Radiofrequency superradiance and cw lasing by nuclear magnetic moments

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A semiclassical approach is used to investigate theoretically superradiance (SR) and continuous (cw) lasing by nuclear magnetic moments. We calculate the measurable lasing parameters—the SR delay time and duration, the voltage at the terminals of the coil, the threshold and final values of the nuclear polarization for pulsed and cw lasing in the case of homogeneous broadening of the NMR line and inexact tuning of the circuit to the NMR frequency. The results are in qualitative and quantitative agreement with experiment.

Resonator effects in systems of nuclear magnetic moments are diligently investigated of late. It has become possible, in particular, to amplify nuclear spin echo signals in magnets,[1] and to observe radiofrequency (rf) cw lasing[2] and superradiance (SR) pulses.[2–6] Note that the latter draws attention in view of the promise of developing sources of ultrashort and very intense coherent rf pulses.

It is known that cavity (resonator) effects are based on the reaction of the field magnetization induced in the cavity to its motion. The quantity indicative of the cavity is the radiation-damping time T_R (Refs. 7, 8). The first description of one of the cavity effects is contained in Ref. 9. Its authors investigated the influence of the rf circuit on the decrease of the free induction following deflection of the nuclear magnetization by pulses of an external alternating field. As shown in Ref. 1, this influence can be substantial also on spin-echo signals in magnets, either amplifying or attenuating them depending on the parameters of the specimen, of the exciting pulses, and of the cavity. Radiofrequency emission from preliminarily inverted nuclear moments was observed in Ref. 2. Different lasing types were obtained: SR was observed in the form of a single delayed high-intensity pulse, as well as cw lasing of considerably lower intensity. Similar phenomena were investigated also in Refs. 3–6, in which the output emission parameters were plotted as functions of the initial polarization of the nuclei.

Radiofrequency SR was theoretically investigated in Refs. 10–12. The calculations there, however, did not agree well enough with the experimental data. The purpose of the present paper is to describe various rf lasing processes and to calculate the output parameters of the radiation observed in experiment. We shall consider the case of homogeneous broadening of NMR and take account of inexact tuning of the rf circuit to the NMR frequency (SR in the case of inhomogeneous NMR broadening and exact tuning was investigated in Ref. 13).

1. BASIC EQUATIONS

To solve our problem we write the Bloch equations for the components of nuclear magnetization precessing in a constant field $\mathbf{H}_0$ parallel to the z axis, supplemented by an equation for the magnetic field $\mathbf{H}$ produced by the motion of the nuclear magnetization of a sample in a coil whose axis is the x axis:

$$\begin{aligned} \dot m_x &= \omega_n m_y - m_x/T_2, \\ \dot m_y &= -\omega_n m_x + \gamma_n m_z H - m_y/T_2, \\ \dot m_z &= -(m_z - m_e)/T_e - \gamma_n m_y H, \\ \ddot H &+ (\omega_c/Q)\dot H + \omega_c^2 H = -\eta_0 \ddot m_x, \end{aligned} \tag{1}$$

where $\omega_n = \gamma_n H_0$, γ_n is the gyromagnetic ratio of the nuclei, η_0 is the filling factor of the coil by the sample, Q the quality factor of the rf tank circuit, ω_c its natural frequency, T_2 the spin-spin relaxation time, and T_e the time to establish the effective value of the nuclear magnetization m_e. The term $-(m_z - m_e)/T_e$ describes the action of the relaxation and of the external pumping producing the negative nuclear polarization.

To solve these equations we assume hereafter that the period of oscillations of the magnetization ($\sim\omega_n^{-1}$) is much shorter than the relaxation times that enter in the magnetization + tank circuit system. It is known[1,2] that in this case it is possible to separate by the averaging method the slow variations of the amplitude and phases from the main high-frequency oscillations. We shall seek hence the solutions of the system (1) in the form

$$\begin{aligned} m_x(t) &= \tfrac{1}{2} A(t) \exp\{i[\omega_n t + \varphi(t)]\} + \text{c.c.}, \\ H(t) &= {}^1/_2 i h(t) \exp\{i[\omega_n t + \vartheta(t)]\} + \text{c.c.}, \end{aligned} \tag{2}$$

where $A(t)$ and $h(t)$ are slowly varying amplitudes, while $\varphi(t)$ and $\vartheta(t)$ are slowly varying phases.

