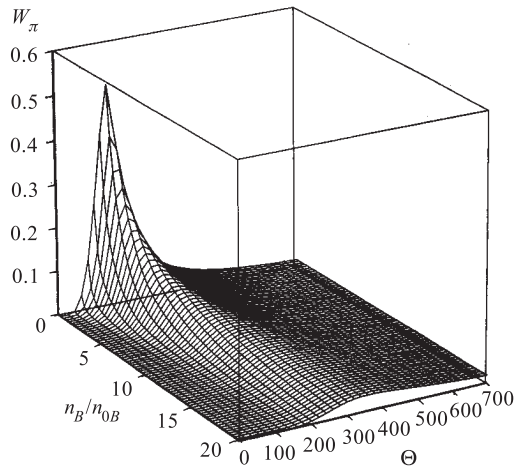

Fig. 12. Pion probability, being the sum of the probabilities of π^+, π^-, and π^0 mesons, with a sharp maximum at the deconfinement crossover

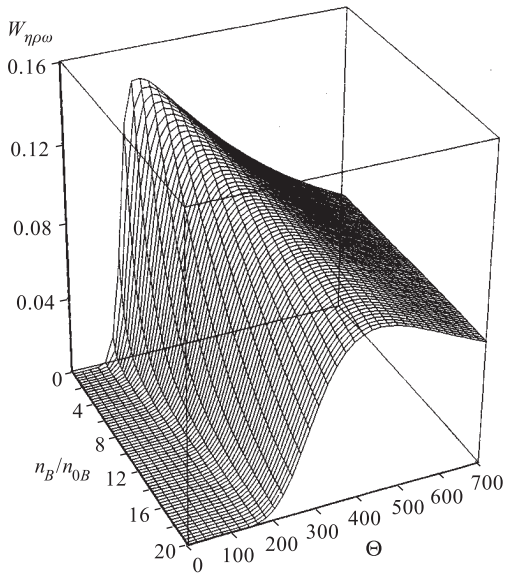

Fig. 13. Summary probability of η, ρ^+, ρ^-, ρ^0, and ω mesons

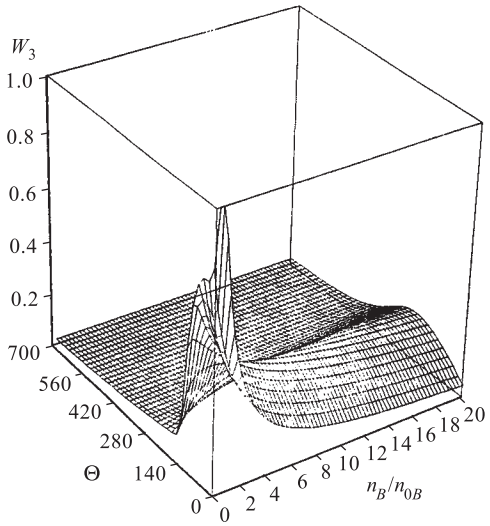

Fig. 14. Nucleon probability, being the sum of the probabilities of neutrons, protons, antineutrons, and antiprotons

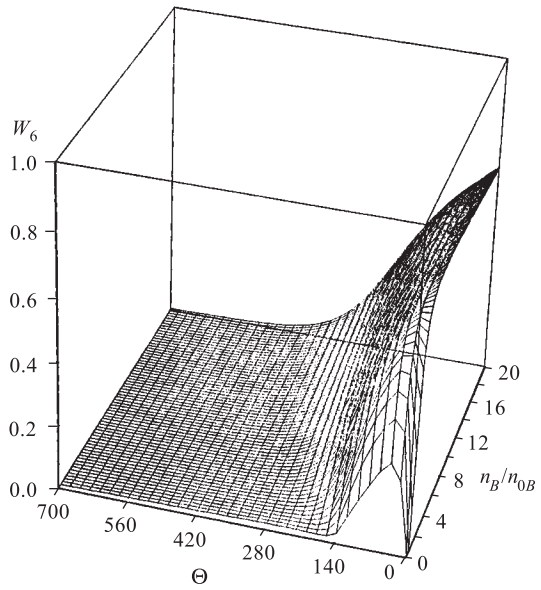

Fig. 15. Dibaryon probability, which is the sum of the dibaryon and antidibaryon weights

