## Feasibility of stationary cw maser effect on thermal and microwave excitation of paramagnetic crystals

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The feasibility of a new type of cw maser scheme with potentially good characteristics is discussed for the particular case of an yttrium ethyl sulfate crystal containing the impurity ions <sup>143</sup>Nd<sup>3+</sup>, <sup>172</sup>Tb<sup>3+</sup>, and <sup>168</sup>Er<sup>3+</sup>. This type of scheme differs from the classical three-level scheme in that the quantum amplification occurs under conditions of partial inversion of the populations among the sublevels of working ions with an effective electron spin  $\frac{1}{2}$  and a nonzero nuclear spin and is sustained mainly by the energy of thermal phonons of the crystal lattice, which is cooled to liquidhelium temperatures. Here the exciting microwave radiation is only the means of producing the population inversion.

1. Spin systems in paramagnetic crystals played a prominant conceptual role in the early development of quantum electronics. The feasibility of a three-level maser scheme was first demonstrated for systems of this type and later carried over into the optical region (the ruby laser).<sup>1</sup> Subsequently, electric-discharge, chemical, and gasdynamic lasers in the optical region utilized methods of obtaining population inversions without the use of optical pumping. These methods employed new, rather general physical ideas and principles, including: 1) the use of many-component active media containing particles of different kinds, each fulfilling a different function in the creation of the population inversion in accordance with its spectroscopic and relaxational properties; 2) the use of resonant energy exchange between particles of certain kinds; 3) partial population inversion; 4) thermal pumping. An example of the simultaneous embodiment of these principles is the thermal gasdynamic IR laser working on vibrational-rotational transitions of diatomic molecules with a partial population inversion.<sup>2</sup>

Meanwhile, for spin systems in paramagnetic crystals there had not been as much progress by the end of the 1970s in methods of obtaining inversion in the microwave region, although several extremely important ideas had been expressed.<sup>3,4</sup> The methods that had been implemented in practice all somehow or other involved microwave or optical pumping.<sup>4–6</sup> Principles 1) and 2) were not widely used,<sup>6</sup> although one way of implementing them was first employed in the Scovil maser back in 1957.<sup>1</sup> The partial inversion achieved in experiments on the slightly off-resonance saturation of the ESR line was too small for practical use.<sup>4</sup>

A certain gap had thus developed in the conceptual principles used to obtain inversion in the optical and microwave regions, prompting me to study the possibility of extending the new ideas that have proved successful in lasers (principally in the gasdynamic laser) to spin systems in paramagnetic crystals. This problem, which is in some sense opposite to that faced in the early development of quantum electronics, is of obvious theoretical and practical interest.

2. As a first step, I proposed theoretically<sup>7</sup> and then, together with Khasanov and Imamutdinov,<sup>8,9</sup> successfully implemented a novel type of pulsed paramagnetic maser in which the quantum amplification is sustained directly by the

energy of thermal phonons in the crystal. The working model of the maser was a crystal of lanthanum-magnesium double nitrate La2Mg3(NO3)24H2O(LaMN) containing impurities of  $\sim 0.1$  at.% <sup>59</sup>Co<sup>2+</sup> (working ion A) and  $\sim 3$  at.%  $Ce^{3+}$  (ion B). The crystal was cooled to  $T_L = 1.7$  K and placed in a static magnetic field  $H_0$  directed at a small angle  $\theta_0$  to the crystalline axis  $C_3 \| Z$ . The values  $H_0 \approx 1.8$  kOe and  $\theta_0 \approx 1^\circ$  were chosen such that the Zeeman splitting  $\delta_A$  $\equiv \delta_A(\mathbf{H}_0)$  of the electronic spins  $S_A = \frac{1}{2}$  of the cobalt fell in the frequency range  $\lambda \approx 3$  cm of the ESR spectrometer, while the small Zeeman splitting  $\delta_{B}(\mathbf{H}_{0})$  of the cerium spins  $S_B = \frac{1}{2}$ , characterized by a highly anisotropic g factor  $(g_{\parallel}^{B} = 0.0235, g_{\perp}^{B} = 1.8264)$ , was equal to A /2, where A is the hyperfine-structure constant of the working ions in the field  $H_0$ . In this case a strong cross relaxational coupling arises, and an effective thermal contact is established between the nuclear spins  $I = \frac{7}{2}$  of the Co<sup>59</sup> and the elec-

