THE CONDITION FOR SELF-EXCITATION OF A LASER

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The problem of the conditions for self-excitation is considered for devices of the type of a molecular generator with a resonating cavity with dimensions much larger than the generated wavelength. The problem is solved by the method of expansion in terms of eigenfunctions of the cavity with ideally conducting walls. The limits of applicability of the results to real cavities are estimated.

THE study of features of the operation of quantum-mechanical generators and amplifiers that use resonating cavities of dimensions much larger than the wavelength has become interesting because of the practical development of such devices in the optical range of wavelengths (lasers).[1,2] One particularly important question is that of the conditions for self-excitation of a laser. Calculations on this have been made by Schawlow and Townes.[3] The problem can, however, be treated by much more rigorous methods than those used by the authors of [3].

Let us consider a system of weakly interacting molecules* with two energy levels, which completely fills the resonating cavity.† The state of the system is characterized by the density of the energy spin $\mathbf{s}(\mathbf{r},t)$, whose components s_1, s_2, and s_3 obey the following equations of motion[5]:

$$\begin{aligned}
&\dot{s}_1 + \omega_0 s_2 + \frac{1}{T_2} s_1 + \frac{1}{\hbar}\sum_\lambda \mathbf{A}_\lambda(\mathbf{r})\,\mathbf{e}_2 s_3 q_\lambda = 0,\\
&\dot{s}_2 - \omega_0 s_1 + \frac{1}{T_2} s_2 - \frac{1}{\hbar}\sum_\lambda \mathbf{A}_\lambda(\mathbf{r})\,\mathbf{e}_1 s_3 q_\lambda = 0,\\
&\dot{s}_3 = \frac{1}{T_1}(s_3^0 - s_3) - \frac{1}{\hbar}\sum_\lambda \mathbf{A}_\lambda(\mathbf{r})\,(\mathbf{e}_1 s_2 - \mathbf{e}_2 s_1)\,q_\lambda;\\
&\ddot{q}_\lambda + \frac{\omega_\lambda}{Q_\lambda}\dot{q}_\lambda + \omega_\lambda^2 q_\lambda = \int_{V_c} \mathbf{A}_\lambda(\mathbf{r})\,(\mathbf{e}_1 s_1 + \mathbf{e}_2 s_2)\,dV.
\end{aligned} \tag{1}$$

Here ω_0 is the frequency of the molecular transition, ω_λ are the natural frequencies of the resonator, Q_λ are the quality factors corresponding to these frequencies, T_1 and T_2 are the Bloch relaxation times, and $\mathbf{e}_1$ and $\mathbf{e}_2$ are vector molecular constants. These constants can be expressed in terms of the matrix elements of the dipole moment. According to [5]

$$\frac{1}{c}\frac{d\hat{\mu}}{dt} = \mathbf{e}_1 r_1 + \mathbf{e}_2 r_2, \tag{2}$$

where $\hat{\mu}$ is the operator for the dipole moment of the molecule, and r_1 and r_2 are spin matrices. In the representation in which the energy operator of the isolated molecule is diagonal we have

$$r_1 = \frac{1}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad r_2 = \frac{1}{2}\begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}.$$

The relation (2) gives the connection between $\mathbf{e}_1$ and $\mathbf{e}_2$ and the matrix elements of $\hat{\mu}$ calculated with the eigenfunctions of the energy operator of the isolated molecule:

$$\mathbf{e}_1 + i\mathbf{e}_2 = (2i\omega_0/c)\,\mu_{21}, \qquad \mathbf{e}_1 - i\mathbf{e}_2 = (-2i\omega_0/c)\,\mu_{12}. \tag{3}$$

We note by the way that the oscillating part of the mean dipole moment is linearly polarized only in cases in which $\mathbf{e}_1$ and $\mathbf{e}_2$ are collinear. In fact,

$$\begin{aligned}
\langle\psi|\hat{\mu}|\psi\rangle_{\text{osc}} &= \langle c_1\psi_1 + c_2\psi_2|\hat{\mu}|c_1\psi_1 + c_2\psi_2\rangle\\
&= c_1^* c_2\,\mu_{12} e^{-i\omega_0 t} + c_1 c_2^*\mu_{21} e^{i\omega_0 t},
\end{aligned} \tag{4}$$

and it becomes obvious that our assertion is correct when one substitutes μ_{12} and μ_{21} from Eq. (3) into Eq. (4).