We take into account one more simplifying circumstance: since the quantity $Q/\omega_c \equiv \tau_c$ is the smallest time scale in most experiments, we can neglect h in the equation for $\dot h$ compared with h/τ_c. After substituting (2) in (1) and averaging over the fast motion (spin precession with frequency ω_n) we obtain the following system of equations for the slow variables:

$$\dot A = -\frac{A}{T_2} + \frac{\gamma_n}{2} m_z h \cos\delta, \tag{3a}$$

$$\dot m_z = -\frac{m_z - m_e}{T_e} - \frac{\gamma_n}{2} A h \cos\delta, \tag{3b}$$

$$\dot\varphi = -\gamma_n \sin\delta \frac{m_z h}{2A}, \tag{3c}$$

where

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$$h=-\eta_0\omega_n\tau_c A\cos\delta, \qquad (3d)$$

$$\vartheta(t)=\varphi(t)-\delta, \qquad (3e)$$

$$\mathrm{tg}\,\delta=\frac{\omega_n^2-\omega_c^2}{\omega_n\omega_c}Q. \qquad (4)$$

The quantity δ takes it into account that if the circuit is not exactly tuned to the NMR frequency ($\omega_n\neq\omega_c$) the field H is shifted in phase relative to m_x by an angle not equal to $\pi/2$. The detuning should be then much smaller than the pass band of the oscillating circuit, i.e., $|\omega_n-\omega_c|\ll\omega_c/Q$. In this case expression (4) can be approximately replaced by

$$\mathrm{tg}\,\delta\approx\frac{\omega_n-\omega_c}{\omega_c}2Q. \qquad (5)$$

On the basis of these equations we shall investigate henceforth in Sec. 2 the case of nonstationary evolution of nuclear magnetization under the action of an rf circuit in the absence of a mechanism that establishes m_e. In Sec. 3 we investigate stationary rf lasing with the pump constantly on. In Sec. 4 the theoretical results are quantitatively compared with the experimental data.

2. NONSTATIONARY rf EMISSION

a. Assume that the pump produces an initial negative nuclear magnetization and is then switched off. When certain threshold conditions are satisfied, a single delayed rf emission pulse is observed.[2] Consider first the case when the nuclear spins have equal spin-spin and spin-lattice relaxation times, i.e., $T_2=T_1$. This situation is known[8] to be realized in liquids, so that in this case we are considering the feasibility of SR in liquid samples. A high polarization of nuclei can be achieved in liquids with the aid of the Overhauser effect,[8] optical excitation,[7] or chemical polarization of the nuclei.[14] Note that the influence of a cavity on the decrease of the free induction of protons in water was observed way back in Ref. 9, while in Refs. 15 and 16 lasing was observed from protons of an aqueous solution of peroxylamine disulfonate. The proton maser operated both in a field on the order of 3000 Oe (Ref. 15) and in the earth's magnetic field, where it was used as a magnetometer.[16] We denote $T_2=T_1=T$,[1)] and take it also into account that when the "pump" is turned off we have $m_e=m_{\rm eq}$, where $m_{\rm eq}$ is the equilibrium value of the nuclear magnetization (in this case, naturally, $T_e=T$). Since we have at the initial instant of time $|m_z(0)|\gg m_{\rm eq}$, we can neglect $m_{\rm eq}$ in the equation for m_z and assume that m_z decreases to zero.

We seek the solution of the system (3) in the form

$$m_z=-|m_0|\exp(-t/T)\cos\psi,$$

$$A=|m_0|\exp(-t/T)\sin\psi,$$

where $|m_0|$ is the modulus of the initial inverted magnetization, and ψ is the angle between **m** and the direction of the z axis. Note that the possibility of representing the solutions in this form means that the presence of a magnetization-vector rotation due to the influence of the rf circuit. It is easily seen that the quantity ψ satisfies the equation

$$\frac{d\psi}{dt}=\frac{\cos\delta}{T_R}\exp\left(-\frac{t}{T}\right)\sin\psi,$$

whose solution is

$$\sin\psi=\mathrm{ch}^{-1}[(\tau-\tau_D)/T_R],$$

where

$$\tau=T[1-\exp(-t/T)],$$

$$T_R=(^1/_2\eta_0\gamma_nQ|m_0|\cos^2\delta)^{-1} \qquad (6)$$

is the time of radiative damping of the resonant circuit with allowance for $\omega_n\neq\omega_c$, since we have from (5)

$$\cos\delta=\frac{\omega_c/2Q}{[\omega_c^2/4Q^2+(\omega_n-\omega_c)^2]^{1/2}}.$$

