Strong EPR saturation under phonon-bottleneck conditions

L. L. Buishvili, N. P. Giorgadze, M. Z. Zviadadze, and A. I. Ugulava

Physics Institute, Georgian Academy of Sciences
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A quantum-statistical theory of strong saturation of EPR under phonon-bottleneck conditions is developed. Time equations for the spin and phonon temperatures are derived and these equations are analyzed in various limiting cases. The expression obtained for the stationary value of the spin temperature in the rotating coordinate frame goes over into the well-known result of Redfield if it is assumed that the resonance phonons are in equilibrium with a thermostat. The conditions for the appearance of a phonon avalanche in pulsed saturation of EPR in oscillating fields of the order of and greater than the local field are discussed.

1. Discussion of the effects associated with heating of phonons in magnetic-resonance saturation and in the dynamical polarization of nuclei began long ago[1]. Recently, interest in these problems has grown in view of the successes of the concept of the dipole-dipole reservoir (DDR) in EPR[2,3]. A systematic theory of the phonon bottleneck (PB) with the DDR taken into account was constructed in the papers[4-6]. In the paper[6] it was shown experimentally that for pulsed saturation of an EPR with detuning there arises a phonon avalanche caused (as follows from the theoretical analysis) by a sharp change in the temperature of the DDR.

In all the above-mentioned papers the oscillating field was assumed to be small compared with the local field due to the spin-spin interaction (the so-called Provotorov case[2]). On the other hand, in experiments on strong EPR saturation radio-frequency fields of the order of the local fields are used (the so-called Redfield case[3,7,8]). In view of this, it is of interest to investigate the effects of phonon heating and of the formation of a phonon avalanche under conditions of strong saturation of the magnetic resonance.

2. Following the paper[5], we write the Hamiltonian of the coupled spin-phonon system in the form ($\hbar = 1$)

$$\mathcal{H}=\mathcal{H}_z+\mathcal{H}_d+\mathcal{H}_{ph}+\mathcal{H}_{s,h}+\mathcal{H}_{s,ph},$$

$$\mathcal{H}_z=\omega_s S_z,\quad \mathcal{H}_{ph}=\sum_{\mathbf{k}j}\omega_{\mathbf{k}j}a_{\mathbf{k}j}^+a_{\mathbf{k}j}, \tag{1}$$

$$\mathcal{H}_{s,h}=\frac{\omega_1}{2}(S^+e^{-i\Omega t}+S^-e^{i\Omega t}),\quad \mathcal{H}_{s,ph}=\frac{1}{2}\sum_{\mathbf{k}j}(L_{\mathbf{k}j}^+S^-+L_{\mathbf{k}j}^-S^+),$$

where $\mathcal{H}_Z$ is the Zeeman energy of the spin system, $\mathcal{H}_d$ is the secular part of the dipole-dipole interaction (this is not written out explicitly), $\mathcal{H}_{ph}$ is the Hamiltonian of the phonons in the harmonic approximation, $\mathcal{H}_{s,h}$ is the energy of interaction of the spin system with the oscillating field $h^{\pm}=\omega_1\gamma^{-1}e^{\pm i\Omega t}$, $a_{\mathbf{k}j}^+$ and $a_{\mathbf{k}j}$ are the creation and annihilation operators for phonons of the j-th branch with wave-vector $\mathbf{k}$ and frequency $\omega_{\mathbf{k}j}$, and $\mathcal{H}_{s,ph}$ is the spin-phonon interaction energy.

In accordance with the assumption of single-phonon relaxation, the lattice operators $L_{\mathbf{k}j}^{\pm}$ can be represented in the form[5,9]

$$L_{\mathbf{k}j}^+=(\mathcal{A}\omega_{\mathbf{k}j})^{1/2}a_{\mathbf{k}j}^+,\quad L_{\mathbf{k}j}^-=(L_{\mathbf{k}j}^+)^+, \tag{2}$$

where $\mathcal{A}$ is the spin-phonon coupling constant[1)].

We note that up to now we have excluded the thermostat from consideration. The relaxation of phonons to the thermostat will be taken into account phenomenologically later.

To eliminate the explicit time dependence of the Hamiltonian we perform the unitary transformation:

$$\Psi^*=U\Psi,\quad U=\exp\left\{i\Omega\left(S^z+\sum_{\mathbf{k}j}a_{\mathbf{k}j}^+a_{\mathbf{k}j}\right)t\right\}. \tag{3}$$

This transformation with respect to the spin system represents a change to a coordinate frame rotating with frequency Ω about the direction of the constant magnetic field (the z axis). With respect to the phonons, it can be regarded as a shift in energy space through a frequency Ω. In the following we shall call the new frame of reference the rotating coordinate frame (RCF).