between the nuclear spins  $I = \frac{7}{2}$  of the Co<sup>59</sup> and the electronic spins of the cerium. The latter are then artificially cooled by adiabatic demagnetization in half-sinusoidal magnetic-field pulses  $H_1^p(t)$  oriented perpendicular to the  $C_3$  axis. After a field pulse has ended, the coupling of the nuclear spins of the Co<sup>59</sup> and the cooled (polarized) spins of the Ce<sup>3+</sup> causes a rapid thermal mixing, establishing a temperature  $T_I$  that is substantially lower than the electronic spin temperatures  $T_S^A \approx T_L$  of the A ions. As a result, the system of spin sublevels in the cobalt acquires a short-term nonequilibrium population distribution of the form<sup>7</sup>

$$n(M,m) = \frac{n_{\rm A} \exp\left(-\delta_{\rm A} M/kT_s^{\rm A} - AMm/kT_I\right) \operatorname{sh}\left(A/4kT_I\right)}{2 \operatorname{ch}\left(\delta_{\rm A}/2kT_s^{\rm A}\right) \operatorname{sh}\left[A\left(2I+1\right)/kT_I\right]},$$
(1)

where  $n_A$  is the concentration of A ions and  $M = \pm \frac{1}{2}$  and  $-I \leqslant m \leqslant I$  are the electronic and nuclear spin quantum numbers. If the gap between  $T_I$  and  $T_S^A$  is sufficiently large, a partial population inversion arises, and stimulated-emission signals appear<sup>8,9</sup> in the ESR spectrum of the <sup>59</sup>Co<sup>2+</sup> ions at the hyperfine transitions  $\frac{1}{2}, m \leftrightarrow -\frac{1}{2}, m$  with quantum numbers m < 0 satisfying the relation<sup>7</sup>

$$-Am/T_{I} > \delta_{A}/T_{S}^{A}.$$
 (2)

It has been shown<sup>10</sup> that a thermal maser of this type, like the gasdynamic laser,<sup>2,3</sup> can be treated as a quantum

heat engine executing a cyclic process which has all the characteristic features of the Carnot cycle. Here the A ion plays the role of a working medium with a discrete energy spectrum. The crystal lattice and the system of cooled B spins are the two heat reservoirs at different (but positive) temperatures. The quantum amplification occurs at the expense of energy from the "hot" reservoir (the lattice). The amplified radiation is analogous to mechanical work.

This maser thus embodies the same ideas and principles used in the partial-inversion gasdynamic laser.

**3.** The most logical next step in this program is to search for ways of achieving steady-state quantum amplification in masers schemes of the type described. In this paper we consider one of the ways of solving this problem, using a combination of thermal excitation and microwave pumping.<sup>1)</sup> The basic idea of our proposal is as follows.

It is perfectly obvious that a maser scheme of this type would be capable of cw operation if the polarization of the spins  $S_B$  were kept constant. This can be achieved through the steady-state dynamic polarization of the spins  $S_B$  by a technique of the type used for the dynamic polarization of nuclei (DPN) of diamagnetic atoms through the familiar solid effect.<sup>12,13</sup> To arrange this the crystal must additionally contain paramagnetic ions of a third kind (C) with suitable spectroscopic and relaxational properties.

