

Concerning the kinetics of stimulated gamma radiation in the transient regime

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The singularities of the kinetics of stimulated γ radiation are considered, for various values of the broadening factor, on the basis of a system of quasiclassical equations in the weak-signal approximation, with allowance for the time dependence of the stimulated-emission cross section. The characteristics of the developing wave are compared in the γ -ray band and in the optical band in the transient regime. The concepts of the optimal time interval and the active-layer length are introduced and their values are calculated for a real case.

INTRODUCTION

In investigation of the problem of the γ laser^[1-3], attention is concentrated on the possibility of obtaining the threshold concentration of the nuclear isomer, which is determined from the condition that the resonant value of the gain $n\sigma$ ($\omega = \omega_0$) exceed the coefficient of the non-resonant losses. This was the principal condition in determination of the parameters of the necessary inversion mechanism in the Mössbauer region of energies and temperatures for isomers with different lifetimes. However, as will be shown below, use of the foregoing conditions for the particular case of short-lived isomers, whose emission and absorption lines are not broadened, is incorrect and leads to significant errors. What is not permissible here is neglect of the kinetic features of the Mössbauer induced γ radiation, which lead to dependence of the cross section of the process on time and on the character of the amplified signal. Consequently, the cross section of the stimulated emission reaches the resonant value within a time comparable with the lifetime of the inverted state, and this exerts a noticeable influence on the kinetics of the γ -wave development. The present paper is devoted to an investigation of the features of the kinetics in the transient regime.

We choose as the model a single-pass two-level γ amplifier described by the usual system of quasiclassical equations for the amplitudes of the vector-potential A ; these equations can be obtained on the basis of the Maxwell and Schrödinger equations without employing any particular model representations, and are also valid in the γ band:

$$\begin{aligned} \frac{1}{c} \frac{\partial A}{\partial t} + \frac{\partial A}{\partial x} + \frac{1}{2l} A &= \frac{2\pi}{\omega} p, \\ \frac{\partial p}{\partial t} + \left(i\epsilon + \frac{\Gamma}{2} \right) p &= \frac{An_f}{ch} |M|^2, \\ \frac{\partial n}{\partial t} + \frac{n-n_0}{T} &= -\frac{2}{ch} (Ap^* + pA^*). \end{aligned} \quad (1)$$

Here c is the velocity of the γ quanta in the medium, f is the Debye-Waller factor, $\epsilon = \omega - \omega_0$, M is the matrix element of the intranuclear transition, n_0 and n are respectively the initial and running values of the inverted population, Γ is the γ -transition linewidth, T is the lifetime of the upper state, p is the current of the nuclear transition, $l = 1/N\sigma_0$ is the mean free path of the non-resonant γ quantum in matter and is determined by the total cross section σ_0 of the photoabsorption and Compton scattering, which are the principal mechanisms of the losses in Mössbauer spectroscopy, and N is the total concentration of all the nuclei. For the case of a poly-

atomic lattice, the value σ_0 must be averaged with allowance for the concentrations of all the components. We have left out of (1) a term that takes the pumping into account, since we shall assume from now on that at the initial instant of time the inverted population of the working transition is n_0 and that after the initial instant the excitation mechanism is turned off (the optical analog of^[4]).

1. CASE OF BROADENED LINE

We consider the evolution of a γ wave in a sample consisting of a long-lived nuclear isomer. It is known that at lifetimes $T > 10^{-5}$ sec the γ -emission (absorption) line comes to be broadened by various types of mechanisms (homogeneous and inhomogeneous), so that $\Gamma T \gg 1$. We shall henceforth consider the development of a sufficiently weak γ wave, which in many cases is the only possible variant, since an intense γ wave results in heating of the sample and in a violation of its homogeneity, both directly and by the products of the nuclear cascade, so that the Debye-Waller factor decreases and the γ line is broadened. Taking the last factor into account, and considering also the process of development of the γ wave for times $t \ll T$ (in this case t can be either smaller or larger than $1/\Gamma$), we assume that the inverted population changes very little during the characteristic time of wave evolution, i.e., $n \approx n_0$.