The values of the slow variables are finally given by the expressions

$$A=|m_0|e^{-t/T}\,\mathrm{ch}^{-1}\left(\frac{\tau-\tau_D}{T_R}\right), \qquad (7a)$$

$$m_z=|m_0|e^{-t/T}\,\mathrm{th}\left(\frac{\tau-\tau_D}{T_R}\right), \qquad (7b)$$

$$h=-\frac{2}{\gamma_nT_R\cos\delta}e^{-t/T}\,\mathrm{ch}^{-1}\left(\frac{\tau-\tau_D}{T_R}\right), \qquad (7c)$$

$$\dot\varphi=\frac{\mathrm{tg}\,\delta}{T_R}e^{-t/T}\,\mathrm{th}\left(\frac{\tau-\tau_D}{T_R}\right), \qquad (7d)$$

which describe rapidly developing rf lasing with a peaked maximum in time. The maximum values of A and h are reached at $\tau=\tau_D$, i.e., at

$$t=T_D=\ln[T/(T-T_D)],$$

where $t=0$ corresponds to the instant when the lasing threshold is reached, T_D is called the delay time, and the value of τ_D can be estimated from the initial condition

$$A(0)=|m_0|\mathrm{ch}^{-1}(\tau_D/T_R),$$

whence we have, with allowance for the relation $\tau_D\gg T_R$ that is usually met in experiment,

$$\tau_D\approx T_R\ln(2|m_0|/A(0)). \qquad (8)$$

We assume that the initial nonzero value $A(0)=(A_n^2)^{1/2}$ of the transverse magnetization is the result of fluctuations, in this case due to Nyquist noise in the resonant circuit. To calculate $(A_n^2)^{1/2}$ we use Eq. (3a) in which we replace h by the "priming" magnetic field in the coil

$$h_n=\frac{N}{l}\frac{(U_n^2)^{1/2}}{R},$$

where

$$(U_n^2)^{1/2}=(2/\pi)k_B\Theta_nR\Delta\omega$$

is the rms voltage due to the Nyquist noise, Θ_n the noise temperature, R the active resistance of the circuit, $\Delta\omega$ the NMR line width, and l its width. Solving the obtained equation in the stationary case and recognizing the noise acts only during the initial instant of time, when $m_z\approx-|m_0|$, we get

$$(A_n^2)^{1/2}=\frac{\gamma_n}{2}T|m_0|\left(\frac{k_B\Theta_n\Delta\omega Q}{2\pi^2r^2l\mu_0\omega_n}\right)^{1/2}, \qquad (9)$$

where $\mu_0=4\pi\cdot10^{-7}$ H/m is the magnetic constant and r is the coil radius. Substituting in (8) the known relation $m_0=cP_i$, where $c=\hbar\gamma_nn/2\mu_0$, n is the density of the nuclear spins, and P_i is their initial polarization, as well as the explicit expression for T_R, we obtain the dependence of τ_D on $|P_i|$ and on the detuning (via $\cos^2\delta$) in the form

$$\tau_D \approx \frac{2}{\eta_0\gamma_n c|P_i|Q\cos^2\delta}\ln\frac{c|P_i|}{(A_n^2)^{1/2}}. \qquad (10)$$

Analysis of the solutions (7) by using the reasoning of Ref. 17 shows that lasing is possible only if the following inequality holds

$$\tau_D<T.$$

The physical reason is quite clear. Superradiance occurs only if the deflection time τ_D of the magnetization from a direction opposite to the z axis to a direction perpendicular to z is shorter than the time T of relaxation of the magnetization to equilibrium. From the condition $\tau_D = T$ and expression (10) we obtain for the threshold value of $|P_i|$

$$|P_i|_{\text{thr}} \approx \frac{4\mu_0}{\hbar\eta_0\gamma_n^2 nTQ\cos^2\delta}\ln\frac{c|P_i|}{(A_n^2)^{1/2}}. \qquad (11)$$

As expected, τ_D and $|P_i|_{\text{thr}}$ increase with increase of the detuning.

It is evident from (2) and (3e) that (7d) is the lasing-frequency shift from ω_n. The absolute value of the SR frequency shift also increases with the detuning (it is zero for exact tuning), but at $t = T_D$, i.e., at the SR maximum, it is zero for any detuning.

Since the SR is revealed by the voltage at the terminals of a coil, we calculate this voltage by equating the electric and magnetic energies of the circuit. Then,

$$|U|=\left(\frac{\mu_0 V_{\text{coil}}}{C}\right)^{1/2} h,$$

where V_{coil} is the volume of the coil and the C the capacitance of the circuit. Substituting here the value of h from (7c) and using the relation

$$V_{\text{coil}}=\mu_0 C\omega_c^2 S^2 N^2,$$

where S is the coil cross-section area, we obtain for the envelope of the SR pulse

$$|U_{\text{SR}}^{\text{env}}|=\frac{2\mu_0 SN\omega_c}{\gamma_n T_R\cos\delta}e^{-t/T}\,\text{ch}^{-1}\left(\frac{\tau-\tau_D}{T_R}\right),$$

which yields readily the maximum voltage in the form

$$|U_{\text{SR}}^{\max}| =\eta_0\mu_0\omega_c SN|m_0|Q\cos\delta(1-\tau_D/T). \qquad (12)$$

