

CONCERNING MODE LOCKING IN LASERS

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It is shown that the effect of saturation in the active medium of a solid-state laser does not lead to locking of axial modes. The conditions necessary for equidistance of the laser-generated frequencies are ascertained.

INTRODUCTION

A number of papers published in recent years^[1-6] report observation of a fine temporal structure in the radiation of solid-state lasers in the regime of free (spike) generation. A similar structure is possessed, according to^[7,8], by giant pulses generated during active Q switching. The cause of the rapid oscillations of the intensity was taken by the authors of the cited papers to be the locking of axial modes.

Argumented objections against such an interpretation were advanced by Malyshev et al.^[9], who noted significant shortcomings in the experimental procedure. A more careful experimental verification undertaken by them did not confirm the presence of the effect of locking of axial modes in ruby and neodymium-glass lasers.

The assumed possibility of mode locking is based on the fact that saturation occurs in the active medium, with the result that the excited modes interact nonlinearly with one another¹⁾. A theoretical analysis should answer two questions: a) is such an interaction capable of ensuring equidistance of the generated frequencies, and, if so, then b) to what phase relations between the modes does it lead?

The indicated questions were discussed in^[2,3,11-13]. The analysis was based on the maximum emission principle formulated by Statz and Tang^[2], and it was concluded that mode locking is possible in principle. The Statz and Tang hypothesis reduces to the fact that at given mode amplitudes the phase relations between them should correspond to the maximum power of the stimulated emission. The incorrectness of this hypothesis was demonstrated in^[14], but the question whether mode locking as a result of saturation of the active medium is possible remained open.

In the present paper we solve the problem by analyzing the self-consistent system of equations for a solid-state laser with homogeneously-broadened gain line without using any additional hypotheses.

1. DERIVATION OF ABBREVIATED EQUATIONS

As is well known, a quantum generator is described by a closed system of equations which includes the equations of the electromagnetic field and the equations

¹⁾As is well known, an analogous mechanism ensures mode locking when a medium possessing saturable absorption is present in the laser resonator^[10].

for the components of the density matrix. When averaged over the period of the oscillations $2\pi/\omega_0$, these equations take the form^[15]

$$\frac{de_k}{dt} + \left[\frac{1}{2T_{ck}} + i(\omega_{ck} - \omega_0) \right] e_k = i\omega_0 d \int_{V_c} \varphi_k \sigma dV, \quad (1.1a)$$

$$\frac{\partial \sigma}{\partial t} + \frac{1}{T_2} \sigma = -\frac{id}{2\hbar} \sum_{k=1}^i \varphi_k e_k N, \quad (1.1b)$$

$$\frac{\partial N}{\partial t} + \frac{N - N_0}{T_1} = -\frac{id}{\hbar} \sum_{k=1}^i \varphi_k (e_k^* \sigma - e_k \sigma^*). \quad (1.1c)$$

The symbols have the following meaning: $e_k(t)$ —field intensity of the mode with number k ; $\sigma(t, z)$ —non-diagonal element of the density matrix referred to a unit volume; $N(t, z) = N_2 - N_1$ —density of the population difference of the working levels; N_0 —equilibrium value of N in the absence of generation; $\varphi_k(z)$ —eigenfunctions of the resonator normalized to its volume V_c ; d —matrix element of dipole moment of the transition; ω_0 —transition frequency; ω_{ck} —natural frequencies of the resonator; $T_{ck} = Q_k/\omega_{ck}$ —relaxation time of the radiation in the resonator; T_1 —relaxation time of the population difference; T_2 —reciprocal of the width of the spectral line of the working transition. All the fields are assumed to be polarized in the same plane.

The frequencies of the axial modes of optical resonators satisfy the condition

$$|\omega_k - \omega_m| T_c \gg 1, \quad (1.2)$$

which makes it possible to average Eqs. (1.1) again, but now over the period of the beats between modes. Such a method was used by Ostrovskii^[15] in the investigation of a two-mode quantum generator.