In writing out the system (1) we have used the expansion of the vector potential of the electromagnetic field in terms of the eigenfunctions of the resonator:

$$\mathbf{A}(\mathbf{r},t) = \sum_\lambda \mathbf{A}_\lambda(\mathbf{r})\,q_\lambda(t), \qquad \int_{V_c} \mathbf{A}_\lambda^2\,dV = 4\pi c^2. \tag{5}$$

Let us introduce the notation

$$\mathbf{A}_\lambda \mathbf{e}_1/\hbar = \alpha_{1\lambda}, \qquad \mathbf{A}_\lambda \mathbf{e}_2/\hbar = \alpha_{2\lambda}, \qquad \alpha_{2\lambda} - i\alpha_{1\lambda} = \alpha_\lambda \tag{6}$$

and make the change of variables $s_1 + is_2 = P_1$, $s_1 - is_2 = P_2$; the system (1) then takes the form

*By molecules we here mean any quantum-mechanical objects with two levels.

†The disk type of resonator used in practice is not a closed system, but for the main modes of oscillation the field is practically all concentrated in the space between the disks. This is confirmed by calculations made by Fox and Li[4]. Therefore our analysis includes the case of disk resonators.

$$\dot{P}_1 + (T_2^{-1} - i\omega_0) P_1 + \sum_\lambda \alpha_\lambda q_\lambda s_3 = 0, \quad (7a)$$

$$\dot{P}_2 + (T_2^{-1} + i\omega_0) P_2 + \sum_\lambda \alpha_\lambda^* q_\lambda s_3 = 0, \quad (7b)$$

$$\dot{s}_3 = \frac{1}{T_1}(s_3^0 - s_3) + \frac{1}{2}\sum_\lambda (P_1\alpha_\lambda^* + P_2\alpha_\lambda) q_\lambda, \quad (7c)$$

$$\ddot{q}_\lambda + \frac{\omega_\lambda}{Q_\lambda}\dot{q}_\lambda + \omega_\lambda^2 q_\lambda = -\frac{i\hbar}{2}\int_{V_c} (P_1\alpha_\lambda^* - P_2\alpha_\lambda)\, dV. \quad (7d)$$

Since, by hypothesis, the working substance uniformly fills the entire volume of the resonating cavity, it is convenient to make an expansion of $P_1(\mathbf{r}, t)$ and $P_2(\mathbf{r}, t)$ in terms of the system of functions α_λ and α_λ^*:

$$P_1(\mathbf{r}, t) = \sum_\lambda \alpha_\lambda(\mathbf{r})\, P_{1\lambda}(t), \qquad P_2(\mathbf{r}, t) = \sum_\lambda \alpha_\lambda^*(\mathbf{r})\, P_{2\lambda}(t). \quad (8)$$

The completeness and orthogonality of the system α_λ follows from the properties of the system $\mathbf{A}_\lambda$.*

Our set problem of finding the conditions of self-excitation of the generator presupposes an analysis of (7) for stability. Let us assume that at the initial time the quantities $P_{1\lambda}$, $P_{2\lambda}$, and q_λ are close to 0 and $s_3 = s_3^0$ does not depend on the space coordinates.† The replacement of s_3 by s_3^0 in (7a) and (7b) linearizes these equations and allows us to go from a system of an infinite number of coupled equations to a finite system. Multiplying both sides of (7a) by α_λ^* and both sides of (7b) by α_λ and integrating them over the cavity volume V_c, we get

$$\dot{P}_{1\lambda} + (T_2^{-1} - i\omega_0) P_{1\lambda} + s_3^0 q_\lambda = 0, \quad (9a)$$

$$\dot{P}_{2\lambda} + (T_2^{-1} + i\omega_0) P_{2\lambda} + s_3^0 q_\lambda = 0. \quad (9b)$$