To calculate the duration Δt_{SR} of the SR pulse, we recognize that in experiment we usually have $\Delta t \ll T$ (Refs. 2–6), and starting from the condition

$$U|_{t=T_D+\Delta t}={}^1\!/_2 U|_{t=T_D},$$

we obtain

$$\Delta t_{\text{SR}} \approx \frac{T_R\ln(2+3^{1/2})}{1-\tau_D/T}. \qquad (13)$$

Finally, using Eqs. (3) we can write the following energy balance equation

$$I_{\text{SR}} = -\frac{d}{dt}(W_{\text{cir}} + W_Z),$$

where

$$W_{\text{coil}} =\mu_0 V_{\text{coil}} h^2/2$$

is the energy magnetic field in the rf circuit;

$$W_Z=-\mu_0 m_z H_0 V_{\text{sp}}$$

is the Zeeman energy of the nuclear spins (V_{sp} is the specimen volume):

$$I_{\text{SR}} =\frac{\mu_0 V_{\text{coil}} h^2}{2\tau_c}+\frac{W_Z}{T} \qquad (14)$$

is the SR intensity, with the second term representing the loss due to the decrease of the length of the vector **m** via spin-lattice relaxation. Substituting (7b,7c) in (14) we get

$$I_{\text{SR}} =\frac{\eta_0\omega_c Q\hbar^2\gamma_n^2 P_i^2 N_s n\cos^2\delta}{8\mu_0}e^{-2t/T}\,\text{ch}^{-2}\left(\frac{\tau-\tau_D}{T_R}\right) -\frac{\mu_0\omega_n V_{\text{sp}}}{\gamma_n T}|m_0|e^{-t/T}\,\text{th}\left(\frac{\tau-\tau_D}{T_R}\right). \qquad (15)$$

where N_s is the number of spins.

b. We consider now the case $T_2 \ll T_1$, which is the situation in a solid, where a negative nuclear polarization is achieved by the effect-solide method[8] or by dynamic cooling.[18] The spin-lattice relaxation in the SR formation can then be neglected. We consider first the initial state of the onset of SR, when $m_z \approx -|m_0|$. In this case the equation (3a) for A takes the form

$$A=(1/T_R+1/T_2)A,$$

from which follows the known[2,16] condition for the growth of A and, correspondingly, h:

$$T_R<T_2. \qquad (16)$$

When the inequality (16) is satisfied, the system (3) has the following solution:

$$A=|m_0|\left(1-\frac{T_R}{T_2}\right)\text{ch}^{-1}\left[(t-T_D)\left(\frac{1}{T_R}-\frac{1}{T_2}\right)\right], \qquad (17a)$$

$$m_z=-|m_0|\frac{T_R}{T_2}+|m_0|\left(1-\frac{T_R}{T_2}\right)\text{th}\left[(t-T_D)\left(\frac{1}{T_R}-\frac{1}{T_2}\right)\right], \qquad (17b)$$

$$h=-\frac{2}{\gamma_n T_R\cos\delta}\left(1-\frac{T_R}{T_2}\right)\text{ch}^{-1}\left[(t-T_D)\left(\frac{1}{T_R}-\frac{1}{T_2}\right)\right], \qquad (17c)$$

$$\varphi=\frac{\text{tg}\,\delta}{T_R}\left\{-\frac{T_R}{T_2}+\left(1-\frac{T_R}{T_2}\right)\text{th}\left[(t-T_D)\left(\frac{1}{T_R}-\frac{1}{T_2}\right)\right]\right\}, \qquad (17d)$$

and the values of δ, T_R, and $(A_n^2)^{1/2}$ is determined as before by expressions (4), (5), (7), and (9), except that (9) contains T_2 in place of T, in which case $(A_n^2)^{1/2}$ coincides with the corresponding expression of Ref. 2. From (16) and (6) we obtain for the threshold initial value of the nuclear polarization

$$|P_i|_{\text{thr}} =4\mu_0/\hbar\eta_0\gamma_n^2 nT_2 Q\cos^2\delta. \qquad (18)$$