It is easy to see that in the RCF the Hamiltonian of the spin-phonon system has the form

$$\mathcal{H}^*=\mathcal{H}_s^*+\sum_j\int_0^{k_j}dk\mathcal{H}_{kj}^*+\frac{1}{2}\sum_j\int_0^{k_j}dk(L_{kj}^+S^-+L_{kj}^-S^+), \tag{4}$$

where

$$\mathcal{H}_s^*=(\omega_s-\Omega)S^z+\omega_1S^x+\mathcal{H}_d,\quad \mathcal{H}_{\mathbf{k}j}^*=(\omega_{\mathbf{k}j}-\Omega)a_{\mathbf{k}j}^+a_{\mathbf{k}j},$$

$$\mathcal{H}_{kj}^*=\int\frac{d\Omega}{4\pi}\frac{k^2}{2\pi^2}\mathcal{H}_{\mathbf{k}j}^*;\quad L_{kj}^{\pm}=\int\frac{d\Omega}{4\pi}\frac{k^2}{2\pi^2}L_{\mathbf{k}j}^{\pm},$$

i.e., does not depend explicitly on time. Here, inasmuch as the oscillating field is not assumed to be small compared with the local field, the interaction of the spin system with the oscillating field is included in the basic Hamiltonian and the role of the small perturbation is reserved for the spin-lattice interaction.

The system under consideration can be described statistically by a density matrix $\rho^*(t)$ satisfying the Liouville equation:

$$i\frac{\partial\rho^*}{\partial t}=[\mathcal{H}^*,\rho^*(t)]. \tag{5}$$

Since the Hamiltonian of the system does not depend explicitly on time, for this description we can use the quantum-statistical method proposed by Zubarev[10] for constructing a nonequilibrium statistical operator (NSO).

3. We shall assume that the time for establishment of internal equilibrium in the spin system and in phonons of specified frequencies is much shorter than the time for establishment of the equilibrium (or stationary) state in the spin-phonon system under consideration. Then in the first (short) stage of the evolution, the spin-phonon system reaches a quasi-equilibrium state in which it is a combination of weakly interacting subsystems—a spin packet and phonon packets of definite frequencies—in internal equilibrium. This state is described by a reduced set of thermodynamic parameters—the inverse temperatures of the spin subsystem (β_s^*) and phonon subsystems ($\beta_{\mathbf{k}j}^*$). The subsequent evolution of the spin-phonon system in the so-called

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macroscopic stage reduces to a slow approach to the equilibrium (or stationary) state as a result of the interaction between the subsystems. At this stage of the evolution, the spin-phonon system can be described by a NSO having the form

$$\rho^{*}=Q_0\exp\left\{-\beta_s^{*}\mathscr{H}_s^{*}-\sum_j\int_0^{k_j}dk\beta_{kj}^{*}\mathscr{H}_{kj}^{*}+\int_{-\infty}^{0}dte^{\varepsilon t}\left[\beta_s^{*}\mathscr{K}_s^{*}(t)+\sum_j\int_0^{k_j}dk\beta_{kj}^{*}\mathscr{K}_{kj}^{*}(t)\right]\right\},\quad \varepsilon>0, \tag{6}$$

where, as can be seen easily, the operators of the fluxes are given by the relations

$$\mathscr{K}_s^{*}=-\frac{1}{2}(\dot{S}^{+}L^{-}+\dot{S}^{-}L^{+}),\quad \mathscr{K}_{kj}^{*}=\frac{1}{2}(S^{-}\dot{L}_{kj}^{+}+S^{+}\dot{L}_{kj}^{-}),$$
$$\dot{S}^{\pm}=\frac{1}{i}[S^{\pm},\mathscr{H}_s^{*}],\quad \dot{L}_{kj}^{\pm}=\frac{1}{i}[L_{kj}^{\pm},\mathscr{H}_{kj}^{*}], \tag{7}$$
$$L^{\pm}=\sum_j\int_0^{k_j}dkL_{kj}^{\pm},\quad L_{kj}^{\pm}=\int\frac{d\Omega}{4\pi}\frac{k^2}{2\pi^2}L_{kj}^{\pm}$$

and satisfy a condition following from the energy conservation law:

$$\mathscr{K}_s^{*}+\sum_j\int_0^{k_j}dk\mathscr{K}_{kj}^{*}=0. \tag{8}$$

Using this relation, confining ourselves to the high-temperature approximation with respect to the spins ($\beta_S^{*}\mathscr{H}_S^{*}\ll 1$), and also taking into account that the thermodynamic fluxes contain the weak interaction, we bring the expression (6) for the NSO to the following form, convenient for practical calculations:

$$\rho^{*}\approx\rho_L^{*}\left\{1-\beta_s^{*}\mathscr{H}_s^{*}+\sum_j\int_0^{k_j}dk\int_0^1 d\lambda\int_{-\infty}^{0}dte^{\varepsilon t}e^{A\lambda}(\beta_{kj}^{*}-\beta_s^{*})\mathscr{K}_{kj}^{*}(t)e^{-A\lambda}\right\}, \tag{9}$$

where

$$\rho_L^{*}=e^{-A}/\mathrm{Sp}\,e^{-A},\quad A=\sum_j\int_0^{k_j}dk\beta_{kj}^{*}\mathscr{H}_{kj}^{*}.$$