Integrating both sides of (7c) and the right side of (7d), we arrive at the equations

$$V_c \dot{s}_3 = \frac{a^2}{2}\sum_\lambda (P_{1\lambda} + P_{2\lambda}) q_\lambda, \quad (9c)$$

$$\ddot{q}_\lambda + \frac{\omega_\lambda}{Q_\lambda}\dot{q}_\lambda + \omega_\lambda^2 q_\lambda = \frac{i\hbar a^2}{2}(P_{2\lambda} - P_{1\lambda}), \quad a^2 = \int_{V_c} \alpha_\lambda \alpha_\lambda^*\, dV. \quad (9d)$$

Let us now assume that there is a small perturbation of the form

$$P_{1\lambda}^0 e^{i\xi_\lambda t}, \qquad P_{2\lambda}^0 e^{i\xi_\lambda t}, \qquad q_\lambda^0 e^{i\xi_\lambda t}, \quad (10)$$

where $\xi_\lambda = \Omega_\lambda + i\delta_\lambda$. Substitution of (10) in (9a)—(9c) leads to a system of homogeneous algebraic equations, which has a non-trivial solution when the determinant is equal to zero:

$$\xi_\lambda^4 - i\xi_\lambda^3(\omega_\lambda/Q_\lambda + 2/T_2) - \xi^2(\omega_\lambda^2 + \omega_0^2 + T_2^{-2} + 2\omega_\lambda/Q_\lambda T_2) + i\xi_\lambda\omega_\lambda[2\omega_\lambda/T_2 + (\omega_0^2 + T_2^{-2})/Q_\lambda] + \omega_\lambda^2(\omega_0^2 + T_2^{-2}) + \hbar a^2\omega_0 s_3^0 = 0. \quad (11)$$

Confining ourselves to the case $|\delta_\lambda| \ll \Omega_\lambda$ and neglecting terms in δ_λ^2, δ_λ^3, and δ_λ^4, we can write instead of Eq. (11) two real equations which determine Ω_λ and δ_λ as functions of ω_λ, s_3^0, and so on. We shall be interested in the occurrence of solutions of the system (9) that increase with the time. From physical considerations it is clear that for $s_3^0 > s_{3\,cr}^0$ there must be increasing solutions characterized by $\delta_\lambda < 0$.* The condition $\delta_\lambda < 0$ leads to the following relations:

$$\Omega_{\lambda\,cr}^2 = \frac{\omega_\lambda(\omega_0^2 T_2 + T_2^{-1} + 2Q_\lambda\omega_\lambda)}{\omega_\lambda T_2 + 2Q_\lambda}, \quad (12)$$

$$s_{3\,cr}^0 = \frac{\omega_\lambda[(\omega_0^2 - \Omega_{\lambda\,cr}^2 + T_2^{-2})^2 + 4T_2^{-2}\Omega_{\lambda\,cr}^2]}{2Q_\lambda T_2^{-1}\omega_0\hbar a^2}. \quad (13)$$

The quantity $s_{3\,cr}^0$ depends on ω_λ, and to a certain characteristic frequency of the cavity there corresponds a value $(s_{3\,cr}^0)_{min}$ below which there is no instability in the system. The quantity $(s_{3\,cr}^0)_{min}$ can be determined from Eq. (13), and the corresponding oscillation frequency is given approximately by $\Omega_\lambda^2 = \omega_0^2 - T_2^{-2}$. If we use the fact that $T_2^{-2} \ll \omega_0^2$, so that without much error we can set $\omega_\lambda = \Omega_\lambda = \omega_0$, we get a simple formula for the boundary of the region of self-excitation:

$$(s_{3\,cr}^0)_{min} \approx 2\omega_0^2/\hbar a^2 Q_\lambda T_2. \quad (14)$$

The quantity a^2 that appears here is given by

$$a^2 = \frac{1}{\hbar^2}\int_{V_c} [(\mathbf{A}_\lambda \mathbf{e}_1)^2 + (\mathbf{A}_\lambda \mathbf{e}_2)^2]\, dV. \quad (15)$$

The simplest special case is that in which $\mathbf{A}_\lambda$ are plane waves and the vectors $\mathbf{e}_1$ and $\mathbf{e}_2$ are collinear. Then the optimal mode, in the sense of its polarization, is that for which $\mathbf{A}_\lambda \parallel \mathbf{e}_1$, and by using the normalization conditions and Eq. (3) one verifies without difficulty that $a^2 = (16\pi\omega_0^2/\hbar^2)|\mu_{12}|^2$. From this we have

$$N_{min} = 2(s_{3\,cr}^0)_{min} = \hbar/4\pi|\mu_{12}|^2 Q_\lambda T_2. \quad (16)$$