To demonstrate this proposal let us consider a crystal of yttrium ethyl sulfate (YES) containing the following impurity ions A, B, and C, respectively: <sup>143</sup>Nd<sup>3+</sup>  $[g_{\parallel}^{A} = 3.665, g_{\perp}^{A} = 1.98$  (Ref. 14),  $A_{\parallel} = 0.0375$  cm<sup>-1</sup>,  $A_{\perp} = 0.0198$  cm<sup>-1</sup> (Ref. 15)], <sup>172</sup>Yb<sup>3+</sup>  $[g_{\parallel}^{B} = 3.328, g_{\perp}^{B} \approx 0.01$  (Ref. 16)], and <sup>168</sup>Er<sup>3+</sup>  $[g_{\parallel}^{C} = 1.5, g_{\perp}^{C} = 8.77$  (Ref. 14)]. In a static magnetic field H oriented in the XZ plane at an angle  $\theta \approx 90^{\circ}$  to the crystalline axis  $C_{3} \parallel Z$ , the Zeeman splitting  $\delta_{C}$  of the Er<sup>3+</sup> spins  $S_{C} = 1/2$  satisfy the relation  $\delta_{C} > \delta_{B} \approx A/2$ . By analogy with ordinary DPN it follows that the steady microwave saturation of the forbidden transitions at one of the frequencies

$$v_p^{\pm} = (\delta_c \pm \delta_B)/h \tag{3}$$

will, under certain conditions,<sup>12,13</sup> lead to a steady positive or negative polarization of the spins  $S_B$ . The problem essentially reduces to one of demonstrating that all the necessary conditions can be satisfied under our peculiar circumstances, mainly: 1) The spins  $S_B$  to be polarized are electronic rather than nuclear; 2) the concentration of the spins  $S_B$  is relatively low; 3) the g factor of the spins  $S_B$  is highly anisotropic; 4) the spin-lattice relaxation time of the spins  $S_B$  is relatively short. Using the theory of the solid effect for conditions of good spectral resolution, one can easily show that with the proper choice of ion concentrations and experimental conditions the undesirable influence of factors 1–4 on the parameters determining the degree of polarization of the spins  $S_B$  can be made to cancel.

In fact, in the case of a pair of spins  $S_B^i$  and  $S_C^j$  coupled by a dipole--dipole interaction, the probability  $W_{ij}$ =  $4|q_{ij}|^2 W$  of the forbidden transition involving a flip of the spin  $S_B^i$  contains (together with the probability W of an allowed transition between the Zeeman sublevels of the spins  $S_C$ ) the quantity

$$|q_{ij}|^{2} = \frac{9\beta^{2}(\boldsymbol{g}_{\perp}^{C})^{2}}{4H^{2}r_{ij}^{6}} \left(\frac{\boldsymbol{g}_{\parallel}^{B}}{\boldsymbol{g}_{\perp}^{B}}\right)^{2} F(\boldsymbol{\theta}) \sin^{2}\boldsymbol{\theta}_{ij} \cos^{2}\boldsymbol{\theta}_{ij} \cos^{2}\boldsymbol{\varphi}_{ij}, \quad (\boldsymbol{4})$$

where the polar and azimuthal angles  $\theta_{ij}$  and  $\varphi_{ij}$  define the position of the radius vector  $\mathbf{r}_{ij}$  joining spins  $S_B^i$  and  $S_C^j$  in a spherical coordinate system,  $\beta$  is the Bohr magneton,

$$F(\theta) = \frac{[\sigma - (1 + \sigma^2)^{\frac{1}{2}}]^2}{\sin^2 \theta (\sigma^2 + 1) \{1 + [\sigma - (1 + \sigma^2)^{\frac{1}{2}}]^2\}^2},$$
 (5)

with  $\sigma = (g_{\parallel}^B/g_{\perp}^B)\cot\theta$ ;  $F(\theta = 90^\circ) = \frac{1}{4}$ . It is seen from (4) that the circumstance that the average value of  $r_{ij}^{-6}$  is relatively small on account of the diluteness of the system of spins  $S_B$  can be compensated by the large factor  $(g_{\parallel}^B/g_{\perp}^B)^2$ , which is absent in the ordinary nuclear case but approximately equal to  $1.1 \cdot 10^5$  for  $^{172}$ Yb<sup>3+</sup>. Therefore, the parameter  $\alpha$  appearing in the theory,  $^{12}$  viz.

$$\alpha = 4 \sum_{i} |q_{ij}|^2 \tag{6}$$

can reach the same values  $(10^{-3}-10^{-4})$  as in the case of DPN.