Averaging now the Hamiltonians of the subsystems under consideration, and the thermodynamic fluxes, over the expression (9), using the relation $\overline{d\mathscr{H}_m/dt}=d\overline{\mathscr{H}}_m/dt$, and taking the condition for the system to be quasi-static, we obtain the following system of equations for the inverse temperatures[2]:

$$\frac{d\beta_s^{*}}{dt}=\frac{\pi}{(\omega_s-\Omega)^2+\omega_1^2+\omega_d^2}\int_0^{\omega_m}d\omega\omega(\omega-\Omega)f^{*}(\Omega-\omega)L^{+-}(\omega)\frac{\beta_0}{\beta^{*}(\omega)}(\beta^{*}(\omega)-\beta_s^{*}),$$
$$\frac{d\beta(\omega)}{dt}=-\pi\frac{c_s}{\bar{c}_p}L^{+-}(\omega)\frac{\omega-\Omega}{\omega}f^{*}(\Omega-\omega)\frac{\beta^{*}(\omega)}{\beta_0}(\beta^{*}(\omega)-\beta_s^{*}). \tag{10}$$

Here,

$$c_s=\frac{1}{3}N_s\omega_s^2S(S+1),\quad \bar{c}_p=\frac{3\omega_s^2}{2\pi^2c^3\beta_0^2},\quad L^{+-}(\omega)=\frac{3A\omega^2}{2\pi^2c^3\beta_0},$$
$$\omega_d^2=\frac{\langle(\mathscr{H}_d)^2\rangle}{\langle(S^z)^2\rangle},\quad f^{*}(\omega)=\frac{1}{2\pi}\int_{\infty}^{\infty}dte^{i\omega t}\frac{\langle S^{-}S^{+}(t)\rangle}{\langle S^{-}S^{+}\rangle},$$
$$S^{\pm}(t)=\exp(i\mathscr{H}_s^{*}t)S^{\pm}\exp(-i\mathscr{H}_s^{*}t),\quad \langle A\rangle=\mathrm{Sp}\,A/\mathrm{Sp}\,1,$$

where N_S is the number of paramagnetic ions in the sample and c is the sound velocity.

Below it is necessary to transform from the phonon temperature in the RCF to the phonon temperature in the laboratory frame.

Starting from the fact that the mean number of phonons is the same in the two coordinate frames, we conclude that

$$\beta(\omega)=\frac{\omega-\Omega}{\omega}\beta^{*}(\omega). \tag{11}$$

Eliminating $\beta^{*}(\omega)$ (by means of this relation) from the system of equations (10), we find

$$\frac{d\beta_s^{*}}{dt}=\frac{\pi}{(\omega_s-\Omega)^2+\omega_1^2+\omega_d^2}\times\int d\omega(\omega-\Omega)^2f^{*}(\Omega-\omega)L^{+-}(\omega)\frac{\beta_0}{\beta(\omega)}\left(\frac{\omega}{\omega-\Omega}\beta(\omega)-\beta_s^{*}\right),$$
$$\frac{d\beta(\omega)}{dt}=-\pi\frac{c_s}{\bar{c}_p}L^{+-}(\omega)\frac{\omega-\Omega}{\omega}f^{*}(\Omega-\omega)\frac{\beta(\omega)}{\beta_0}\left(\frac{\omega}{\omega-\Omega}\beta(\omega)-\beta_s^{*}\right)-\frac{\beta(\omega)}{\beta_0}\frac{\beta(\omega)-\beta_0}{T_0}. \tag{12}$$

The last term in the right-hand side of the second equation of the system (12) has been added phenomenologically and describes the simplest form of relaxation of the phonons to the thermostat, with a frequency-independent relaxation time T_0.[3)]

The system (12), which describes the rate of change of the inverse temperatures β_S^{*} and $\beta(\omega)$, is the starting point. With regard to it, it must be noted that the correlator appearing here is a nonsymmetric function of the frequency and differs substantially from the correlator encountered in the theory of intermediate saturation of EPR[2,3,5]. The first and second moments of this correlator, which are of interest from the standpoint of the present treatment, respectively have the form

$$M_1=\omega_s-\Omega,\quad M_2=(\omega_s-\Omega)^2+{}^1/_2\omega_1^2+2\omega_d^2. \tag{13}$$

We proceed now to examine different particular cases.

4. We assume first that the PB effect is absent and the resonance phonons remain in equilibrium with a thermostat at temperature β_0^{-1}. Then, taking the relations (13) into account, we can bring the first equation of the system (12) to the form

$$\frac{d\beta_s^{*}}{dt}=-\frac{1}{T_{sp}^{*}}(\beta_s^{*}-\beta_s^{*st}), \tag{14}$$

$$\beta_s^{*st}=\frac{\omega_s(\omega_s-\Omega)}{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2+2\omega_d^2}\beta_0, \tag{15}$$

$$\frac{1}{T_{sp}^{*}}=\frac{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2+2\omega_d^2}{(\omega_s-\Omega)^2+\omega_1^2+\omega_d^2}\frac{1}{T_{sp}}, \tag{16}$$

where $T_{sp}=[\pi L^{+-}(\omega_S)]^{-1}$ is the ordinary spin-lattice relaxation time.