Using (17b), we can write for the final value of the nuclear polarization $P_f = P|_{t\to\infty}$

$$P_f=|P_i|(1-2T_R/T_2).$$

This expression yields a known result,[6,12] the linear dependence of P_f on $|P_i|$, which is observed in experiment,[5,6] with the polarization reversed at

$$T_R < T_2/2,$$

whence

$$|P_i|_{\text{r.p.}} = 2|P_i|_{\text{thr}} .$$

We consider now the lasing frequency shift due to detuning of the rf circuit from the NMR frequency. It follows from (17d) that in the general case the SR frequency is given by

$$\omega_{\text{SR}} = \omega_n + 2(\omega_n - \omega_c)\tau_c\left\{-\frac{1}{T_2} + \left(\frac{1}{T_R} - \frac{1}{T_2}\right)\right.$$
$$\left.\times \text{th}\left[(t-T_D)\left(\frac{1}{T_R} - \frac{1}{T_2}\right)\right]\right\}, \quad (19)$$

from which we see that at various instants of time the SR frequency shift has different values and even signs. Thus, at the initial lasing instant

$$\omega_{\text{SR}}|_{t=0} \approx \omega_n + 2(\omega_n - \omega_c)\tau_c/T_R,$$

at the maximum of the SR

$$\omega_{\text{SR}}|_{t=T_D} \approx \omega_n - 2(\omega_n - \omega_c)\tau_c/T_2,$$

and at the termination of the SR pulse it depends on whether the reversal condition is satisfied, viz.

$$\omega_{\text{SR}}|_{t\to\infty} \approx \omega_n - |1/T_R - 2/T_2| 2(\omega_n - \omega_c)\tau_c$$

in the absence of reversal and

$$\omega_{\text{SR}}|_{t\to\infty} \approx \omega_n + |1/T_R - 2/T_2| 2(\omega_n - \omega_c)\tau_c$$

in the presence of reversal. Note, however, that since we have assumed the detuning to be small, the frequency shifts obtained are small. Nonetheless, our allowance for the detuning accounts qualitatively for the small discrepancies between the experiment and the computer results obtained in Ref. 2 for exact tuning. Namely, allowance for the detuning explains the shift of the SR maximum to the right along the time axis, and also the decrease of the generated voltage on the SR wings.

The expressions for T_D, I_{SR}, $U_{\text{SR}}^{\text{env}}$, and Δt are determined here as in case **a**.[2)] Since the results obtained in case **b** will be compared with the experimental results of Refs. 5 and 6, where these quantities are represented as functions of P_i, it follows that by introducing the notation

$$T_R = 1/k|P_i|\cos^2\delta,$$

where $k = \eta_0\gamma_n Qc/2$, we express these quantities as functions of $|P_i|$, and also of $\cos^2\delta$, we obtain the dependence on the detuning:

$$T_D = \frac{1}{k|P_i|\cos^2\delta - T_2^{-1}} \ln\frac{c(k|P_i|\cos^2\delta - T_2^{-1})}{k\cos^2\delta\,(A_n^2)^{1/2}}, \quad (20)$$

$$I_{\text{SR}} = \frac{\eta_0\omega_c Q\hbar^2\gamma_n^2 P_i^2 N_s n\cos^2\delta}{8\mu_0}[1-(T_2k|P_i|\cos^2\delta)^{-1}]^2$$
$$\times \text{ch}^{-2}[(t-T_D)(k|P_i|\cos^2\delta - T_2^{-1})], \quad (21)$$

$$|U_{\text{SR}}^{\text{env}}| = \frac{2\mu_0 SN\omega_c}{\gamma_n\cos\delta}(k|P_i|\cos^2\delta - T_2^{-1})$$
$$\times \text{ch}^{-1}[(t-T_D)(k|P_i|\cos^2\delta - T_2^{-1})], \quad (22)$$

$$\Delta t_{\text{SR}} \approx \frac{T_R\ln(2+3^{1/2})}{1-T_R/T_2} = \frac{\ln(2+3^{1/2})}{k|P_i|\cos^2\delta - T_2^{-1}}. \quad (23)$$

As should have been expected, with increase of the detuning (with decrease of $\cos^2\delta$), the values of T_D, Δt_{SR} and of the lasing frequency increase while U_{SR} and I_{SR} decrease. Comparing $U_{\text{SR}}^{\max}$ and Δt_{SR} in cases **a** and **b**, we obtain

$$\frac{(U_{\text{SR}}^{\max})^{\text{b}}}{(U_{\text{SR}}^{\max})^{\text{a}}} = \frac{1-T_R/T_2}{1-\tau_D/T}, \quad \frac{\Delta t_{\text{SR}}^{\text{b}}}{\Delta t_{\text{SR}}^{\text{a}}} = \frac{1-\tau_D/T}{1-T_R/T_2},$$

from which it can be seen when account is taken of the inequality $T_R \ll \tau_D$, usually satisfied in the experiment, that in case **b**, for identical values of $|P_i|$ the generated voltage of the SR pulse will be larger, and its duration shorter (this is physically understandable, inasmuch as energy is transferred irreversibly to the lattice in case **a**). The theoretical and experimental investigations of case **a** are, however, of interest, since observation of intense SR signals in liquids yields valuable information on its spin dynamics.

