

## SPECTRAL PROPERTIES OF STIMULATED EMISSION IN A BROAD PUMPING RANGE

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The spectrum of stimulated emission in a stationary generation regime is obtained in the axial-mode model of a plane resonator. The shape of the emission lines is found as a function of the pumping. The existence of a limiting spectral width is established. An additional analysis of the effect of narrowing of the stimulated-emission spectrum during movement of the active centers along the resonator axis is presented.

TANG et al.<sup>[1]</sup> were the first to show that the multiplicity of the axial modes in the spectrum of stationary stimulated emission of lasers causes, even in the case of homogeneous broadening of the luminescence line of the active centers, a spatial inhomogeneity of these modes on the resonator axis. It was established in subsequent investigations<sup>[2,3]</sup> that the extremely weak spatial relaxation of the inverse population in an active medium such as ruby cannot greatly reduce the spatial inhomogeneity of the excitation occurring during the course of generation, and consequently also the multiplicity of modes of the stimulated emission. The results of the cited papers pertain to the case of weak pumping, that is, they describe the spectral laws governing laser emission near the generation threshold.

In a recent paper, Anan'ev and Sedov<sup>[4]</sup> attempted to increase the quantitative accuracy of the results obtained by Tang et al.<sup>[1]</sup> Their calculations were tabulated for a broader pumping range than in<sup>[1]</sup>, and were experimentally verified with a laser  $\text{Sm}^{2+}$  in  $\text{CaF}_2$ . If we disregard the various investigations of the spectral composition of semiconductor laser emission, the latter paper is the only experimental attempt to verify the consistency of the axial-mode model assumed by Tang et al.<sup>[1]</sup> for a qualitative description of the spectral laws governing the stimulated emission in stationary generation.

In the present paper we present an analytic solution of the problem of the spectrum of stationary stimulated emission, using the approximation employed in<sup>[4]</sup>. We present by the same token a refinement of the initial equation which was introduced in<sup>[1]</sup>, an important factor for subsequent quantitative and qualitative comparison of theory and experiment. It will be shown below that the ana-

lytic solution of the problem over a wide range of pumping makes it possible to predict a new effect of the so-called saturation of the spectrum by the axial modes, that is, the existence of a limiting spectral width of laser emission.

In addition, continuing our earlier work,<sup>[5]</sup> we consider in the present article the influence of the pump power on the spectral narrowing of the emission of a laser with active centers that move relative to the resonator mirrors. This effect was predicted for the case of low pumping, and then experimentally confirmed in one of our earlier investigations.<sup>[6]</sup>

1. We start with equations of the balance type for a system of two levels, corresponding to the working transition. The possibility of such a description was investigated earlier.<sup>[7-9]</sup> Assume that  $n_1$  and  $n_2$  are the populations of the lower and upper levels of the working transition, respectively, referred to unit length of the active medium,  $n = n_2 - n_1$ ,  $n_\Sigma = n_1 + n_2 = \text{const}$ ,  $t$  is the time of spontaneous decay of the second level,  $g_i$  is the ordinate of the luminescence line at the frequency of the  $i$ -th mode,  $D$  is a quantity proportional to the stimulated-emission probability,  $L$  is the resonator length,  $P_i = N_i(1 - \cos(2\pi m_i z/L))$  is the energy density of the  $i$ -th mode at the point  $z$  inside the planar resonator,  $N_i$  is the number of photons in the  $i$ -th mode, and  $\gamma_i$  the relative photon loss in the  $i$ -th mode per unit time. If we assume that the rate of change of the population of the lower level due to the pumping is described by a term  $\xi W_p n_1$  ( $W_p$  is the pump power), then

$$\frac{dn_1}{dt} = -\xi W_p n_1 + \frac{n_2}{\tau} + \sum_i D g_i P_i n. \quad (1)$$

Going back to the inverse population  $n$ , we obtain

$$\frac{dn}{dt} = n_{\Sigma} \left( \xi W_p - \frac{1}{\tau} \right) - n \left( \xi W_p + \frac{1}{\tau} \right) - 2 \sum_i Dg_i n P_i. \quad (2)$$