Connecting the matrix element of the intranuclear transition, with the lifetime T of the upper state on the basis of the detailed-balancing principle, we transform (1) into

$$\begin{aligned} \frac{1}{c} \frac{\partial A}{\partial t} + \frac{\partial A}{\partial x} + \frac{1}{2l} A &= C_1 \int_0^t A \exp \left[- \left(i\epsilon + \frac{\Gamma}{2} \right) (t-t') \right] dt', \\ C_1 &= \frac{\pi f^2 n_0}{2\omega^2 T (1+\alpha)} \frac{1+2J_2}{1+2J_1}, \end{aligned} \quad (2)$$

where α is the internal-conversion coefficient and J_2 and J_1 are the spins of the upper and lower states of the nucleus.

The solution of Eq. (2) should satisfy the conditions

$$A(0, t) = A_0 e^{-t/T}, \quad t > 0; \quad A(x, 0) = 0; \quad A(x \geq ct) = 0. \quad (3)$$

Applying the Laplace-Carson transformation with respect to the variable t to Eqs. (2) and (3), we obtain

$$\begin{aligned} \bar{A} &= A_0 \frac{a}{a+1/2T} \exp \left(-\frac{x}{2l} - \frac{ax}{c} + \frac{C_1 x}{a+i\epsilon+\Gamma/2} \right), \\ \bar{A} &= a \int_0^\infty e^{-at} A dt. \end{aligned}$$

Using the inversion formula, we obtain an expression for A:

$$A = \frac{A_0}{2\pi i} e^{-x/2l} \int_{\delta-i\infty}^{\delta+i\infty} \frac{1}{a+1/2T} \exp \left[a \left(t - \frac{x}{c} \right) + \frac{C_1 x}{a+ie+\Gamma/2} \right] da.$$

The integrand has a pole at the point $a = -1/2T$ and an isolated essential singularity $a = -i\epsilon - \Gamma/2$, so that the integration is carried out along a line located to the right of the imaginary axis, i.e., $\delta > 0$. We close the integration contour with an arc lying in the left half-plane, with allowance for the fact that the integrand at $t - x/c > 0$ satisfies the Jordan-lemma condition on this arc. Then, using the generating function for the modified Bessel function

$$\exp \left[\frac{1}{2} u \left(v + \frac{1}{v} \right) \right] = \sum_{m=-\infty}^{\infty} v^m I_m(u)$$

and expanding the integrand in a Laurent series, we obtain

$$A = A_0 e^{-x/2l} \left[\exp \left(-\frac{t-x/c}{2T} + \frac{C_1 x}{ie+\Gamma/2-1/2T} \right) - \exp \left[-\left(ie + \frac{\Gamma}{2} \right) \left(t - \frac{x}{c} \right) \right] \sum_{n=1}^{\infty} \left[\frac{1}{ie+\Gamma/2-1/2T} \left(\frac{C_1 x}{t-x/c} \right)^{1/2} \right]^n \right] \times I_n \left(2 \left(C_1 x \left(t - \frac{x}{c} \right) \right)^{1/2} \right). \quad (4)$$

It follows from analysis of (4) that the first term in the square brackets for the case $\Gamma T \gg 1$ is a solution of the usual balance equation of a quantum amplifier, and that the expression $2C_1/n_0 (i\epsilon + \Gamma/2)$ has the meaning of the usual induced-transition cross section σ . However, the presence of the remaining terms complicates the picture. The result (4) is equivalent to the fact that the cross section of the induced radiation for the active center depends on the time relative to the instant of passage of the leading front of the amplified signal through the location of the center considered, i.e., the delay time $(t - x/c)$, and the establishment of the stationary cross section takes place over a time exceeding $1/\Gamma$. This result has a complete analogy in the optical band.

Figure 1 shows the spatial evolution of an optical step signal in a one-dimensional ruby amplifier, obtained from (4) by summing the first few terms of the rapidly-converging series, for two instants of time and for realistic parameters ($\Gamma = 10^{11} \text{ sec}^{-1}$, $C_1 = 10^{10} \text{ cm}^{-1} \text{ sec}^{-1}$, $n_0 = 0.8 \times 10^{19} \text{ cm}^{-3}$, and $l = 30 \text{ cm}$).

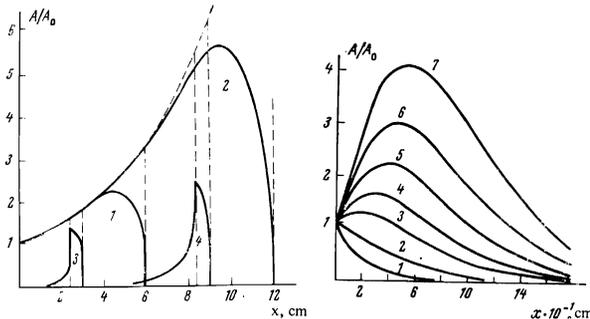


FIG. 1

FIG. 2

FIG. 1. Spatial amplification of optical signal: 1, 2—stepwise signal at $t = 2 \times 10^{-10}$ and 4×10^{-10} sec, respectively; 3, 4—short pulse-type signal at $t = 10^{-10}$ sec and 3×10^{-10} sec.