We make the change of variable

$$e_k = v_k \exp[-i(\omega_k - \omega_0)t] \quad (1.3)$$

and expand σ in a series

$$\sigma = \sum_{k=1}^i \sigma_k(t, z) \exp[-i(\omega_k - \omega_0)t]. \quad (1.4)$$

If the generation frequency spectrum ω_k is equidistant, the difference of the populations can be represented in the form of the series

$$N = N(t, z) + \sum_{l=1}^{i-1} N_l(t, z) \exp\{il\Delta\omega t\} + N_i^*(t, z) \exp\{-il\Delta\omega t\}. \quad (1.5)$$

Substituting (1.3)–(1.5) in (1.1b) and neglecting the derivative $\partial \sigma_k / \partial t$, we obtain after averaging

$$\sigma_k = -\frac{idT_2 g_k}{2\hbar} \left(\varphi_k v_k \bar{N} + \sum_{l=1}^{k-1} \varphi_{k-l} v_{k-l} N_l^* + \sum_{l=1}^{s-k} \varphi_{k+l} v_{k+l} N_l \right),$$

$$g_k = [1 - iT_2(\omega_k - \omega_0)]^{-1} \quad (1.6)$$

Expressions for N_l can easily be found by substituting (1.3)–(1.6) in Eq. (1.1c). It is necessary to bear in mind here that N_0 will not depend on the time $| \dot{N}_l / N_l | \ll \Delta\omega$, and that for all the crystals and glasses used in lasers the following inequality holds true:

$$|\omega_k - \omega_m| T_1 \gg 1. \quad (1.7)$$

We present the approximate formula

$$N_i = \bar{N} \frac{id^2 T_2}{2\hbar^2 l \Delta\omega} \sum_{m=1}^{s-i} \varphi_m \varphi_{m+i} (g_m + g_{m+i}^*) v_m v_{m+i}^*, \quad (1.8)$$

the region of validity of which is limited not only by (1.2) and (1.7), but also by the condition $|N_l / \bar{N}| \ll 1$.

This condition, as seen from (1.8), is satisfied when

$$sd^2 T_2 |v_{max}|^2 \ll \hbar^2 \Delta\omega. \quad (1.9)$$

Estimates show that the inequality (1.9) is satisfied up to powers attained in giant pulses. In the same approximation, the following equation is valid for \bar{N} :

$$\frac{\partial \bar{N}}{\partial t} + \frac{\bar{N} - N_0}{T_1} = -\frac{d^2 T_2}{\hbar^2} \sum_k \varphi_k^2 |v_k|^2 \text{Re } g_k \bar{N}. \quad (1.10)$$

The equation for the complex amplitudes of the fields is obtained by substituting (1.3) and (1.4) in (1.1a) and then averaging:

$$\frac{dv_k}{dt} + \left[\frac{1}{2T_{ck}} + i(\omega_{ck} - \omega_k) \right] v_k = i\omega_0 \chi_k v_k. \quad (1.11)$$

The quantity

$$\chi_k = d \int_{V_c} \varphi_k \sigma_k dV / v_k \quad (1.12)$$

is the susceptibility of the active medium at the frequency ω_k .

It will be convenient in what follows, after introducing the mode phases θ_k ,

$$v_k = |v_k| \exp i\theta_k,$$

to change over to real variables. Equation (1.1) then breaks up into two. The obtained closed system of laser equations will be written in dimensionless form

$$\frac{\partial n}{\partial \tau} = \alpha - n \left[1 + \sum_k (1 - \Delta_k^2) \varphi_k^2 x_k^2 \right], \quad (1.13a)$$

$$\frac{d}{d\tau} (x_k^2) = -G x_k^2 (2 \text{Im } \kappa_k + C_k), \quad (1.13b)$$

$$\frac{d\theta_k}{d\tau} = -G_2 (\Delta_{ck} - \Delta_k) + G \text{Re } \kappa_k. \quad (1.13c)$$

The new variables and coefficients are connected with the old ones by the relations

$$\tau = t / T_1, \quad x_k = |v_k| d(T_1 T_2)^{1/2} / \hbar, \quad n = \omega_0 T_2 T_{c1} d^2 \bar{N} / \hbar,$$

$$\alpha = n N_0 / \bar{N}, \quad \kappa_k = \omega_0 T_{c1} \chi_k, \quad G = T_1 / T_{c1}, \quad G_2 = T_1 / T_2,$$

$$C_k = T_{c1} / T_{ck}, \quad \Delta_k = T_2 (\omega_k - \omega_0), \quad \Delta_{ck} = T_2 (\omega_{ck} - \omega_0), \quad \Delta = T_2 \Delta\omega. \quad (1.14)$$