The expressions (15) and (16) for the stationary value of the spin temperature and the spin-lattice relaxation time in the RCF were first obtained by Redfield[7], and later by a number of other authors on the basis of semi-phenomenological arguments[3,8]. The appearance of the numerical coefficients ½ and 2 is associated with this specific treatment.

5. We assume now that the PB effect occurs. We shall assume that the spectral diffusion in the phonon spectrum in the frequency interval of the order of EPR linewidth proceeds faster than the exchange between the spin system and the phonons. Then we can assign a single temperature β_p^{-1} to the resonance phonons interacting with the spin system, and regard them as a thermodynamic subsystem. The remaining phonons must be assigned to the thermostat.

The system of equations that describes the time evolution of β_S^{*} and β_p can be obtained easily from the initial system and has the form[4)]

$$\frac{d\beta_s^{*}}{dt}=-\frac{1}{T_{sp}^{*}}\left\{\beta_s^{*}-\frac{\omega_s(\omega_s-\Omega)}{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2+2\omega_d^2}\beta_p\right\}\frac{\beta_0}{\beta_p},$$

$$\frac{d\beta_p}{dt}=-\frac{1}{T_{ps}}\left\{\beta_p-\frac{\omega_s-\Omega}{\omega_s}\beta_s^{\cdot}\right\}\frac{\beta_p}{\beta_0}-\frac{\beta_p-\beta_0}{T_0}\frac{\beta_p}{\beta_0}, \qquad (17)$$

where $1/T_{ps} = (c_s/2\Delta\bar{c}_p)(1/T_{sp})$, and 2Δ is the width of the frequency spectrum of the resonance phonons.

The stationary solution of the system of equations (17) is given by the expressions

$$(\beta_s^{\cdot})^{st}=\frac{\omega_s(\omega_s-\Omega)}{(\omega_s-\Omega)^2+(1+\sigma)(^1/_2\omega_1^2+2\omega_d^2)}\beta_0,$$
$$(\beta_p)^{st}=\frac{(\omega_s-\Omega)^2+^1/_2\omega_1^2+2\omega_d^2}{(\omega_s-\Omega)^2+(1+\sigma)(^1/_2\omega_1^2+2\omega_d^2)}\beta_0, \qquad (18)$$

where $\sigma = T_0/T_{ps}$ is the PB coefficient.

In the absence of the PB effect ($\sigma \ll 1$), the second of these expressions gives $(\beta_p)^{st} \approx \beta_0$, while the Redfield result (15) follows naturally from the first. The presence of a PB leads to considerable heating of the resonance phonons, and also to an increase of the spin temperature in the RCF.

In conclusion, we remark that the relations (18) contain, as a particular case, Borghini's result referring to intermediate saturation ($\omega_1 \ll \omega_d$) under PB conditions[4]. In fact, for $\omega_1 \ll \omega_d$, the temperature $(\beta_S^*)^{-1}$ simply coincides with the DDR temperatures β_d^{-1}, and is connected with the Zeeman temperature by the relation

$$\beta_s^{-1}=\frac{\omega_s}{\omega_s-\Omega}(\beta_s^{\cdot})^{-1}.$$

6. We now consider the case when, under PB conditions, there is no spectral diffusion in the phonon spectrum (the so-called case of weak spectral diffusion). We shall assume first that the spin system is excited by a sufficiently short UHF pulse, during which it can be regarded as isolated from the lattice. Then, on expiry of this plate, it reaches a state described in the RCF by the inverse temperature

$$\beta_s^{\cdot}=\frac{\omega_s(\omega_s-\Omega)}{(\omega_s-\Omega)^2+\omega_1^2+\omega_d^2}\beta_0. \qquad (19)$$

The behavior of the resonance phonons over short times following the end of the pulse can be described by an equation for the deviation of the number of resonance phonons from the equilibrium value, i.e., for

$$Z(\omega)=\frac{n(\omega)-n_0}{n_0}\approx\frac{\beta_0-\beta(\omega)}{\beta(\omega)}.$$

This equation is of the form[5)]

$$\dot{Z}(\omega)+\frac{Z(\omega)}{T_0}=\frac{1}{T_{ps}^0}\frac{\omega^2}{\omega_s^2}\frac{f^{\cdot}(\Omega-\omega)}{f^{\cdot}(0)}\frac{(\omega_s-\Omega)(\omega_s-\omega)(\Omega/\omega)+\omega_1^2+\omega_d^2}{(\omega_s-\Omega)^2+\omega_1^2+\omega_d^2}$$
$$+\frac{1}{T_{ps}^0}\frac{\omega}{\omega_s}\frac{f^{\cdot}(\Omega-\omega)}{f^{\cdot}(0)}\frac{(\omega_s-\Omega)(\Omega-\omega)}{(\omega_s-\Omega)^2+\omega_1^2+\omega_d^2}Z(\omega), \qquad (20)$$
$$\frac{1}{T_{ps}^0}=\frac{c_s}{\bar{c}_p}f^{\cdot}(0)\frac{1}{T_{sp}}.$$