In the absence of stimulated emission, the stationary value of the inverse population is

$$n_0 = \frac{\xi W_p - 1/\tau}{\xi W_p + 1/\tau} n_{\Sigma} = \frac{W_p - W_{eq}}{W_p + W_{eq}} n_{\Sigma}, \quad (3)$$

where  $W_{eq}$  is the pump power at which the level populations become equalized,  $n_0 = 0$ . Introducing in (2) the parameter  $n_0$  and

$$\tau_{eff} = \frac{\tau}{1 + W_p/W_{eq}}, \quad (4)$$

we obtain the equation

$$\frac{dn}{dt} = -\frac{n - n_0}{\tau_{eff}} - 2 \sum_i Dg_i n P_i. \quad (5a)$$

The equation for the number of photons  $N_i$  is of the form

$$\frac{dN_i}{dt} = -\gamma_i N_i + \int_0^L Dg_i n P_i dz. \quad (5b)$$

The essential difference between (5a) from the analogous equation of Tang et al.<sup>[11]</sup> is the dependence of  $\tau_{eff}$  on the pump power.

2. In the stationary case, solving Eq. (5a) with respect to  $n$  and substituting the resulting expression in (5b), we get

$$-\frac{\gamma}{Dg_i n_0} + \int_0^L \frac{1 - \cos(2\pi m_i z/L)}{1 + \Sigma Q_k [1 - \cos(2\pi m_i z/L)]} dz = 0, \quad (6)$$

$$Q_i = 2Dg_i \tau_{eff} N_i.$$

Using the expansion<sup>1)</sup>

$$\frac{1}{1 + \Sigma Q_k [1 - \cos(2\pi m_i z/L)]} \approx \frac{1}{1 + \Sigma Q_k} + \frac{\Sigma Q_k \cos(2\pi m_i z/L)}{(1 + \Sigma Q_k)^2}, \quad (7)$$

we transform (6) and obtain

$$-\frac{\gamma_i}{Dg_i L n_0} + \frac{1}{1 + \Sigma Q_k} - \frac{1}{2} \frac{Q_i}{(1 + \Sigma Q_k)^2} = 0. \quad (8)$$

We shall assume that the resonator is tuned to the center of the homogeneously broadened luminescence line of Lorentz shape. Then

$$g_i = \frac{g}{1 + \beta(j - i + 1)^2}, \quad (9)$$

where  $\beta = (\delta\nu/\Delta\nu)^2$ ,  $\delta\nu$  is the difference in the

frequencies of the neighboring axial modes which go into lasing,  $2\Delta\nu$  is the half-width of the luminescence line, and  $g = (\pi\Delta\nu)^{-1}$ ; we shall henceforth assume also that  $\gamma_i \approx \gamma$ . Solving the system (8), we find that

$$Q_i = 2(1 + \Sigma Q_k) \left[ 1 - \frac{g}{\alpha g_i} (1 + \Sigma Q_k) \right], \quad (10)$$

$$\alpha = DgLn_0/\gamma, \quad (11)$$

$$1 + \Sigma Q_k = \frac{\alpha}{4(2j+1)[1 + \sqrt[3]{\beta j(j+1)}]} \{4j+1 + ((4j+1)^2 + 8\alpha^{-1}(2j+1)[1 + \sqrt[3]{\beta j(j+1)}])^{1/2}\}. \quad (12)$$

From the condition  $Q_{2j+1} \geq 0$  we obtain those values for  $\alpha$  for which generation of the  $(2j+1)$ -st mode begins. Hence

$$\alpha \geq \frac{(1 + \beta j^2)^2}{1 - (\beta/3)j(8j^2 - 3j - 2)}. \quad (13)$$

The generation of the first mode, as seen from (13), sets in for  $\alpha = 1$ . Consequently,  $\gamma = DgLn_0^{thr}$ , where  $n_0^{thr}$  is the inverse population corresponding to the threshold pump power  $W_{thr}$ . Hence

$$\alpha = n_0/n_0^{thr} \quad (14)$$

Using (3), we transform (14) into

$$\alpha = \frac{W_p/W_{eq} - 1}{W_p/W_{eq} + 1} \frac{W_{thr}/W_{eq} + 1}{W_{thr}/W_{eq} - 1}. \quad (15)$$

When  $W_p \rightarrow W_{thr}$ , Eq. (15) goes over into the formula given in<sup>[11]</sup> for small pumping.