The susceptibility κ_k is made up of linear and nonlinear parts, the expressions for which are obtained from (1.6), (1.8), and (1.12):

$$\kappa_k^L = 1/2 n(kk) (1 - \Delta_k^2) (\Delta_k - i), \quad (1.15)$$

$$\text{Re } \kappa_k^{NL} = \frac{1}{4G_2 \Delta} \left\{ - \sum_{l=1}^{k-1} \sum_{m=1}^{s-l} \frac{x_{k-l} x_m x_{m+l}}{l x_k} n(k-l, k, m, m+l) \right.$$

$$\times [2 \cos \theta_- - (2\Delta_k + \Delta_{m+l} - \Delta_m) \sin \theta_-] + \sum_{l=1}^{s-k} \sum_{m=1}^{s-l} \frac{x_{k+l} x_m x_{m+l}}{l x_k} \times n(k, k+l, m, m+l) [2 \sin \theta_+ + (2\Delta_k - \Delta_{m+l} + \Delta_m) \sin \theta_+] \left. \right\}, \quad (1.16)$$

$$\text{Im } \kappa_k^{NL} = -\frac{1}{4G_2 \Delta} \left\{ \sum_{l=1}^{k-1} \sum_{m=1}^{s-l} \frac{x_{k-l} x_m x_{m+l}}{l x_k} n(k-l, k, m, m+l) \times [2 \sin \theta_- + (2\Delta_k + \Delta_{m+l} - \Delta_m) \cos \theta_-] + \sum_{l=1}^{s-k} \sum_{m=1}^{s-l} \frac{x_{k+l} x_m x_{m+l}}{l x_k} \times n(k, k+l, m, m+l) [2 \sin \theta_+ - (2\Delta_k - \Delta_{m+l} + \Delta_m) \cos \theta_+] \right\}. \quad (1.17)$$

To abbreviate the notation we introduce, in addition to (1.14), the symbols

$$\theta_-(k, m, l) = \theta_{k-l} - \theta_k - \theta_m + \theta_{m+l}, \quad \theta_+(k, m, l) = \theta_k - \theta_{k+l} - \theta_m + \theta_{m+l},$$

$$n(kk) = V_c^{-1} \int_{V_c} n \varphi_k^2 dV, \quad n(k, k', m, m') = V_c^{-1} \int_{V_c} n \varphi_k \varphi_{k'} \varphi_m \varphi_{m'} dV. \quad (1.18)$$

The quantity g_k is expanded in powers of Δ_k :

$$g_k = 1 + i\Delta_k - \Delta_k^2 - i\Delta_k^3,$$

with all terms included in κ_k^L , and only the first two terms in κ_k^{NL} .

Although all the active media used in solid-state lasers, as already mentioned, have large relaxation times T_1 , it is of interest in principle to consider the case of inertialess active media, for which

$$|\omega_k - \omega_m| T_1 \ll 1. \quad (1.19)$$

The equations (1.13) remain valid in this case, too, but the nonlinear susceptibility is no longer described by formulas (1.16) and (1.17). Calculating N_l , we can disregard the derivative $\partial N / \partial t$ in (1.1c), and obtain in place of (1.8) the expression

$$N_i = \bar{N} \frac{d^2 T_1 T_2}{2\hbar^2} \sum_{m=1}^{s-i} \varphi_m \varphi_{m+i} (g_m + g_{m+i}^*) v_m v_{m+i}^*. \quad (1.20)$$