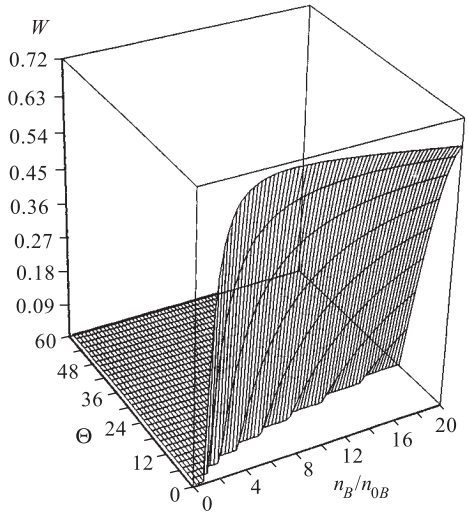

Fig. 16. Probability of the Bose–Einstein condensate of dibaryons

is given in MeV, $n_{0B} = 0.167$ fm^{-3} is the normal baryon density. These results demonstrate that the deconfinement transition at finite baryon density, and at conditions typical of heavy-ion collisions, is a gradual crossover. The deconfinement transition can be associated with a point where some reduced quantities display a maximum. The largest amount of pions is produced around the deconfinement temperature $\Theta_d = 160$ MeV and at the low baryon density $n_B < n_{0B}$. A high concentration of dibaryons can be achieved at low temperature $\Theta < 20$ MeV and the baryon density $n_B \approx (5 - 20)n_{0B}$. Dibaryons can form the Bose–Einstein condensate, thus, being in a coherent state. Producing high concentrations of pions or dibaryons, under the corresponding conditions, can be employed for creating pion and dibaryon lasers [119]. For producing a large amount of gluons, very high temperatures, $\Theta > 160$ MeV, are required. But gluons cannot be emitted as free particles.

CONCLUSION

We presented a detailed description of coherent radiation by nuclear spins. It is important to stress that pure spin superradiance was first experimentally observed in Dubna. The theoretical consideration of coherent nuclear spin radiation is based on the theory developed by the authors. The mathematical basis of this theory is the scale separation approach.

We show that at sufficiently strong external magnetic fields in the system of nuclear spins there appear nuclear spin waves, which play the role of the triggering mechanism starting the incoherent motion of spins. Neither magnetodipole radiation nor the resonator Nyquist noise can initiate spin motion. After the spins started moving, the transverse transition coherence of their evolution is due to the action of the feedback magnetic field formed by the resonant electric circuit. This resonator field is crucial for developing coherent spin motion. Different regimes of coherent spin radiation are investigated. Electron-nuclear hyperfine coupling can lead to an essential enhancement of nuclear spin radiation. Superradiance by magnetic molecules is feasible, which necessarily requires the presence of a resonant electric circuit and cannot be achieved without it. The time dependence of the transition frequency, caused by the single-site magnetic anisotropy, can be compensated by the chirping effect. The possibility of pion and dibaryon radiation from excited nuclear matter is discussed.

The mathematical techniques, presented in this review for treating strongly nonequilibrium spin systems, are rather general and can be employed for analyzing nonlinear dynamics of arbitrary spin or quasispin essemblies. A very interesting application of these techniques would be to investigating the nonequilibrium phenomena occurring in dilute gases of cold trapped atoms [95, 132–135]. Such atoms can be spin-polarized, forming at low temperatures the spinor Bose–Einstein

condensates. The nonequilibrium dynamics of polarized spinor condensates can exhibit coherent phenomena similar to those happening in nuclear spin systems.

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