Figure 1 shows the variation of the number of modes with  $\alpha$  for different  $\beta$ .

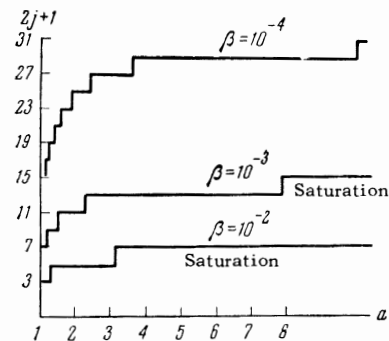


FIG. 1. Number of modes going into lasing at different values of  $\alpha$ .

3. It follows from (13) that no matter how large the parameter  $\alpha$ , the number of modes which participate in the generation is limited (effect of saturation of the spectrum by the modes), and does not exceed  $1.4\beta^{-1/3}$ . Consequently, the maximum width of the stimulated-emission spectrum is

<sup>1)</sup>An analogous approximation was used by Anan'ev and Sedov<sup>[4]</sup>.

$$B \approx 1.4\beta^{-1/3}\delta\nu = 1.4\delta\nu^{1/3}\Delta\nu^{2/3}. \quad (16)$$

On the basis of formula (10) we can obtain the half-width  $\Gamma$  of the stimulated-emission line, which turns out to be equal to  $0.7B$ . Thus, for ruby  $10 \text{ cm}$  long  $B \sim 1.5 \text{ cm}^{-1}$  at room temperature and  $B \sim 0.2 \text{ cm}^{-1}$  at liquid-nitrogen temperature.

In the foregoing estimates we have assumed that  $\alpha$  is unbounded, but it follows from (15) that  $\alpha$  is bounded and tends with increasing pumping to

$$\alpha_{\max} = \frac{W_{\text{thr}}/W_{\text{eg}} + 1}{W_{\text{thr}}/W_{\text{eg}} - 1}. \quad (17)$$

In the case when  $\alpha_{\max}$  is small, it may turn out that  $B$  is not determined by formula (16), but by the quantity  $(2j_{\max} + 1)\delta\nu$ , where  $j_{\max}$  for  $\alpha = \alpha_{\max}$  is calculated by means of (13). Of course, the presence of  $\alpha_{\max}$  does not signify saturation of the radiation intensity with increasing pump power. Since

$$N_i \sim \frac{Q_i}{\tau_{\text{eff}}} = \left( \frac{W_p}{W_{\text{eq}}} + 1 \right) \frac{Q_i}{\tau},$$

the number of photons in the  $i$ -th mode depends linearly on the pump power also when  $\alpha \sim \alpha_{\max}$ .<sup>2)</sup>

In the derivation of the system (8) we made use of an expansion of  $n(z)$  in terms of the parameter

$$(1 + \sum Q_k)^{-1} \sum Q_k \cos(2\pi m_k z / L).$$

The greatest deviation from the exact solution arises in such an approximation in the single-mode case, since the expansion parameter is close to unity at the point  $z = L/2\pi m_j$ . The larger the number of generating modes, the better the agreement with the exact solution. In the case of one mode, Eq. (6) can be solved exactly and

$$Q_1 = 1/4 [4\alpha - 1 - (8\alpha + 1)^{1/2}]. \quad (18)$$

Figure 2 shows plots of  $Q_1$  for the case when one retains in the expansion (7) two terms (our approximation), three terms, the exact solution (18) and the approximation of Tang et al.<sup>[1]</sup> formally continued to the region of large  $\alpha$ . It is seen from the plots that even under the most unfavorable case, that of one mode, there is satisfactory qualitative agreement between the exact and approximate solutions.

Although, as shown above, the width of the stimulated-emission spectrum is limited, nevertheless a large number of modes participate in the

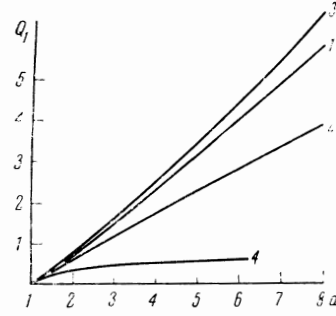


FIG. 2. Dependence of  $Q_1$  on  $\alpha$  in the one-mode regime, obtained 1) for the exact solution, 2), when two terms are left in the expansion of  $n$ , 3), when three terms are left in the expansion of  $n$ , and 4) in the approximation of Tang et al.<sup>[1]</sup>

generation, and the power distribution over the modes is described, as seen from (10) by a parabolic law.