It follows from this equation that, under conditions of a strong PB effect ($\sigma_0 = T_0/T_{ps}^0 \gg 1$) for the resonance phonons whose frequencies satisfy the inequalities

$$(\omega_s-\Omega)(\omega_s-\omega)+\omega_1^2+\omega_d^2>0,\qquad(\omega_s-\Omega)(\Omega-\omega)>0,$$

$Z(\omega)$ increases first linearly, and then exponentially with time. In other words, in the frequency range indicated a phonon avalanche arises. The logarithmic increment of the avalanche is determined by the expression

$$\gamma(\omega)=\frac{\dot{Z}(\omega)}{Z(\omega)}\approx\frac{1}{T_{ps}^0}\frac{|\omega_s-\Omega||\Omega-\omega|}{(\omega_s-\Omega)^2+\omega_1^2+\omega_d^2}\frac{f^{\cdot}(\Omega-\omega)}{f^{\cdot}(0)}. \qquad (21)$$

In the limit of intermediate saturation ($\omega_1 \ll \omega_d$), the result of[6] follows from this.

We now consider stationary saturation of the EPR under conditions of a weak PB effect ($\sigma_0 \ll 1$). In this case, we can neglect the effect of heating of the resonance phonons on the behavior of the spin system, as a result of which the stationary value of the spin temperature in the RCF will be determined by the expression (15). As regards the stationary value of $Z(\omega)$, it can be obtained without difficulty from the second equation of the system (12), in which the relation (15) must be substituted for β_S^*. As a result, we have

$$Z(\omega)\approx\sigma_0\frac{f^{\cdot}(\Omega-\omega)}{f^{\cdot}(0)}\frac{\Omega^2}{\omega^2}\frac{(\omega_s-\Omega)(\omega_s-\omega)+^1/_2\omega_1^2+2\omega_d^2}{(\omega_s-\Omega)^2+^1/_2\omega_1^2+2\omega_d^2}. \qquad (22)$$

Hence it follows that, for strong saturation of the EPR at the center of the line ($\omega_S = \Omega$) under conditions of a weak PB effect, $Z(\omega)$ duplicates the nonsymmetric shape of the EPR line. But for saturation with detuning ($\omega_S \neq \Omega$), the spectral distribution of the resonance phonons has a fairly complicated form. Thus, e.g., for $\omega_S > \Omega$ the resonance phonons from the frequency interval

$$\omega<\omega_s+(\omega_s-\Omega)^{-1}(^1/_2\omega_1^2+2\omega_d^2)$$

are heated up, whereas in the opposite wing of the line cooling of the resonance phonons occurs[6]. In the limit of intermediate saturation the relation (22) goes over, naturally, into the result[6].

7. We proceed now to examine the case of ultrastrong saturation, when $\omega_1 \gg \omega_d$. First of all, by the unitary transformation

$$\tilde{S}^x=S^x\cos\theta-S^z\sin\theta,\quad \tilde{S}^y=S^y,\quad \tilde{S}^z=S^x\sin\theta+S^z\cos\theta, \qquad (23)$$

where

$$\sin\theta=\frac{\omega_1}{\omega_e},\quad \cos\theta=\frac{\omega_s-\Omega}{\omega_e},\quad \omega_e=\sqrt{(\omega_s-\Omega)^2+\omega_1^2},$$

we go over from the RCF to the so-called effective coordinate frame (ECF) in which the axis $\tilde{z}$ is directed along the effective magnetic field $\mathbf{H}_e = \gamma^{-1}[\mathbf{k}(\omega_S - \Omega) + \mathbf{i}\omega_1]$ (k and i are unit vectors directed respectively along the z and x axes of the RCF). The Hamiltonian of the system under consideration in the ECF can be obtained easily from the expression (4) and has the form

$$\tilde{\mathscr{H}}=\tilde{\mathscr{H}}_z+\lambda(\theta)\tilde{\mathscr{H}}_d+\sum_j\int_0^{k_j}dk\mathscr{H}_{kj}^{\cdot}+\tilde{\mathscr{H}}_{s,ph}, \qquad (24)$$

where $\tilde{\mathscr{H}}_Z = \omega_e\tilde{S}^Z$ is the Zeeman energy in the ECF, $\lambda(\theta)\tilde{\mathscr{H}}_d$ is the secular part of the dipole-dipole interaction in this same coordinate frame, and finally $\tilde{\mathscr{H}}_{s,ph}$ describes the spin-phonon interaction[6)]:

$$\tilde{\mathscr{H}}_{s,ph}=^1/_2\Lambda_+(\theta)(L^+\tilde{S}^-+L^-\tilde{S}^+)+^1/_2\Lambda_-(\theta)(L^+\tilde{S}^++L^-\tilde{S}^-).$$