The leakage processes in the system are determined by the quantity

$$(T_{\rm B}')^{-1} \approx T_{\rm B}^{-1} + \frac{n_{\rm A}}{n_{\rm B}} T_{\rm A}^{-1} [1 - (P_{\rm A}^{L})^2], \quad P_{\rm A}^{L} = - \operatorname{th} (\delta_{\rm A}/2kT_{\rm L}),$$
(7)

which characterizes the spin-lattice relaxation rate of the spins  $S_B$  under conditions of effective thermal contact of these spins with the nuclear spins I of the working ions A.<sup>7</sup> Here  $n_{\gamma}$  denotes the concentration of ions of species  $\gamma = A$ , B, and C, and  $T_{\gamma}$  is the electronic spin-lattice relaxation time of the spins  $S_{\gamma}$  on account of the direct electron-phonon interaction of the corresponding ions species with lattice vibrations. Even at rather low temperatures,  $(T'_B)^{-1}$  can exceed the leakage rate by 2–3 orders of magnitude in the ordinary nuclear case, but the undesirable increase in the leakage factor<sup>12</sup>

$$f = n_{\rm B} (T_{\rm B}')^{-1} / n_{\rm C} \alpha T_{\rm C}^{-1}$$
(8)

can be compensated by the correspondingly smaller (by virtue of circumstance 2) ratio  $n_B/n_C$ .

The relatively high (by virtue of circumstance 1) rate of spin diffusion will lead only to a faster establishment of a uniform polarization of the spins  $S_B$  over the volume of the crystal.

Thus the familiar conditions<sup>12</sup> for the solid effect,

$$\alpha^{-1}T_{c}^{-1} \gg W \gg f T_{c}^{-1}, \ T_{c}^{-1},$$
(9)

under which the polarization  $P_B = -\tanh(\delta_B/2kT_s^B)$  of the spins  $S_B$  and the thermal-equilibrium polarization  $P_C^L$  $= -\tanh(\delta_C/2kT_L)$  of the spins  $S_C$  reach the limiting relationship<sup>12</sup>

$$P_{\rm B} = \pm P_{\rm C}^{L}, \tag{11}$$

are not any more stringent than in the case of DPN (if we ignore several undesirable factors described below). This assertion is also supported by numerical estimates made for the YES crystal under the following conditions:  $n_A = 1$  at.%

 $(\approx 1.9 \cdot 10^{-19} \text{ cm}^{-3}), n_B = 2 \text{ at.} \%, n_C = 1 \text{ at.} \%, T_L = 1.5$ K, H = 2.908 kOe, and  $\theta = 88^{\circ}45'$ . Under these conditions  $\delta_B/h = A_{\perp}/2h \approx 300$  MHz, ensuring thermal contact of the electronic spins of  $^{172}\text{Yb}^{3+}$  with the nuclear spins  $I = \frac{7}{2}$  of the  $^{143}\text{Nd}^{3+}$  ions. At the same time, the pump frequency  $v_p^+ = (\delta_C + \delta_B)/h = 36$  GHz and the signal frequency  $v_s^+ = 10.1$  GHz at the transition  $\frac{1}{2}, + I \leftrightarrow -\frac{1}{2}, + I$  of the neodymium ions at maximum inversion fall in technically well-mastered ranges; the latter frequency is determined from the relation

$$v_s^{\pm} = \left(\delta_A \pm AI\right) / h \approx \left(g_{\perp}^{A} \beta H \pm A_{\perp}I\right) / h.$$
(12)