FIG. 2. Spatial evolution of γ wave at $\Gamma T \gg 1$ for t : 1—plot of the function $e^{-x/2l}$, 2—0.04, 3—0.08, 4—0.1, 5—0.12, 6—0.14, 7—0.16 sec.

It follows from Fig. 1 that the evolution of the stationary cross section (exponential curve) occurs within a time $t \gtrsim 1/\Gamma$. Owing to the very large value of Γ in the optical band, this effect is difficult to observe experimentally and it exerts no essential influence on the amplification of signals whose envelope varies slowly in a time $t > 1/\Gamma$. However, amplification of pulses of duration $\tau < 1/\Gamma$ leads to a modification of their wave form, namely, to a certain lag of the leading front of the signal, and also to a fundamentally new phenomenon as stretching of the trailing edge over a time much longer than the duration of the initial pulse. It is as if the quantum system "remembered" all the parameters of the amplified pulse during a prolonged interval ($\sim 1/\Gamma$), which can be much longer than τ , until dephasing of the quantum-transition current takes place, and then emitted quanta coherent with this pulse even after this pulse has completely passed, in space, through the location of the active center. The same figure shows the result of the spatial amplification of a short pulse of duration $\tau = 2 \times 10^{-11}$ sec in a ruby amplifier. This result can easily be obtained from (4) if the input signal is represented in the form of two step-like signals with identical variation of the amplitude (in this case, of constant amplitude A_0), but of opposite polarity, shifted by a time τ . The delay of the leading front of the amplified pulse will compete, as the signal progresses along the amplifying medium, with the forward shift of the leading front of the signal as a result of the saturation phenomenon. These effects will predominate at very small and very large lengths of the amplifiers, although the concept of small and large length depends strongly in this case on the amplitude of the amplified signal.

For the Mössbauer effect with allowance for certain singularities of the nuclear isomers, the nuclear-transition linewidth can reach very small values, so that the transients will be very long and play an important role. Everything noted above concerning the singularities of the amplification of ultrashort optical pulses is fully applicable to the process of amplification of γ radiation, but all the characteristic times are longer by many orders of magnitude.

Using the expression for the generating function of the Bessel function, we can easily transform (4) to an equivalent form that is more convenient for calculation of the characteristics of the γ wave at $t < 1/\Gamma$:

$$A = A_0 e^{-x/2l} \exp \left[-\left(ie + \frac{\Gamma}{2} \right) \left(t - \frac{x}{c} \right) \right] \times \sum_{n=0}^{\infty} \left[\left(ie + \frac{\Gamma}{2} - \frac{1}{2T} \right) \left(\frac{t-x/c}{C_1 x} \right)^{1/2} \right]^n I_n \left(2 \left(C_1 x \left(t - \frac{x}{c} \right) \right)^{1/2} \right). \quad (5)$$

Figure 2 shows the spatial development of the γ wave, calculated from (5), for the real parameters $C_1 = 10^2 \text{ cm}^{-1} \text{ sec}^{-1}$, $\Gamma = 0.2 \text{ sec}^{-1}$, $T = 10^4 \text{ sec}$, $l = 0.1 \text{ cm}$, $\omega = 10^{18} \text{ sec}^{-1}$, $n_0 = 10^{23} \text{ cm}^{-3}$, and $f = 1$, at which (5) takes the simpler form

$$A = A_0 e^{-x/2l} \exp \left[-\left(ie + \frac{\Gamma}{2} \right) \left(t - \frac{x}{c} \right) \right] I_0 \left(2 \left(C_1 x \left(t - \frac{x}{c} \right) \right)^{1/2} \right).$$

It is seen from the figure that although the leading front of the γ wave moves with velocity c , the region of maximal radiation moves with a very low velocity $v = 4C_1 l^2 = 4 \text{ cm} \cdot \text{sec}^{-1}$.