Here,

$$\lambda(\theta)=^1/_2(3\cos^2\theta-1),\qquad \Lambda_\pm(\theta)=^1/_2(\cos\theta\pm1).$$

Since $\omega_e \gg \omega_d$ in the case considered, we must represent the spin system in the ECF in the form of a combination of two subsystems—the Zeeman subsystem and the DDR, assigning to each of them its inverse temperature ($\tilde{\beta}_S$ and $\tilde{\beta}_d$, respectively). As a result, we obtain the following expression for the NSO:

$$\tilde{\rho}\approx\rho_L^{\cdot}\Big\{1-\tilde{\beta}_s\tilde{\mathscr{H}}_z-\tilde{\beta}_d\tilde{\mathscr{H}}_d$$
$$+\int_0^1 d\lambda\int_{-\infty}^0 dte^{\varepsilon t}e^{A\lambda}\tilde{\mathscr{H}}_s^{\cdot}(t)e^{-A\lambda}(\tilde{\beta}_s-\tilde{\beta}_d)+\int_0^1 d\lambda\int_{-\infty}^0 dte^{\varepsilon t}e^{A\lambda}\mathscr{H}_{kj}^{\cdot}(t)e^{-A\lambda}(\beta_{kj}^{\cdot}-\tilde{\beta}_d)\Big\}. \qquad (25)$$

Averaging by means of this expression the flux operators

$$\tilde{\mathcal{H}}_s=\frac{\omega_e}{2i}\{\Lambda_+(\theta)(L^-\tilde{S}^+-L^+\tilde{S}^-)+\Lambda_-(\theta)(L^+\tilde{S}^+-L^-\tilde{S}^-)\},$$

$$\tilde{\mathcal{H}}_d=-\frac{1}{2}\{\Lambda_+(\theta)(L^+\dot{S}^-+L^-\dot{S}^+)+\Lambda_-(\theta)(L^+\dot{S}^++L^-\dot{S}^-)\},\qquad (26)$$

$$\tilde{\mathcal{H}}_{kj}=-\frac{1}{2}\{\Lambda_+(\theta)(L_{kj}^+\tilde{S}^-+L_{kj}^-\tilde{S}^+)+\Lambda_-(\theta)(L_{kj}^+\tilde{S}^++L_{kj}^-\tilde{S}^-)\}$$

and the Hamiltonians of the different subsystems, and also going over, for the phonons, to the laboratory frame in accordance with the relation (11), we obtain a system of equations describing the change in time of the inverse temperatures:

$$\frac{d\beta_s}{dt}=-\int d\omega\{\Phi(\omega,\omega_e)+\Phi(\omega,-\omega_e)\},$$

$$\frac{d\beta_d}{dt}=\int d\omega\left\{\frac{\Omega+\omega_e-\omega}{\omega_e}\Phi(\omega,\omega_e)-\frac{\Omega-\omega_e-\omega}{\omega_e}\Phi(\omega,-\omega_e)\right\},\qquad (27)$$

$$\frac{d\beta(\omega)}{dt}=\frac{c_e}{c_p'}\frac{\omega_e}{\omega}\{\Phi(\omega,\omega_e)-\Phi(\omega,-\omega_e)\}-\frac{\beta(\omega)}{\beta_0}\frac{\beta(\omega)-\beta_0}{T_0},$$

where

$$\Phi(\omega,\omega_e)$$
$$=\pi\Lambda_+^2(\theta)\tilde{f}(\Omega+\omega_e-\omega)L^{+-}(\omega)\frac{\beta_0}{\beta(\omega)}\left(\beta_s-\frac{\Omega+\omega_e-\omega}{\omega_e}\beta_d-\frac{\omega}{\omega_e}\beta(\omega)\right),$$

$$\tilde{f}(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dte^{i\omega t}\frac{\langle S^-\tilde{S}^+(t)\rangle}{\langle S^-S^+\rangle},\quad \tilde{S}^\pm(t)=e^{i\lambda(\theta)\tilde{\mathcal{H}}_d t}\tilde{S}^\pm e^{-i\lambda(\theta)\tilde{\mathcal{H}}_d t},$$

$$c_e=\frac{1}{3}N_s\omega_e^2S(S+1),\qquad c_p'=\frac{3\omega_e^2}{2\pi^2c^3\beta_0^2}.$$

It follows from the structure of the equations obtained that the spin system interacts with two groups of phonons, having central frequencies $\Omega-\omega_e$ and $\Omega+\omega_e$ and clearly separated by a frequency interval $2\omega_e$ that is very much greater than the EPR linewidth. The width of each group of resonance phonons is small compared with the width of the magnetic-resonance line, since the correlator $\tilde{f}(\omega)$ contains the factor $\lambda(\theta)$ in the outer factors of the operator $\tilde{S}^+(t)$. The presence of the two narrow groups of resonance phonons is in accordance with the fact that for ultrastrong saturation at frequency Ω resonance interaction with the detecting field occurs near the frequencies $\Omega\pm\omega_e$[11].