The spin-lattice relaxation rates  $T_{\gamma}^{-1}$  were determined by extrapolation from the experimental curves given in Refs. 14 and 16:  $T_A^{-1} \approx 1.2 \text{ sec}^{-1}$ ,  $T_B^{-1} \approx 0.5 \text{ sec}^{-1}$ ,  $T_C^{-1} \approx 1900$ sec<sup>-1</sup>. For an approximate estimate of the lattice sum in (6) one can replace  $\cos^2 \varphi_{ij}$  in (4) by the average value  $\frac{1}{2}$  and use the results of Ref. 17. This procedure gives for this sum an expression of the form  $(C_B/2) \cdot 16.25a^{-6}$  (Å<sup>-5</sup>), where  $a \approx 14.05$  Å (Ref. 17),  $C_B = 0.02$  is a parameter indicating the fraction of the Y<sup>3+</sup> ions that have been replaced by ions of species *B*. We finally get  $\alpha \approx 1.4 \cdot 10^{-3}$ ,  $(T'_B)^{-1} \approx 1.1$ sec<sup>-1</sup>,  $f \approx 0.8$ . Condition (10) holds easily, and, in view of the large value of  $g_{1}^{c}$ , condition (9), where  $W \propto (g_{1}^{c})^{2}$ , is also easily satisfied.<sup>2)</sup>

Figure 1 shows the population distribution in the spin system of our YES crystal for the cases of positive and negative temperatures  $T_{S}^{B}$  of the spins  $S_{B}$ .

4. Let us now discuss the maser characteristics of this class of paramagnetic crystals. In the idealized limiting case, Eq. (11) with  $\delta_B = A/2$  implies  $T_S^B = \pm AT_L/2\delta_C$ .

Further assuming that  $m = \mp I$ ,  $T_I = T_S^B$ , and  $T_S^A = T_L$  in (2) and taking (3) and (12) into account, we transform the inversion condition (2) into

$$v_s^{\pm}/v_p^{\pm} < 2I. \tag{13}$$

It follows from (13) that maser schemes of this type in principle admit a large (for  $I = \frac{7}{2}$ , almost seven-fold) excess of the amplified-signal frequency  $\nu_s^{\pm}$  (12) over the microwave jump frequency  $\nu_p^{\pm}$  (3); this excess is considerably greater than the 1.5-fold excess permissible in the maser schemes known previously.<sup>6</sup> The signal frequency can thus be tuned over wide limits by changing the magnitude and direction of the field **H**, even at a fixed pump frequency.

Because  $g_{\perp}^{A} < g_{\perp}^{C}$  in the <sup>143</sup>Nd<sup>3+, 172</sup>Yb<sup>3+, 168</sup>Er<sup>3+, 172</sup>YES crystal, the relation  $v_{s}^{\pm} > v_{p}^{\pm}$  cannot be satisfied for this particular object.<sup>3)</sup> However, in this material one can apparently demonstrate another important advantage of the proposed maser schemes when  $hv_{p}^{\pm} > hv_{s}^{\pm} \sim kT_{L}$ , namely, the high values of the inversion coefficient  $K_{\pm I}$  for the transitions with  $m = \pm I$ . In particular, a calculation by Eq. (1) for the conditions used in the limiting case described above gives  $K_{\pm I} \approx 14.6$ . For comparison we note that in the idealized three-level scheme with the same pump and signal frequencies the inversion coefficient is  $\approx 2.6$ .

Quantum amplification can be achieved simultaneously for several neighboring hyperfine transitions 1/2,  $m \leftrightarrow -1/2, m$  with numbers *m* of the same sign, but the inversion coefficient falls off with decreasing |m|. Maser amplification is also possible for  $T_S^B < 0$ , at the low frequency  $v_B = \delta_B / h \approx A / 2h$ .

In the case when the vector  $\mathbf{H}_1$  of the magnetic component of the microwave signal is oriented along the Z axis, the