Note should be taken of the following: the value $\Gamma = 0.2 \text{ sec}^{-1}$, taken by way of example, is as yet quite far from the experimentally obtained results for

Mössbauer isomers ($\Gamma \sim 10^5 \text{ sec}^{-1}$); however, as has been shown in a number of papers [1, 6-8], this value is perfectly obtainable in principle when the temperature is lowered and a radio-frequency field that restores the transition-line profile is applied. Moreover, it was possible, in an experiment on induced narrowing of the NMR line by applying a series of coherent high-frequency pulses in the radio band to the substance [9, 10], to suppress the dipole broadening, which is the main mechanism, and to ensure narrowing of the line to 10 sec^{-1} , which constitutes significant progress in this direction.

It should be noted that inasmuch as the establishment of the stationary cross section is decisively connected with the change of the spectral density of the amplified signal, owing to the change in its spectral width $\Delta\omega \sim 1/(t - x/c)$ at $t - x/c \lesssim 1/\Gamma$, the subsequent main part of the signal (i.e., $t - x/c > 1/\Gamma$), for which $\Delta\omega < \Gamma$, will interact with a medium characterized by a stationary cross section $\sigma = 2C_1/n_0(i\epsilon + \Gamma/2)$, since the effective equivalent cross section tends to the stationary value at $t - x/c > 1/\Gamma$. It follows therefore (leaving aside the peculiar behavior of the developing wave near its leading front $t - x/c < 1/\Gamma \ll T$), that the threshold condition for amplification on a strongly broadened transition is exactly the same as in the usual balance approach, namely $\sigma n_0 - 1/l > 0$.

2. CASE OF NONBROADENED LINE

We consider the development of a γ wave in an active medium consisting of a relatively short-lived nuclear isomer (i.e., with a lifetime $T \lesssim 10^{-6}$ sec), the emission (absorption) line of which has the natural width as a result of the Mössbauer effect, so that we have $\Gamma T = 1$ at a stable lower level. We make a number of assumptions when solving the system (1) for this case. It is seen from the foregoing calculations that, owing to the very small velocity of the maximum of the γ wave, it is perfectly permissible to neglect the retardation effect. In addition, we assume that because of the sufficiently small gain the change in the population of the working transition resulting from the amplification of the γ wave is small in comparison with the action of the spontaneous decay.

Then, assuming that the inverted population varies with time like $n = n_0(2\eta e^{-t/T} - 1)$, where η is the initial concentration of the excited nuclei, we obtain from (1)

$$\frac{\partial}{\partial t} \left(\frac{\partial A}{\partial x} + \frac{1}{2l} A \right) + \left(i\epsilon + \frac{\Gamma}{2} \right) \left(\frac{\partial A}{\partial x} + \frac{1}{2l} A \right) = C_1(2\eta e^{-t/T} - 1)A, \quad (6)$$

$$A(0, t) = A_0 e^{-t/2T}, \quad t > 0; \quad A(x, 0) = 0; \quad A(x \geq ct) = 0; \quad A = A_1 e^{-x/2l}.$$

Carrying out the Laplace-Carson transformation with respect to the variable x , we obtain for the resonant case $\epsilon = 0$

$$\bar{A}_1 = A_0 \exp \left[-\frac{2C_1\eta}{\Gamma a} (e^{-r_1} - 1) - \frac{C_1 t}{a} - \frac{\Gamma}{2} t \right].$$

Expanding the expression for \bar{A}_1 in Bessel functions and applying the inversion formula, we obtain

$$A = A_0 e^{-x/2l - r_1/2} I_0 \left\{ 2 \left(C_1 \left[\frac{2\eta}{\Gamma} (1 - e^{-r_1}) - t \right] x \right)^{1/2} \right\}. \quad (7)$$

Figure 3 shows the spatial evolution, calculated from (7), of a γ wave stemming from the spontaneous decay of nuclei situated in the surface layer, for the following real parameters: $C_1 = 6 \times 10^7 \text{ cm}^{-1} \text{ sec}^{-1}$, $\Gamma = 10^6 \text{ sec}^{-1}$, and $l = 0.1 \text{ cm}$. It is seen from the figure that the region of maximum radiation (maximum density of the γ wave)