Comparing Eq. (16) with the condition for self-excitation of an ordinary molecular generator obtained by Basov and Prokhorov,[6] one readily verifies that they are identical. This fact is not unexpected, since the calculation of Basov and Prokhorov was made on the same assumptions about the character of the polarization of the field in the cavity and about the properties of the dipole

*We are confining ourselves to the case of greatest practical importance, for which $\mathbf{A}_\lambda/|\mathbf{A}_\lambda|$ is a constant vector.

†$2s_3$ has the meaning of the number of active molecules per unit volume, and $s_3 > 0$ describes a state with a preponderance of molecules in the upper level.

*This can be shown directly, because $d\delta/ds_3^0 < 0$.

moment as have been used in obtaining Eq. (16), and is due to the fact that the minimum point on the curve of $s_{3\,cr}^0(\omega_\lambda)$ corresponds to the excitation of only one mode of the cavity. For $s_3^0 > (s_{3\,cr}^0)_{min}$ adjacent modes also become unstable.

In conclusion we remark that the treatment given here has been based essentially on the assumption that the volumes of the matter and the field are the same. It is just owing to this that we could go from the system (7) to the system (9). If we take into account the finite conductivity of the walls of the cavity this assumption is incorrect, owing to the penetration of the field into the metal to the extent of the skin depth. In this case our derivation remains valid, though only approximately, provided a condition which will now be indicated is satisfied.

Let A_λ be the eigenfunctions of a hypothetical cavity with ideally conducting walls and with the same geometry as the actual cavity. Then we have the equation

$$\int\limits_{V_c+V_{sk}} \sum_\mu A_\lambda A_\mu\, dV = \int\limits_{V_c} \sum_\mu A_\lambda A_\mu\, dV + \int\limits_{V_{sk}} \sum_\mu A_\lambda A_\mu\, dV, \quad (17)$$

in which V_c is the volume of the cavity and V_{sk} is the volume of the skin-depth layer. It follows from Eq. (17) that the system of functions A_λ can be used if

$$\int\limits_{V_c} \sum_\mu A_\lambda A_\mu\, dV \equiv \int\limits_{V_c} A_\lambda^2\, dV \gg \int\limits_{V_{sk}} \sum_\mu A_\lambda A_\mu\, dV. \quad (18)$$

Since

$$\sum_\mu \int\limits_{V_{sk}} A_\lambda A_\mu\, dV \leqslant \sum_\mu \int\limits_{V_{sk}} \{\max A_\mu\}^2\, dV = n\{\max A_\mu\}^2 V_{sk},$$

we can replace Eq. (18) by the stronger inequality

$$\int\limits_{V_c} A_\lambda^2\, dV \gg n\{\max A_\lambda\}^2 V_{sk}. \quad (19)$$

It is natural to take n to mean the total number of characteristic frequencies of the cavity that fall within the line width of the molecules. Violation of the inequality (19) is to be expected only in the case of large n, i.e., when the dimensions of the cavity are much larger than the wavelength. Furthermore, according to the normalization condition, $\{\max A_\lambda\}^2 \sim 4\pi c^2/V_c$. Using the fact that the ratio V_c/V_{sk} is approximately equal to the quality figure (cf., e.g., [7]), we finally get the condition for the validity of our approximation in the form

$$Q \gg n. \quad (20)$$

The criterion (20) is satisfied clear up to the optical region. In fact, the number of types of vibrations of a resonator such as a Fabry-Perot interferometer which have sufficiently high values of Q is given, according to [8], by the formula

$$n = LD^4\omega^4\Delta\omega/2\pi^2c^5Q^2.$$

Here $Q = L\omega/c(1-\alpha)$, L is the distance between the plates and D their diameter, and α is the reflection coefficient. For L = 4 cm, D = 0.5 cm, $\omega \approx 3 \times 10^{15}$, $\Delta\omega/\omega = 10^{-3}$, $\alpha = 0.95$, we get $n/Q \approx 0.2$.

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