4. Inasmuch as the spatial inhomogeneity is responsible for the multiple-mode nature of the stimulated-emission, it is clear that any effect that leads to a smoothing or reduction of such an inhomogeneity should automatically lead to a reduction (for a given pump power) in the number of generated modes. Thus, it was shown in a number of papers<sup>[2, 3, 10]</sup> that a noticeable decrease in the number of generating modes can be expected in the presence of strong diffusion of the excitation, and in the case of very large diffusion, the generation becomes a single-mode. Unfortunately, in solid-state media the amount of diffusion is governed by the interaction existing between neighboring active centers, and does not lend itself to any noticeable change. One of the methods of equalizing the inhomogeneity of inverse population is the motion of active centers along the axial mode, that is, the motion of active centers relative to the resonator mirrors. A similar problem was considered by us earlier<sup>[5]</sup> for the case of low pumping. In the present article we generalized the formulas of<sup>[5]</sup> for a moving crystal to include the case of large pumping.

Stationary generation, when the active centers of the medium move at constant velocity  $v$  relative to the system of standing waves of a resonator of mirrors on the ends of the crystal, is described by the equations

$$v \frac{dn}{dz} = - \frac{n - n_0}{\tau_{\text{eff}}} - 2 \sum_k Dg_k n N_k \left( 1 - \cos \frac{2\pi m_k z}{L} \right), \quad (19a)$$

$$0 = - \gamma_i + \int_0^L Dg_i n \left( 1 - \cos \frac{2\pi m_i z}{L} \right) dz. \quad (19b)$$

Taking into account the boundary condition  $n(0)$

<sup>2)</sup> It must be borne in mind, of course, that at very large pumps the two-level model employed here is no longer valid.

$= n(L)$  we obtain from (19a) for the case when  $\kappa_i \Sigma Q_k \ll 1$ , where  $\kappa_i = L/2\pi m_i \tau_{\text{eff}} v$  and  $Q_i = 2D\tau_{\text{eff}} g_i N_i$ ,

$$n = n_0 \left\{ \frac{1}{1 + \Sigma Q_h} + \frac{\Sigma Q_h \kappa_h \sin(2\pi m_h z/L)}{1 + \Sigma Q_h} + \Sigma Q_h \kappa_h^2 \cos \frac{2\pi m_h z}{L} \right\}. \quad (20)$$

Substituting (20) in (19b) we obtain the equation

$$-\frac{\gamma_i}{Dg_i n_0 L} + \frac{1}{1 + \Sigma Q_h} - \frac{1}{2} Q_i \kappa_i^2 = 0. \quad (21)$$

Assuming all  $\kappa_i \approx \kappa$ ,  $\gamma_i \approx \gamma$ , and that the number of modes entering into generation is  $2j + 1$ , we get

$$Q_i = \frac{2}{\kappa^2} \left\{ -\frac{1}{\alpha} [1 + \beta(j - i + 1)^2] + \frac{[B^2 + 2\kappa^2(2j + 1)]^{1/2} + B}{2(2j + 1)} \right\},$$

$$B = \alpha^{-1}(2j + 1) [1 + 1/3\beta j(j + 1)] - 1/2\kappa^2, \quad (22)$$

from which follows a criterion for the vanishing of the  $(2j + 1)$ -st mode:

$$\kappa^2 \leq \frac{2j(2j + 1)(2j - 1)(1 + \beta j^2)\beta}{3\alpha(\alpha - 1 - \beta j^2)} \quad (23)$$

It follows from the inequality (23) that when  $v$  increases the number of modes that enter into generation becomes continuously smaller and at sufficiently large velocities the single-mode operation sets in. Figure 3 illustrates well the variations in the spectral composition of generation with increasing velocity.