FIG. 1. Population distribution in the spin system of an yttrium ethyl sulfate crystal containing impurity ions <sup>143</sup>Nd<sup>3+</sup>, <sup>172</sup>Yb<sup>3+</sup>, and <sup>168</sup>Er<sup>3+</sup> (species A, B, and C, respectively) at saturation of the forbidden transitions at frequencies  $v_p^- = (\delta_C - \delta_B)/h$  and  $v_p^+$  $= (\delta_C + \delta_B)/h$ . The energies are  $E_A = \delta_A M + AMm$ ,  $E_B = \delta_B M_B$ ,  $E_{CB} = \delta_C M_C + \delta_B M_B$ ;  $M = \pm \frac{1}{2}$ ,  $M_B = \pm \frac{1}{2}$ , and  $M_C = \pm \frac{1}{2}$  are the electron spin quantum numbers  $(S_A = S_B = S_C = \frac{1}{2})$ ; the quantization direction in this case is along the field H. The dot-and-dash curves in the left-hand parts of a and b correspond to  $\exp(-\delta_A/kT_L)$ , where  $T_L$  is the lattice temperature, the dashed curve corresponds to  $\exp(-A/2kT_I)$ , and the dashed curve in the middle and left-hand parts of a and b corresponds to  $\exp(-\delta_B/kT_S^B)$ . The spin temperature  $T_S^B \approx T_I$ . The right-hand parts of a and b shows the population distribution in a system of pairs of spins  $S_B^i$  and  $S_C^i$  coupled by a dipole-dipole interaction. The population distribution over the sublevels of the ions <sup>143</sup>Nd<sup>3+</sup> and <sup>172</sup>Yb<sup>3+</sup> at a positive temperature  $T_S^B$  (part a) is analogous to that which occurs in the crystal <sup>59</sup>Co<sup>2+</sup>: Ce<sup>3+</sup>: LaMN.<sup>8,9</sup> imaginary part  $\chi''$  of the complex magnetic susceptibility of our material at the frequency of the hyperfine transition with m = + I can be found from the formula<sup>19</sup>

$$4\pi\chi'' = \frac{\pi (g_{\parallel}^{A})^{2}\beta^{2}K_{+I}n_{A}\Delta\tilde{n}_{L}(+I)}{2\hbar (\Delta\nu)_{A}}$$
(14)

Here  $\Delta \tilde{n}_{I}$  (+ I) is the thermal-equilibrium value of the difference of the relative (per ion) populations at the transition with m = +I, and  $(\Delta v)_A \approx 30$  MHz<sup>19</sup> is the width of the individual hyperfine components in the ESR spectrum of <sup>143</sup>Nd<sup>3+</sup>. For the conditions described above, the limiting value of  $4\pi\chi''$  is  $\approx 0.3$ , more than an order of magnitude greater than the typical value  $4\pi \chi'' = 0.01$  for the ruby maser.<sup>6</sup> It follows that our material can be better than ruby in one of the most important characteristics  $(\chi'')$  provided only that the optimum values of the concentration  $n_A$  and temperature  $T_r$  will not be too different from the values used in the calculations. In regard to this last assumption, however, we note the following.

5. We have ignored a number of factors capable of complicating the operation of the proposed maser scheme in the YES crystal and affecting the optimum values of  $n_A$  and  $T_I$ . These factors include: a) the "phonon bottleneck" in the spin-lattice relaxation of the  $Er^{3+}$  ions; b) appreciable inhomogeneous broadening the the ESR lines of  $Er^{3+}$  and  $Yb^{3+}$ ; c) cross relaxation processes between the electronic spins of  $Nd^{3+}$  and  $Er^{3+}$ ; e) cross relaxation in the system of hyperfine lines of the <sup>143</sup>Nd<sup>3+</sup> ions. Similar factors can come into play in other objects as well, and so a discussion of their role, even on a qualitative level and for a particular example, is of definite interest.

Judging from the experimental studies,<sup>20,21</sup> factors a and b can be important under the conditions described above; this circumstance, as in the case of DPN,<sup>12,13</sup> is undesirable. It follows from Ref. 13, however, that even a very narrow phonon bottleneck does not increase the polarization attainable in the solid-effect method to below  $2P_{C}^{L}/3$ . Estimates based on the data of Ref. 21 show that the polarization decrease due to factor b should not be catastrophic either.