Let us ascertain the conditions under which the described RF lasing is coherent. Obviously, this occurs when the motion of the emitting nuclear moments is phase-coherent, i.e., according to Ref. 19, the nutation period of the nuclear spins in the effective field in coordinate frame rotating at a frequency ω_c will be shorter than the phase memory of the spins:

$$\tau_n = \{(\gamma_n h_{max}/2)^2 + (\omega_{\text{SR}}|_{t=T_D} - \omega_n)^2\}^{-1/2} < T_2.$$

We shall therefore characterize the SR coherence by the quantity

$$\zeta = T_2/\tau_n ,$$

which reduces in case **b**, for exact tuning, to

$$\zeta \sim T_2/2T_R.$$

The larger ζ, the more phase-coherent is the motion of the spins and their radiation. The last expression agrees also with the fact [which follows from Eqs. (20)–(23)] that the quadratic dependence of the intensity and the inverse proportionality of the SR pulse duration on the number of active particle, both of which are indicative of the SR phenomenon, take place if $T_R \propto N_s^{-1}$ is substantially lower than the threshold value $T_R = T_2$.

3. STATIONARY rf LASING

Consider now the case when the nuclear spins of a solid specimen ($T_2 \ll T_1$) are continuously pumped in the course of lasing, for example when they are in contact with an electron dipole-dipole reservoir (EDDR) whose level inversion is supported by nonresonant EPR saturation. To investigate this lasing we turn to the system of equations (1), where $T_e \neq \infty$. The time T_e coincides in this case with the time to produce nonequilibrium nuclear polarization by the dynamic cooling method. When the condition is satisfied for saturation of an EPR line by a microwave field with detuning Δ from the EPR line center, and in the absence of extraneous nuclear relaxation (not connected with the EDDR), we have

$$T_e^{-1}=\tau_f^{-1}=T_{Id}^{-1}(\alpha'\omega_d^2+\Delta^2)\Big/\left[\left(1+\frac{C_I}{C_d}\frac{T_{dL}}{T_{Id}}\right)\alpha'\omega_d^2+\Delta^2\right],$$

where C_I and C_d are, respectively, the specific heats of the nuclear spins and the EDDR participating in the lasing, respectively; ω_d is the average EDDR quantum; $\alpha' = T_{SL}/T_{dL}$; T_{SL} and T_{dL} are the times of the electron spin and dipole relaxations to the lattice; T_{Id} is the relaxation time of the nuclei to the EDDR.

We assume as before that τ_c is the smallest time scale, therefore

$$h=-2A/\gamma_n T_{Re}|m_e|\cos\delta,$$

where $T_{Re}^{-1} = 1/2\eta_0\gamma_n|m_e|Q\cos^2\delta$. The equations for A and m_z can then be rewritten as

$$\dot{A}=\left(\frac{1}{T_{Re}}-\frac{1}{T_2}\right)A-\frac{A}{T_{Re}}\frac{m_z+|m_e|}{|m_e|},$$

$$\dot{m}_z=-\frac{m_z+|m_e|}{T_e}+\frac{A^2}{T_{Re}|m_e|}. \qquad (24)$$

The system (24) has two stationary solutions. The nonradiative stationary solution of this system ($A=0$, $m_z = -|m_e|$) becomes unstable at $T_{Re} < T_2$. The radiative solution ($A^2 = m_e^2(1 = -T_{Re}/T_2)T_{Re}/T_e$, $m_z = -|m_e|T_{Re}/T_2$), is valid only if $T_{Re} < T_2$ and is stable if $1/T_e > 1/T_{Re} - 1/T_2$. Continuing the analogy of the motion of a beforehand-inverted nuclear magnetization with the descent of a pendulum from its upper unstable equilibrium state,[17] it can be stated that the stationary lasing regime corresponds to the fall of a pendulum in the presence of a "spring" that maintains the pendulum near the upper equilibrium state. In the situations of Refs. 2–6 the role of the spring is performed by thermal contact between the nuclei and an EDDR which is a continuous-cooling state. Let us consider the establishment of a stationary lasing regime.

Assume that the following case is realized

$$\frac{1}{T_{Re}}>\frac{1}{T_2},\qquad \frac{1}{T_{Re}}-\frac{1}{T_2}<\frac{1}{T_e}.$$

We stipulate also that

$$\frac{1}{T_{Re}}-\frac{1}{T_2}\ll\frac{1}{T_e}. \qquad (24a)$$

The evolution of m_z is then faster than the evolution A, and accordance to the subordination principle[20] we can put $\dot{m}_z = 0$. [Note that if we make in (24) a change of variables $U = A|m_e|$ and $S = (m_z + |m_e|)/|m_e|$, and use the dimensionless time $t' = t/T_R$, the system (24) has the same form as Eqs. (7.1.1–2) in Ref. 20.] To satisfy inequality (24a) it is necessary that the system be close to the lasing threshold, i.e., $|m_e|\approx|m_e|^{\text{thr}}$. As will be shown below, in cw lasing the value $|m_z|\approx -|m_e|^{\text{thr}}$ remains constant in this case.