The condition for its validity, which follows from the condition $|N_l / \bar{N}| \ll 1$, is

$$sd^2 T_1 T_2 |v_{max}|^2 \ll \hbar^2. \quad (1.21)$$

Relation (1.20) leads to the following formulas for the components of the nonlinear susceptibility:

$$\text{Re } \kappa_k^{NL} \approx -\frac{1}{2} \left\{ \sum_{l=1}^{k-1} \sum_{m=1}^{s-l} n(k-l, k, m, m+l) \frac{x_{k-l} x_m x_{m+l}}{x_k} \sin \theta_- - \sum_{l=1}^{s-k} \sum_{m=1}^{s-l} n(k, k+l, m, m+l) \frac{x_{k+l} x_m x_{m+l}}{x_k} \sin \theta_+ \right\}, \quad (1.22)$$

$$\text{Im } \kappa_k^{NL} \approx \frac{1}{2} \left\{ \sum_{l=1}^{k-1} \sum_{m=1}^{s-l} n(k-l, k, m, m+l) \frac{x_{k-l} x_m x_{m+l}}{x_k} \cos \theta_- + \sum_{l=1}^{s-k} \sum_{m=1}^{s-l} n(k, k+l, m, m+l) \frac{x_{k+l} x_m x_{m+l}}{x_k} \cos \theta_+ \right\}. \quad (1.23)$$

2. INERTIAL ACTIVE MEDIUM. CASE OF THREE MODES

If the number of excited modes is equal to three, then the right-hand sides of Eqs. (1.13) depend on the single phase combination

$$\theta = \theta_1 - 2\theta_2 + \theta_3$$

and relations (1.16) reduce to

$$\begin{aligned} \operatorname{Re} \kappa_1^{NL} &= \frac{1}{4G_2\Delta} \left\{ n(1122)x_2^2 + \frac{1}{2}n(1133)x_3^2 \right. \\ &\quad \left. + n(1223)\frac{x_2^2x_3}{x_1} [2\cos\theta + (2\Delta_1 - \Delta)\sin\theta] \right\}, \\ \operatorname{Re} \kappa_2^{NL} &= \frac{1}{4G_2\Delta} \{ n(2233)x_3^2 - n(1122)x_1^2 + 2n(1223)x_1x_3\Delta\sin\theta \}, \\ \operatorname{Re} \kappa_3^{NL} &= -\frac{1}{4G_2\Delta} \left\{ n(2233)x_3^2 + \frac{1}{2}n(1133)x_1^2 \right. \\ &\quad \left. + n(1223)\frac{x_2^2x_1}{x_3} [2\cos\theta + (2\Delta_1 + \Delta)\sin\theta] \right\}. \end{aligned} \quad (2.1)$$

The equation for the determination of θ is a linear combination of the equations (1.13c)

$$d\theta/d\tau = -G_2(\Delta_{c1} - 2\Delta_{c2} + \Delta_{c3}) + G \operatorname{Re}(\kappa_1 - 2\kappa_2 + \kappa_3). \quad (2.2)$$

We note that by virtue of the equidistance of the frequencies ω_k the unknown quantities Δ_k have dropped out from the first term of the right hand side of (2.2).

When $d\theta/d\tau = 0$, Eq. (2.2) determines the dependence of θ on the other variables and on the laser parameters. We shall not consider the problem in general form, but confine ourselves to the most interesting particular case of modes with close amplitudes. Such an assumption makes it possible to put $n(1122) = n(2233)$ and to neglect the difference of x_k in a number of terms. Substituting (2.1) in (2.2), we obtain, with allowance for the remarks already made, the equation

$$\begin{aligned} G_2(\Delta_{c1} - 2\Delta_{c2} + \Delta_{c3}) - G \operatorname{Re}(\kappa_1 - 2\kappa_2 + \kappa_3) \\ = \frac{G}{4G_2\Delta} \left\{ \left[2n(1122) - \frac{1}{2}n(1133) \right] (x_1^2 - x_3^2) \right. \\ \left. - 2n(1223)(x_1^2 - x_3^2)\cos\theta - 10n(1223)x_1^2\Delta\sin\theta \right\}. \end{aligned} \quad (2.3)$$

Equation (2.3) does not always have real solutions.

There are certainly no such solutions if the inequality

$$G_2|\Delta_{c1} - 2\Delta_{c2} + \Delta_{c3}| \leq \max \left\{ \frac{G}{G_2}|n(1223)|x_1^2, \frac{G}{G_2\Delta}|(x_1^2 - x_3^2)n(1223)| \right\}. \quad (2.4)$$

which limits the permissible nonequidistance of the natural frequencies of the resonator, is not satisfied.