The experimental data of Ref. 22 imply that the rate of process c under the chosen conditions is negligibly low. Processes d can be as undesirable here as the analogous cross relaxation processes are in ruby,<sup>6</sup> although situations are possible in which these processes will increase the inversion. In view of the appreciable difference in the splittings  $\delta_A$  and  $\delta_C$  it seems unlikely that these processes will play an important role at the chosen concentrations  $n_A$  and  $n_C$ . If this turns out not to be the case, then the undesirable influence of this cross relaxation (like that of the possible phonon bottleneck in the spin--lattice relaxation of Er<sup>3+</sup>) can in principle be weakened by decreasing the concentration  $n_{\rm C}$  of the  $Er^{3+}$  ions. [The corresponding undesirable increases in the leakage factor (8) can be compensated either by making a small increase in the angle  $\theta$ , which would lead to an increase in the parameter  $\alpha$  (6) on account of the strong angular dependence of the function  $F(\theta)$  (5), or by replacing the  $^{172}$ Yb<sup>3+</sup> ions by  $^{162}$ Dy<sup>3+</sup> ions  $(g_{\parallel}^{B} = 10.9, g_{\perp}^{B} \approx 0.08)^{23}$  and choosing  $\theta = 90^\circ$ .]

It follows from the physics of the solid effect that for fixed  $n_B$  and  $n_C$  there should be a concentration  $n_A$  that optimizes  $\chi''$  (14) even in the absence of processes d and e. However, these processes can actually turn out to be the most important factors determining the where the optimum occurs and the value reached there. Judging from a comparison of the ESR data for  $^{143}Nd^{3+}$  ions in YES and for X- $^{59}\text{Co}^{2+}$  in LaMN and from the fact<sup>9</sup> that cross relaxation of type e does not manifest itself in the latter at a concentration  $1.6 \cdot 10^{16} \text{ cm}^{-3}$ , one can conclude that processes of type e are also unimportant in the YES crystal at the same concentration of neodymium ions. It follows that the optimum value of  $n_A$  is greater than  $1.6 \cdot 10^{18}$  cm<sup>-3</sup>, at which value Eq. (14) gives  $4\pi \gamma'' \approx 0.03$ .

Thus, it cannot be ruled out that the optimum values of  $n_A$  and  $T_I$  can be substantially different from the values used in the calculations, but it can apparently be assumed that inversion can nevertheless be achieved and that the optimum value of  $\gamma''$  will turn out to be of the same order of magnitude as in the ruby maser.<sup>4)</sup>

6. From the thermodynamic and quantum-electronics standpoint, the proposed maser scheme is a combination of a spin refrigerator, in which microwave excitation is used to create a gap between the temperatures of two quantum subsystems (the crystal lattice and the  $S_B$  spin system), and a thermal paramagnetic maser,<sup>7-9</sup> in which a partial population inversion is achieved on account of this temperature gap (Fig. 2).

The actual maser part of this scheme there should execute a cyclic process like that described in Sec. 2.<sup>5)</sup> However, unlike the case of Refs. 7–9, here it is possible, for  $T_{S}^{B} < 0$ , to have a quantum analog of the Carnot cycle between two heat reservoirs having temperatures of opposite sign but finite and minimally different absolute values. This circumstance is of direct interest in connection with questions<sup>3,25</sup> concerning the fundamental analogy between masers and heat engines. (Such a possibility has never before been realized in any quantum-electronic device).

In classical masers and lasers with microwave or optical pumping the exciting radiation performs two interrelated

FIG. 2. Idealized equivalent thermodynamic scheme of the <sup>143</sup>Nd<sup>3+</sup> <sup>172</sup>Yb<sup>3+</sup>;<sup>168</sup>Er<sup>3+</sup>:YES crystal maser. The arrows show the direction of the main energy flows between the different quantum subsystems, which are denoted as follows: He) liquid helium, L) crystal lattice, R) amplified microwave radiation, A) spin system of working ions of species A, B) system of polarized spins  $S_B$ , C) system of electron spins  $S_C$ , P) microwave pump. The dashed arrows correspond to the case of a negative temperature of spins  $S_B$ .



functions, serving both as the means of creating the population inversion and as the energy source sustaining the quantum amplification. In the proposed scheme the exciting radiation performs mainly just the first of these functions. For  $T_S^B > 0$  the amplification is sustained solely by the energy of the thermal phonons of the lattice, while for  $T_S^B < 0$  it is also sustained partly at the expense of energy from the reservoir of polarized spins  $S_B$  (see Fig. 2), but the latter contribution is relatively small.