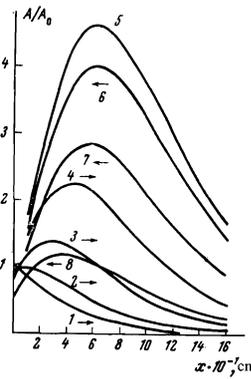


FIG. 3

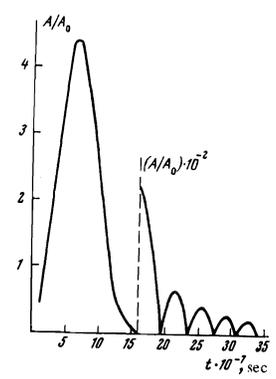


FIG. 4

FIG. 3. Spatial evolution of γ wave at $\Gamma T = 1$ for the following values of t : $1-0.6 \times 10^{-7}$, $2-10^{-7}$, $3-2 \times 10^{-7}$, $4-3.5 \times 10^{-7}$, $5-6.5 \times 10^{-7}$, $6-7 \times 10^{-7}$ sec, $7-10^{-6}$, $8-1.2 \times 10^{-6}$ sec.

FIG. 4. Temporal evolution of γ wave for the optimal value x_0 .

moves with a variable velocity $v = 4C_1 l^2 (2\eta e^{-\Gamma t} - 1)$ equal to $4C_1 l^2 (2\eta - 1)$ at the initial instant and decreasing to zero at the instant $t = \Gamma^{-1} \ln 2\eta$ (turning point), after which it reverses sign. The direction of motion of the maximum is shown by the arrows. The maximum value of A from (7), under the condition $r \gg 1$, when $I_0(r) \approx I_1(r)$, is reached for a sample of length

$$x_0 = \frac{C_1(2l)^2}{\Gamma} \left[2\eta - 1 - \frac{\Gamma}{4C_1 l} - \ln \left(\frac{2\eta}{1 + \Gamma/4C_1 l} \right) \right] \quad (8)$$

at the instant of time

$$t_0 = \frac{1}{\Gamma} \ln \left(\frac{2\eta}{1 + \Gamma/4C_1 l} \right), \quad (9)$$

which is always less than $T = 1/\Gamma$.

For real parameters of Mössbauer γ transitions, the optimal value of x_0 lies in the range $0.1-10 \text{ cm}$, i.e., is a perfectly acceptable quantity. For the foregoing parameters, which satisfy the condition $r \gg 1$, we have $x_0 = 0.7 \text{ cm}$, $t_0 = 6.5 \times 10^{-7} \text{ sec}$. Substituting (8) and (9) in (7), we obtain an expression for the maximum amplitude of the amplified wave at a given value of the inverted population and for the concrete nuclear parameters:

$$A = A_0(2\eta)^{-1/2} \exp[-2C_1 l T (2\eta - 1 - \ln 2\eta)] \cdot I_0[4C_1 l T (2\eta - 1 - \ln 2\eta)]. \quad (10)$$

If the argument of the Bessel function is small, $r \ll 1$, then the optimal values of x_0 and t_0 can be obtained from the equations

$$\frac{\Gamma}{2} = C_1 x_0 (2\eta e^{-r_1} - 1); \quad \frac{1}{2l} = C_1 \left[\frac{2\eta}{\Gamma} (1 - e^{-r_1}) - t_0 \right]. \quad (11)$$

Figure 4 shows the dependence of the amplitude of the γ wave on the time for the optimal value $x_0 = 0.7 \text{ cm}$. It appears that one of the essential features of quantum amplifiers operating in the transient regime is the possibility of temporal amplification of the signal, unlike the pure spatial amplification that predominates in the optical and microwave bands. The characteristic oscillations of the γ -wave amplitude are due to the response of the resonantly-absorbing medium to the decreasing γ radiation. They set in at the instant when the inverted population goes through zero as a result of spontaneous decay, and the modified Bessel function turns into the usual function. We note that a similar effect follows from Hamermesh's theory [11], which describes the absorption of γ quanta by a medium on the basis of a classical model of damped oscillations. It is easy to verify that all of Hamermesh's results [11] can be obtained from

formulas (4) and (5) following an obvious simplification of the latter.

The condition for amplification of the γ wave differs significantly from the usual balance condition and takes the form

$$\ln \left\{ I_0 \left(2 \left(C_1 \left[\frac{2\eta}{\Gamma} (1 - e^{-\Gamma t}) - t \right] x \right)^{1/2} \right) \right\} - \frac{\Gamma t}{2} - \frac{x}{2l} > 0. \quad (12)$$

By fixing the coordinate x , we can determine the interval of values of t at which temporal amplification of the input signal takes place. Analogously, by fixing t , we can find the interval of values of x at which the amplified signal exceeds the input signal, i.e., the range in which spatial amplification takes place.