For a numerical estimate it is necessary to know the order of magnitude of x_k^2 and $n(1223)$. It follows from (1.13) that $n < \alpha$ and in the stationary regime $x_k^2 \lesssim \alpha - 1$. Specifying the characteristic values of the relaxation times $T_1 = 10^{-3}$ sec, $T_2 = 10^{-12}$ sec, $T_C = 10^{-8}$ sec ($G = 10^5$, $G_2 = 10^9$), assuming the modes to be separated in frequency by $\Delta\omega = 10^9$ sec $^{-1}$ ($\Delta = 10^{-3}$) and with a certain margin assuming that $|n(1223)| = \alpha$, $x_1^2 - x_3^2 \sim \alpha - 1$, we obtain from (2.4) the condition

$$|\omega_{c1} - 2\omega_{c2} + \omega_{c3}| \ll 10^2\alpha(\alpha - 1)\text{sec}^{-1}.$$

The excess over the generation threshold α usually does not exceed several times ten, and consequently the requirements concerning the equidistance of the natural frequencies of the resonator are exceedingly stringent. Failure to meet them means that the generated frequencies will likewise be nonequidistant.

The most noticeable deviations of the frequencies ω_k from an equidistant distribution are caused by the same factors that lead to selection of axial modes. The

end faces of the active element, which are parallel to the mirrors, the plane-parallel substrates of the mirrors, the plane-parallel plates or Fabry-Perot interferometers in the space between the mirrors, all these transform the resonator into a system of coupled resonators. As a consequence, the frequencies of the axial modes turn out to be shifted by different and considerable distances from their unperturbed values^[16,17].

It is clear from the foregoing that the effect of mode locking can certainly not be observed in lasers unless great care is taken to eliminate all the factors that lead to mode selection. None of the experiments of this kind described in the literature satisfy this requirement, with the exception of^[9].

If mode selection is completely eliminated, then the generation spectrum is equidistant and the question arises as to the phases of the excited modes. Since in this case $C_k = 1$, the following equation is deduced from (1.13b) for the stationary generation regime:

$$\operatorname{Re} \kappa_k^L = \Delta_k(1 + \operatorname{Im} \kappa_k^{NL})/2. \quad (2.5)$$

In the specified approximation we have

$$\begin{aligned} \operatorname{Im} \kappa_1^{NL} &= -\frac{n(1223)}{2G_2\Delta} \frac{x_2^2x_3}{x_1} \sin\theta, \quad \operatorname{Im} \kappa_3^{NL} = \frac{n(1223)}{2G_2\Delta} \frac{x_2^2x_1}{x_3} \sin\theta, \\ \operatorname{Im} \kappa_2^{NL} &= 0 \end{aligned} \quad (2.6)$$

and (2.3) reduces to

$$\begin{aligned} [2n(1122) - 1/2n(1133)](x_1^2 - x_3^2) \\ = 2n(1223)[(x_1^2 - x_3^2)\cos\theta + 3x_1^2\Delta\sin\theta]. \end{aligned} \quad (2.7)$$

When $|x_1^2 - x_3^2| \gg \Delta$, the solution of (2.7) is

$$\cos\theta = [2n(1122) - 1/2n(1133)]/2n(1223), \quad (2.8)$$

and in the opposite case

$$\sin\theta = 0. \quad (2.9)$$

Each of these solutions gives two possible values of $\bar{\theta}$. In order to ascertain which of them is realized, it is necessary, generally speaking, to investigate the system (1.13) for stability. However, definite information can also be obtained by a simpler method, by specifying a small deviation of the phase $\psi = \theta - \bar{\theta}$, and linearizing Eq. (2.2). In the case $|x_1^2 - x_3^2| \gg \Delta$, such an approach leads to the conclusion that the state with $n(1223)(x_1^2 - x_3^2)\sin\bar{\theta} > 0$, is unstable, and in the case $|x_1^2 - x_3^2| \ll \Delta$ the unstable state is the one for which $n(1223)\cos\bar{\theta} < 0$.