7. The compound YES is an extremely convenient object for both theoretical constructions and experimental studies, which can be done by the usual maser techniques<sup>6</sup> with the adoption, where necessary, of suitable methods borrowed from DPN.<sup>12,13</sup> Even so, since YES is crystal hydrate it is of dubious practical interest. However, successful demonstrations with this material, besides satisfying a purely scientific interest, could stimulate a purposeful search for new objects of a similar kind, having substantially better maser characteristics than ruby and rutile in frequency regions with a mature technology<sup>6</sup> and/or being suitable for use in the frequency region ~  $10^{11}$  Hz, where present-day maser technology is poorly developed.

ly rules out cross relaxation of types d and e, but for this same reason  $\chi''$  will be extremely small (this, however, is not very important for a first demonstration).

<sup>5)</sup>In the refrigerator part of the scheme a second cyclic process is executed, involving the spins  $S_c$ . This process (in the idealized version) includes successive repetition of the steps: 1) absorption of a photon of energy  $(\delta_c \pm \delta_B)$  from the microwave pump in a forbidden transition (involving a flip of spin  $S_B$ ); 2) an allowed relaxation transition with the transfer of a quantum of energy  $\delta_c$  to the lattice; 3) exchange of a quantum of energy  $\delta_B \approx A/2$  with the spin system of the working ions (absorption of a quantum for  $T_S^B > 0$  or transfer of a quantum for  $T_S^B < 0$ ), accompanied by the depolarizing flip of a spin  $S_c$ .

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Translated by Steve Torstveit

<sup>&</sup>lt;sup>1)</sup>Definite possibilities for achieving cw operation are also being examined in connection with the idea<sup>11</sup> of using the principles of quantum electronics to directly transform mechanical work into coherent microwave radiation by rotating crystals such as <sup>59</sup>Co<sup>2+</sup>:Ce<sup>3+</sup>:LaMN in a static magnetic field.

<sup>&</sup>lt;sup>2)</sup>A steady-state dynamic polarization of the spins of Ce<sup>3+</sup> ions in the crystal <sup>59</sup>Co<sup>2+</sup>:Ce<sup>3+</sup>:LaMN has been achieved experimentally through microwave saturation of one of the cobalt hyperfine transitions with  $m = \pm \frac{1}{4}$  under conditions of thermal contact between the nuclear spins of Co<sup>59</sup> and the spins of the Ce<sup>3+</sup> ions. On the strength of this it is natural to suppose that a polarization of the spins  $S_B$  in systems of the type under discussion can be achieved in an analogous way through the use of ions of species C enriched with some odd isotope under conditions of simultaneous thermal contact between the spins  $S_B$  and the nuclear spins of the A and C ions. Discussion of this paper.

<sup>&</sup>lt;sup>3)</sup>A suitable crystal for demonstrating this possibility might be  ${}^{143}Nd^{3+}:{}^{172}Yb^{3+}:{}^{159}Tb^{3+}:YES$ , where  ${}^{159}Tb^{3+}$  is used as the C ion.

<sup>&</sup>lt;sup>4)</sup>A more suitable system for eliminating the undesirable factors of types a-e is Sm<sup>3+</sup>:<sup>172</sup>Yb<sup>3+</sup>:<sup>142</sup>Nd<sup>3+</sup>:YES (with  $n_A \approx 0.25$  at.%,  $n_B \approx 1$  at.%,  $n_C \approx 1$  at.%, and  $v^{\pm} \leq 36$  GHz), in which the role of the active centers is played by <sup>147</sup>Sm<sup>3+</sup> or <sup>149</sup>Sm<sup>3+</sup> (Ref. 14). In this system the phonon bottleneck effect should not be manifested in the spin--lattice relaxation of the C ions (Nd<sup>3+</sup>), as far as one can tell from the experimental studies.<sup>19,24</sup> The ESR lines of the Nd<sup>3+</sup> ions are considerably narrower<sup>24</sup> than in the case of Er<sup>3+</sup>. The small g factor of Sm<sup>3+</sup> ( $g^{+}_{\parallel} g^{-}_{\perp} \approx 0.6$ )<sup>14</sup> practical-

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