It should be noted that the condition (12) imposes more stringent requirements on the parameters of the active medium than does the balance condition. This result has a simple physical explanation. In parallel with the process of establishment of the cross section, which leads to a lowering of the amplification threshold (to the balance threshold in the limit), there is a spontaneous decay of the excited nuclei, which leads to the opposite result. It is clear that the presence of such competing effects makes optimal a certain instant of time $t < T$ at which the value of the cross section is less than the stationary and the value of the inversion is less than the initial $n_0(2\eta - 1)$, so that the amplification threshold is in this case always higher than the idealized balance value.

We emphasize that for real parameters of short-lived γ transitions the threshold value of the inverted population exceeds the balance value by several times. For example, the method of obtaining the inverted state proposed by Gol'danskiĭ and Kagan^[2] makes it possible, according to the authors' estimate, to obtain an amplification effect on the unbroadened transition of Ta^{181} ($T = 6.8 \times 10^{-6}$ sec) on an inverted population $\xi = 0.1$ ^[2] corresponding to a value $\eta = 0.55$ ($n_0 = 3 \times 10^{19}$ cm⁻³). A simple calculation based on the foregoing theory shows that amplification at the data of^[2] is possible only if $\xi \gtrsim 0.4$ ($\eta \gtrsim 0.7$). For example, at $\xi = 0.6$ ($\eta = 0.8$), $C_1 = 7.6 \times 10^6$ cm⁻¹sec⁻¹ and $\Gamma = 1.47 \times 10^5$ sec⁻¹ the amplitude of the amplified wave for optimal values $x_0 = 0.6$ cm, $t_0 = 0.44$ and $T = 3.0 \times 10^{-6}$ sec exceeds the initial amplitude of the input signal A_0 by a factor 1.3, and the amplitude at $t = t_0$ by a factor 2.1. On the other hand, an estimate assuming a constant resonant cross section for Ta^{181} , namely $\sigma = 1.7 \times 10^{-18}$ cm²^[12], and assuming $\xi = 0.6$ and $x = x_0 = 0.6$ cm leads to an excess of the amplified signal over the input signal, using the customarily employed balance relations, of more than 10^3 times. Analogously, at $\xi = 0.8$ ($\eta = 0.9$) we have $x_0 = 1$ cm, $t_0 = 3.7 \times 10^{-6}$ sec, and the amplitude of the amplified γ wave exceeds the initial input amplitude by four times and the input amplitude at $t = t_0$ by 7 times, whereas a control calculation for the balance case and for a constant cross section at $x = x_0$ yields an amplification of 10^6 times at $t \ll T$.

The foregoing numerical estimates, in spite of their somewhat idealized character (neglect of the saturation effect), confirm the essential influence of the transient phenomena on the kinetic processes using natural-width lines, and shows that the mechanism of excitation of the inverted state of short-lived nuclei should be more effective than hitherto supposed. In addition, the rather

small amplification of the γ wave justifies the approximation used in the present paper, wherein the population of the working transition was assumed to depend little on the kinetics of the process.

CONCLUSION

The considered singularities of the kinetics of induced γ radiation make it possible to examine the problem of development of a γ laser more realistically. The results offer evidence that the evolution of the stationary resonant cross section occurs within a time exceeding $1/\Gamma$. Starting from this, we see clearly the singularities of both cases considered above. The spatial amplification curve in the case of a strongly broadened line becomes identical, after a certain time during which no essential change takes place in the population of the working transition, with the exponential curve obtained from the balance relations, but the value of the cross section is in this case, as is well known, less than the resonant value by the broadening factor ΓT . It is therefore obvious that the threshold value of the inverted population for the case of a strongly broadened line corresponds to the usual balance relation. In the case of amplification that proceeds with the use of a nonbroadened line, the limiting value of the cross section is equal to the resonant value, but spontaneous decay of this state takes place during the evolution of this cross section, and the radiation maximum is obtained at $t_0 < T$, i.e., at a cross section far from the stationary resonant value. The threshold condition then takes the form (12), which differs strongly from the balance condition.

The case of the nonbroadened line, as follows from the foregoing, leads to the concept of the optimal thickness x_0 of the active layer. It is clear that at a small value of x_0 it is no longer necessary to produce a γ resonator to increase the effective thickness of the active layer. Therefore the study of the problem of the γ laser on short-lived γ transitions must be limited in a number of cases to the traveling-wave regime.

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