For $\dot{m}_z = 0$, the evolution of A is described by the equation

$$\dot{A}=-\alpha A-\beta A^3, \qquad (25)$$

where

$$\alpha=-\left(\frac{1}{T_{Re}}-\frac{1}{T_2}\right),\qquad \beta=\frac{T_e}{T_{Re}^2 m_e^2}.$$

Equation (25) has the solution

$$A=\pm\left(-\frac{\alpha}{\beta}\right)^{1/2}\left(1-\frac{A^2(0)+\alpha/\beta}{A^2(0)}e^{2\alpha t}\right)^{-1/2}, \qquad (26)$$

which describes at $\alpha > 0$ a decrease of A to zero, and at $\alpha < 0$ (i.e., $T_{Re} < T_2$) a change of A to the value

$$A=\pm\left(-\frac{\alpha}{\beta}\right)^{1/2}=\pm|m_e|\left(1-\frac{T_{Re}}{T_2}\right)^{1/2}\left(\frac{T_{Re}}{T_e}\right)^{1/2}. \qquad (27)$$

This value of A produces at the terminals of the coil a stationary voltage

$$U_{\text{Mr}}=\frac{2\mu_0 SN\omega_c}{\gamma_n(T_{Re}T_2)^{1/2}\cos\delta}\left(1-\frac{T_{Re}}{T_2}\right)^{1/2}.$$

Since $T_e \gg T_R$, the intensity of the stationary lasing will be much lower than in the SR pulse, where in case **b** we have

$$U_{\text{SR}}^{\max}=\frac{2\mu_0 SN\omega_c}{\gamma_n T_R\cos\delta}\left(1-\frac{T_{Re}}{T_2}\right).$$

Calculating in the case of stationary lasing the quantity

$$\frac{|\gamma_n h_{max}|}{\Delta\omega}=\frac{T_2}{(T_{Re}T_e)^{1/2}}\left(1-\frac{T_{Re}}{T_2}\right)^{1/2},$$

where $\Delta\omega$ is the NMR width, and recognizing that $T_{Re} < T_2$, and $T_e \gg T_2$, we find that the generated rf field cannot govern all the spins over the NMR linewidth, i.e., the motion of the spins is not coherent. This lasing should be interrupted when the condition $T_{Re} < T_2$ is violated but is in force again when this condition is restored (just as the spring that maintains a pendulum near its upper equilibrium state is always in action). The features listed indicate that the described lasing is incoherent maser generation (MG). The value of m_z that sets in as a result of MG is equal to

$$m_z=-|m_e|T_{Re}/T_2. \qquad (28)$$

As was assumed above, $|m_e|\approx|m_e|^{\text{thr}}$, and then $T_{Re}\approx T_2$ and $m_z \approx -|m_e|^{\text{thr}}$, i.e., in the stationary MG system is near threshold all the time, while the stationary value of the nuclear polarization

$$P_f^{\text{MG}}=-2/c\eta_0\gamma_n T_2 Q\cos^2\delta$$

is independent of $|P_i|$.

4. QUANTITATIVE COMPARISON WITH THE EXPERIMENTAL DATA

We carry out a quantitative comparison with the experimental results of Refs. 2, 5, and 6, where the case $T_2 \ll T_1$ was realized. (Note that our calculations cannot be compared with the experiments of Refs. 3 and 4, in which τ_c is not the very shortest time.) We consider first the results of Ref. 2, where a single delayed pulse of high intensity was observed after producing an above-threshold negative nuclear polarization and tuning the rf circuit to the NMR frequency, followed after some time by stationary rf generation of much lower intensity. Substituting the experimental data[2]

$T_R = 2.6\cdot10^{-5}$ s, $T_2 = 3\cdot10^{-5}$ s, $\omega_n = 2\pi\cdot12.2\cdot10^6$ rad/s,

$|m_0| = 2.8$ A/m, $\eta_0 = 0.575$, $Q = 60$,

$V_{\text{cat}} = 1.74\cdot10^{-6}\ \text{m}^3$,

$(A_n^2)^{1/2}\approx10^{-7}$ A/m,

we obtain from Eq. (20) $T_D \approx 3\cdot 10^{-3}$ s, which is close to the measured $2\cdot 10^{-3}$ s, while from Eq. (23) we obtain $\Delta t_{SR} = 0.26\cdot 10^{-3}$ s, which agrees with the measured value $0.25\cdot 10^{-3}$ s. The data in Ref. 2 are insufficient to calculate U_{SR}^{max}, and it can only be stated that the experimental value $U_{SR}^{max} = 2.5$ V agrees with that calculated from Eq. (22) at a circuit capacitance 55 pF. (The values of U_{MG} below, calculated for $C = 55$ pF, agree with experiment.) Calculations of the final value of the longitudinal nuclear magnetization in SR yield

$$m_z(\infty) = |m_0|(1 - 2T_R/T_2) = -2,$$

which agrees with the measured $m = -2.05$ (see Fig. 15 of Ref. 2).