We note that the system (27) was obtained by neglecting the term S^zL^z in the Hamiltonian of the spin-phonon interaction. Allowance for this term would lead to excitation of phonons near the frequency ω_e (corresponding to the audio range), and this of interest from the point of view of rotational saturation. However, in the present paper we confine ourselves to treating processes associated with excitation of phonons in the UHF range.

In conclusion, we call attention to the fact that each different group of resonance phonons has its corresponding effective spin temperature. In particular, the effective spin temperature corresponding to resonance phonons of frequencies $\Omega\pm\omega_e$ is determined by the relation

$$\beta_{\text{eff}}^{\pm}(\omega)=\pm\frac{\omega_e}{\omega}\beta_s-\frac{\Omega\pm\omega_e-\omega}{\omega}\beta_d.$$

Below we shall see that this circumstance predetermines the appearance of the phonon avalanche in pulsed saturation of the EPR.

8. We proceed now to analyze the different limiting cases. First of all we assume that the spectral-diffusion length in the phonon spectrum is of the order of the width of the correlator $\tilde{f}(\omega)$. Then to the resonance phonons $\Omega\pm\omega_e$ we can assign a single temperature (β^+ and β^- respectively). The system of equations that describes the time evolution of $\tilde{\beta}_s$, β^+ and β^- can be obtained easily from Eqs. (27) in the standard way and has the form

$$\frac{d\beta_s}{dt}=-\frac{1}{T_{sp}^+(\theta)}\frac{\beta_0}{\beta^+}\left(\beta_s-\frac{\Omega+\omega_e}{\omega_e}\beta^+\right)-\frac{1}{T_{sp}^-(\theta)}\frac{\beta_0}{\beta^-}\left(\beta_s+\frac{\Omega-\omega_e}{\omega_e}\beta^-\right)$$
$$\frac{d\beta^\pm}{dt}=\pm\frac{1}{T_{ps}^\pm(\theta)}\frac{\omega_e}{\Omega\pm\omega_e}\left(\beta_s\mp\frac{\Omega\pm\omega_e}{\omega_e}\beta^\pm\right)\frac{\beta^\pm}{\beta_0}-\frac{\beta^\pm-\beta_0}{T_0}\frac{\beta^\pm}{\beta_0},\qquad (28)$$

where

$$\frac{1}{T_{sp}^\pm(\theta)}=\pi\Lambda_\pm^2(\theta)L^{+-}(\Omega\pm\omega_e),\qquad \frac{1}{T_{ps}^\pm(\theta)}=\frac{c_e}{\tilde{c}_p'\cdot 2\delta}\frac{1}{T_{sp}^\pm(\theta)},$$

and 2δ is the width corresponding to the correlator $\tilde{f}(\omega)$ (i.e., the width of the resonance phonon packet).

Hence it is easy to obtain that the stationary regime is characterized by the following distribution of temperatures:

$$\tilde{\beta}_s=\frac{\omega_s(\omega_s-\Omega)}{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2(1+\sigma'\omega_1^2/4\omega_e^2)}\beta_0,$$
$$\beta^+\approx\beta^-\approx\frac{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2}{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2(1+\sigma'\omega_1^2/4\omega_e^2)}\beta_0,\qquad (29)$$

where

$$\sigma'=\pi L^{+-}(\Omega)\frac{c_e}{c_p'2\delta}T_0.$$

These relations are analogous in many respects to the relations (18), if in the latter we put $\omega_1\gg\omega_d$. The essential point is that both groups of resonance phonons are heated equally.

We shall consider now the pulsed regime of EPR saturation, i.e., we shall assume that the spin system is excited by a sufficiently short UHF pulse, during which the spin system can be regarded as isolated from the lattice. Taking into account the fact that, in the limit of ultra-strong saturation ($\omega_1\gg\omega_d$) exchange of energy between the Zeeman subsystem and the DDR is difficult, we find that by the time the UHF pulse ends the state of the spin system will be characterized by the temperatures

$$\tilde{\beta}_s=\frac{\omega_s(\omega_s-\Omega)}{(\omega_s-\Omega)^2+\omega_1^2}\beta_0,\qquad \beta_d=\beta_0,\qquad (30)$$

while the deviations in the number of resonance phonons from the equilibrium values Z^+ and Z^- satisfy the equations

$$\dot{Z}^\pm+\frac{Z^\pm}{T_0}=\frac{1}{T_{ps}^\pm}\left[\frac{\omega_e(\Omega\pm\omega_e)\mp\omega_s(\omega_s-\Omega)}{\omega_e(\Omega\pm\omega_e)}\mp\frac{\omega_s(\omega_s-\Omega)}{\omega_e(\Omega\pm\omega_e)}Z^\pm\right].\qquad (31)$$