Acting in accordance with the maximum emission principle, we should have calculated the radiation power averaged over the period of the oscillations $W = -\sum_k x_k^2 \operatorname{Im} \kappa_k$. The part of W that depends explicitly

on the phase, starting from (2.6), is equal to zero²⁾. Only allowance for the small terms leads to expression $W(\theta) \sim n(1223)x_1x_2^2x_3\cos\theta$. The quantity $W(\theta)$ reaches a maximum at $\theta = 0$ if $n(1223) > 0$ and $\theta = \pi$ if $n(1223) < 0$. These values of θ are solutions of (1.13c) only if the natural frequencies of the reso-

²⁾An expression $W(\theta) \neq 0$ is obtained in [2] with a similar approximation; this is apparently due to an error in the calculations.

nator are equidistant and the amplitudes of the outermost modes are equal. In all other cases the maximum emission principle leads to an incorrect result.

3. CASE OF LARGE NUMBER OF MODES

The selecting factors are eliminated to the greatest degree when the resonator mirrors are coated directly on the end surfaces of the active element. This is the laser variant which we shall investigate below. To answer the question as to the stationary phases we shall, as in the preceding section, consider the system of equations

$$F_k - F_{k+1} - F_m + F_{m+1} = 0, \quad (3.1)$$

where F_k denotes the right-hand sides of (1.13c). Since the natural frequencies of the resonator are equidistant, and the contribution of the terms $\text{Re } \kappa_k^L$ is small in accord with (2.5), Eqs. (3.1) reduce to

$$\text{Re}(\kappa_k^{NL} - \kappa_{k+1}^{NL} - \kappa_m^{NL} + \kappa_{m+1}^{NL}) = 0. \quad (3.2)$$

The number of equations in (3.2) coincides with the number of phase combinations of the type $\theta_k - \theta_{k+l} - \theta_m + \theta_{m+l}$, and the latter must be found when the system is solved. When $s \gg 1$, however, this procedure is quite laborious and is furthermore unnecessary. It is possible to verify that the values $|\theta(k', m', l')| \ll 1$ corresponding to the locked modes do not satisfy the equations by substituting these values directly in (3.2). From the symmetry of the problem follows equality of the amplitudes of the modes that are located at equal distances from the center of the gain line. As shown in^[18], the intensity of the mode depends on the detuning in accordance with a quadratic law. If now we turn to formulas (1.16), then we can see that, accurate to small terms of higher order, the following relation holds for symmetrically arranged modes:

$$\text{Re } \kappa_k^{NL} = -\text{Re } \kappa_{s-k+1}^{NL}$$

Consequently, the values $|\theta(k', m', l')| \ll 1$ satisfy only those equations of (3.2) in which $m = s - k - l + 1$. This proves the absence of the effect of mode locking in lasers with an inertial active medium.

The situation is somewhat different if the active medium is inertial. Substituting in (3.2) the values of $\text{Re } \kappa_k^{NL}$, which are determined by formulas (1.22), we verify that $\theta_+(k, m, l) = \theta_-(k, m, l) = 0$ is one of the possible solutions of the system. The center of gravity of the problem now shifts to an investigation of the stability. Linearizing the phase equations (1.13c) relative to this stationary value of the phases, we obtain for small deviations $\delta_k = \theta_k - \bar{\theta}_k$ the equation

$$\frac{1}{\theta_k} \frac{d\theta_k}{d\tau} \approx \text{Im } \kappa_k^{NL} - \frac{1}{2} \left[\sum_{l=1}^{k-1} n(k-l, k-l, k, k) x_{k-l}^2 + \sum_{l=1}^{s-k} n(k, k, k+l, k+l) x_{k+l}^2 \right]. \quad (3.3)$$

We see from it that $d\delta_k/d\tau > 0$, i.e., the regime of generation with locked modes is unstable.

Stable mode locking can be obtained only by reversing the sign of the difference of the populations of the medium in which the nonlinear interaction takes place. In practice it is necessary to this end to place in the resonator, in addition to the amplifying element, also a saturating absorber^[10]. Since the interaction of the modes occurs not only in the absorber but also in the amplifying medium, and the latter prevents synchronization, the difference between the absorber level populations should exceed a certain finite value.

In conclusion, we note that in cases when $\theta_{\pm}(k, m, l) = 0, \pi$ satisfy the quantum-generator equations, the maximum emission principle leads to correct phase relations. In these cases this principle may turn out to be a convenient means of solving individual problems, for example determining the dependence of the waveform of the emitted signal on the position of the cell with saturable absorber in the resonator^[19].

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