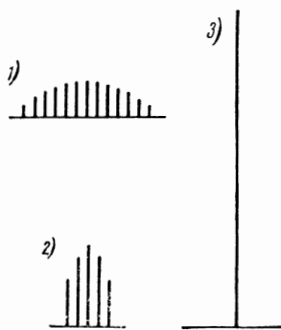


FIG. 3. Change in number of modes and in their relative intensity with increasing velocity  $v$ : 1)  $v = 1.7$  cm/sec, 2)  $v = 6$  cm/sec, 3)  $v = 37$  cm/sec (the estimate of the velocities is for the case of ruby,  $\alpha = 10$ ,  $\beta = 10^{-6}$ ).

To obtain single-mode operation, the relative velocity of the active centers must satisfy the inequality.

$$v \geq \frac{\lambda}{4\pi\tau_{\text{eff}}} \left[ \frac{\alpha(\alpha - 1 + \beta)}{2\beta(1 + \beta)} \right]^{1/2} = \frac{\lambda}{4t_0},$$

$$t_0 = 2\pi\tau_{\text{eff}} \beta(1 + \beta) [\alpha(\alpha - 1 + \beta)]^{-1}. \quad (24)$$

Using furthermore (15) and (17), we obtain

$$v \geq \frac{\lambda}{4\pi\tau} \alpha_{\text{max}} \left[ \left( \frac{W_p}{W_{\text{eq}}} - 1 \right) \left( \frac{W_p}{W_{\text{eq}}} - \frac{W_{\text{thr}}}{W_{\text{eq}}} \right) / 2\beta \right]^{1/2}. \quad (25)$$

It is seen from (25) that, at low pumping,  $v$  should be the smaller the more the threshold pump  $W_{\text{thr}}$  differs from the pump power  $W_{\text{eq}}$  equalizing the level populations of the working transition, that is, the larger the resonator loss. At large pumps  $v \sim \alpha_{\text{max}} W_p$ . According to (17), the smaller the resonator energy loss, the larger the coefficient of proportionality between the velocity and the pump power. Thus, the effect of narrowing down of the stimulated-emission spectrum is easier to realize with the aid of low- $Q$  resonators.

One can expect in the case of the nonstationary operating condition, the generation to be likewise single-mode when the spike duration is  $\Delta t > t_0$ . This effect was observed by us together with V. P. Nazarov and L. K. Sidorenko, for pulsed operation at velocities  $v \sim 30$  cm/sec and small  $\alpha$ .<sup>[6]</sup> The critical time  $t_0$  was of the order of  $10^{-6}$  sec. Of outstanding interest is an experimental verification of the obtained theoretical results for the case of stationary operation of rare-earth or semiconductor lasers. It is probable that the relative motion of the active centers and of the system of standing electromagnetic waves in semiconductor lasers should be realized with a stationary crystal, by moving external mirrors.

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<sup>1</sup>C. L. Tang, H. Statz, and G. de Mars, *J. Appl. Phys.* **34**, 2289 (1963).

<sup>2</sup>B. L. Livshitz and V. N. Tsikunov, *DAN SSSR* **162**, 314 (1965), *Soviet Phys. Doklady* **10**, 446 (1965).

<sup>3</sup>H. Statz, C. L. Tang, and J. M. Lavin, *J. Appl. Phys.* **35**, 2581 (1964).

<sup>4</sup>Yu. A. Anan'ev and B. M. Sedov, *JETP* **48**, 779 (1965), *Soviet Phys. JETP* **21**, 515 (1965).

<sup>5</sup>B. L. Livshitz and V. N. Tsikunov, *DAN SSSR* **163**, 870 (1965), *Soviet Phys. Doklady* **10**, 745 (1965).

<sup>6</sup>B. L. Livshitz, V. P. Nazarov, L. K. Sidorenko, and V. N. Tsikunov, *JETP Letters* **1**, No. 5, 23 (1965), *transl.* **1**, 136 (1965).

<sup>7</sup>C. L. Tang, *J. Appl. Phys.* **34**, 2935 (1963).

<sup>8</sup>J. A. Fleck and R. E. Kidder, *J. Appl. Phys.* **35**, 2825 (1964).

<sup>9</sup>H. Statz and C. L. Tang, *J. Appl. Phys.* **35**, 1377 (1964).

<sup>10</sup>B. L. Livshitz, S. N. Stolyarov, and V. N. Tsikunov, *DAN SSSR*, in press.