An analysis (which we do not pause to give) of these equations under conditions of a strong PB effect shows that for $\omega_s-\Omega<0$ the avalanche encompasses the resonance phonons of frequency $\Omega+\omega_e$, and for $\omega_s-\Omega>0$ encompasses those of frequency $\Omega-\omega_e$. The logarithmic increments of the avalanches are determined by the relations

$$\gamma^\pm=\frac{\dot{Z}^\pm}{Z^\pm}=\frac{1}{T_{ps}^\pm(\theta)}\frac{\omega_s|\omega_s-\Omega|}{\omega_e(\Omega\pm\omega_e)}.\qquad (32)$$

The appearance of a phonon avalanche is connected, as before, with negative values of the effective spin temperature. For $\omega_s-\Omega<0$ the effective spin temperature for the resonance phonons of frequency $\Omega+\omega_e$ becomes negative, while for $\omega_s-\Omega>0$ the spin temperature of the resonance phonons of frequency $\Omega-\omega_e$ is negative.

In connection with the results obtained, we must note the following. As has been observed, the appearance of a phonon avalanche in the Provotorov case is due to the DDR, which plays an active role in the saturation of the EPR. It is precisely the sharp increase of the DDR temperature that leads to the result that, depending on the sign of the detuning, the effective spin temperature for part of the resonance phonons takes a negative value. In particular, an avalanche develops in the phonons lying in the wing near to Ω. In strong oscillating fields, when $\omega_1 \gg \omega_d$, the Zeeman energy and the DDR defined in the ECF are thermodynamically no longer coupled with each other. The role of the DDR now reduces to that of a source of randomization within the spin system, while the DDR temperature is practically unchanged in the absence of rotational saturation at the frequency ω_e. Nevertheless, the effective spin temperature for the group of resonance phonons lying in the wing near to Ω remains, as before, negative, giving rise to the appearance of an avalanche.

Naturally, an avalanche-like increase in the number of resonance phonons also occurs in the limit of weak spectral diffusion in the system of phonons. The corresponding analysis is trivial and leads to the following values of the logarithmic increments of the avalanche:

$$\gamma^\pm(\omega) \approx \frac{1}{T_{ps}^{\pm'}} \frac{\omega_s|\omega_s-\Omega|}{\omega_e(\Omega\pm\omega_e)} \frac{\bar{f}(\Omega\pm\omega_e-\omega)}{\bar{f}(0)}, \qquad \frac{1}{T_{ps}^{\pm'}} = \frac{c_e}{c_p'}\bar{f}(0)\frac{1}{T_{sp}^\pm}. \tag{33}$$

In conclusion, we shall dwell briefly on the treatment of the stationary regime of saturation in the limit of weak spectral diffusion in a system of phonons with a weak PB effect ($\sigma^\pm = T_0/T_{ps}^{\pm'} \ll 1$). Neglecting, as we did in the Redfield case, the heating of the phonons in the calculation of the stationary spin temperature, we find

$$\tilde\beta_s = \frac{\omega_s(\omega_s-\Omega)}{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2}\beta_0, \qquad \tilde\beta_d=\beta_0,$$

after which, as a result of simple calculations, from the third equation of the system (27) we obtain

$$Z(\omega) \approx \sigma^+\frac{\bar f(\Omega+\omega_e-\omega)}{\bar f(0)}\left\{1-\frac{\omega_s}{\Omega+\omega_e}\frac{\omega_e(\omega_s-\Omega)}{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2}\right\} + \sigma^-\frac{\bar f(\Omega-\omega_e-\omega)}{\bar f(0)}\left\{1+\frac{\omega_s}{\Omega-\omega_e}\frac{\omega_e(\omega_s-\Omega)}{(\omega_s-\Omega)^2+{}^1/_2\omega_1^2}\right\}.$$

This means that the spectral distribution of the resonance phonons in both groups is practically symmetric about the central frequencies $\omega \approx \Omega \pm \omega_e$. In this case, heating occurs for both groups.

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1)Here, we have from the outset discarded the second term in $L_{kj}^\pm$, which leads later to terms oscillating with time, and have also taken the long-wave approximation ($\exp(\pm i\mathbf{k}\cdot\mathbf{r}_n) \approx 1$), which does not limit the generality.

2)We omit here the details of the calculations, which are analogous in many respects to those performed earlier [5]. We note that the high-temperature approximation is also taken with respect to the resonance phonons.

3)The addition of this term is discussed in more detail in [5].

4)For this the second equation of the system (12) must be multiplied by $-3\omega^2 d\omega/2\pi^2c^3\beta^2(\omega)$ and integrated over the spectrum of the resonance phonons. Here, in both equations, we must put $\beta(\omega) = \beta_0$.

5)To obtain this equation it is necessary, in the second equation of the system (12), to go over from $\beta(\omega)$ to $Z(\omega)$ and to use the expression (19) for β_s^*.

6)The terms with $\tilde S^z L^\pm$ which are of no interest have been discarded.

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