We substitute now in the very same equations the experimental data of Refs. 5 and 6:

$$\omega_n = 2\pi\cdot 54\cdot 10^6 \text{ rad/s}, \quad N = 3, \; l = 8\cdot 10^{-3} \text{ m},$$
$$S = 1.13\cdot 10^{-4} \text{ m}^2, \quad |P_i|_{\text{r.p.}} = 0.32,$$

NMR line width $\Delta\omega = 80$ kHz, which corresponds to $T_2 \approx 6\cdot 10^{-6}$ s for a Gaussian NMR line. The duration of the SR pulse, calculated using these data in the equation

$$\Delta t_{SR} \approx \frac{T_2 \ln(2 + 3^{1/2})}{2|P_i|/|P_i|_{\text{r.p.}}^{-1}}$$

(with $|P_i| = 0.55$ and $\Delta t_{SR} \approx 3\cdot 10^{-6}$ s) turns out to be much lower than the experimental ($\approx 80\cdot 10^{-6}$ s). Note, however, that we have calculated the SR duration at constant detuning, whereas in the experiments[5,6] a linear scan of the constant magnetic field was used. It seems to us therefore that the measured SR-pulse duration reflects also the rate of this scan and the variable detuning, and there should be no agreement here with experiment.

Calculating the maximum voltage at the terminals of the coil under SR conditions

$$U_{SR}^{max} = \frac{4\mu_0 SN\omega_c}{T_2\gamma_n |P_i|_{\text{r.p.}} \cos\delta}\left(|P_i| - \frac{|P_i|_{\text{r.p.}}}{2}\right),$$

at $|P_i|_{\text{r.p.}}$, we obtain[3)] for U_{SR}^{max} in volts

$$U_{SR}^{max} \approx 140\left(\frac{\omega_c^2}{4Q^2} + (\omega_n - \omega_c)^2\right)^{1/2} \Big/ \frac{\omega_c}{2Q},$$

which yields, with allowance for detuning, a value of the same order as observed in experiment at the point of value reversal

$$U_{SR}^{max}(|P_i|_{\text{r.p.}}) = 900 \text{ V}.$$

We calculate now the rf lasing voltage in the stationary regime with the external pump turned on under the experimental conditions of Ref. 2. We obtain first the value of T_e. To this we substitute in the ratio of the voltages $U_{MG} = 0.06$ V and $U_{SR}^{max} = 2.5$ V,

$$\frac{U_{MG}}{U_{SR}^{max}} = \frac{|m_e|}{m_0}\frac{(1 - T_{Re}/T_2)^{1/2}}{1 - T_{Re}/T_2}\left(\frac{T_{Re}}{T_2}\right)^{1/2},$$

the experimental parameters cited above,[2] as well as our estimates $|m_e| \approx |m_e|^{\text{thr}} \approx 2.43$ A/m. We substitute the resultant values $T_{Re} = 2.99\cdot 10^{-5}$ s and $T_e = 4.47\cdot 10^{-3}$ s together with the capacitance $C = 55$ pF estimated above for the rf circuit in the expression for U_{MG}:

$$U_{MG} = \eta_0 Q|m_e|\left(\frac{\mu_0 V_{\text{cat}}}{C}\right)^{1/2}\left(\frac{T_{Re}}{T_e}\right)^{1/2}\left(1 - \frac{T_{Re}}{T_2}\right)^{1/2} = 0{,}05 \text{ V},$$

which agrees with the measured value $U_{MG} = 0.06$ V.

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[1)] Note that numerical calculations[7] show that the small difference between T_2 and T_1 (within the order of magnitude) does not influence the qualitative results and alters little the quantitative ones connected with the radiative damping of the resonant circuit.

[2)] Note that the magnetization components and the delay time were calculated in Ref. 12 for the case $T_2 \ll T_1$. They agree with the results of the present paper for case **b**, but the nature of $A(0)$ was not explained in Ref. 12. The reason for the delay time therefore remained unclear. In addition, the values of U_{SR} and Δt were not calculated in Ref. 12.

[3)] Since $|P_i|_{\text{r.p.}} \propto 1/\cos^2\delta$, we have in fact $U_{SR}^{max} \propto \cos\delta$, i.e., it decreases with increase of the detuning.

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