

# Two-photon laser amplifiers

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Optical amplification by multiphoton transitions is considered theoretically. The threshold conditions, the shape of amplified pulse, the efficiency, and the gain are obtained for a standing wave amplifier when the driving laser operates in the  $Q$ -switched or free-running regime. Amplification of a running light wave in a nonlinearly amplifying medium is analyzed on basis of the kinetic equations. The possibility of "igniting" an inverse-populated extended medium by a narrow laser beam at half-frequency is demonstrated qualitatively.

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The necessary condition for the generation of coherent radiation is the inversion of the populations of the phototransition levels. However, this condition is far from sufficient for realization of an actual laser, which calls also for high gains  $\alpha$ : for a number of reasons it is practically impossible to generate coherent emission at  $\alpha \lesssim 10^{-4} \text{ cm}^{-1}$ . On the other hand, in the study of systems that are promising for laser development, one encounters with greater frequency cases in which, at appreciable attainable per-unit values of the population inversion of the working levels, an estimate of the gain yields values  $\sim 10^{-4} \text{ cm}^{-1}$  and lower. Examples are the suggested photorecombination lasers,  $\text{H}_2$  ortho-para conversion lasers,<sup>[4]</sup> IR lasers using  $\text{H}_2$  and  $\text{N}_2$ ,<sup>[5]</sup> and others.

The smallness of the gain at appreciable inversion levels is due to the low values of the stimulated-emission cross section  $\sigma$ , which in turn is the consequence of either a forbidden transition or of the width of the emission spectrum (the large number of sublevels). From this point of view, it is of interest to analyze the possibility of converting large per-unit energies that are inversely stored in the medium, and the stimulated emission on many-photon and particularly two-photon transitions. The selection rules for such transitions differ from the selection rules for single-photon transitions, and they may turn out to be more allowed.<sup>[6]</sup> The presence of a large number of closely lying sublevels also contributes to an increase of the cross section  $\beta_k$  of a  $k$ -photon transition ( $k \geq 2$ ).<sup>[7]</sup>

Two-frequency lasers have been proposed quite some time ago, a particular case of which (at  $\omega_1 = \omega_2$ ) are two-photon lasers.<sup>[8]</sup> Most recently, an attempt was made to consider the self-excitation regime of a multiphoton laser.<sup>[9]</sup> However, in view of the dependence of the gain in the multiphoton transition on the light intensity,  $\alpha \sim I^{k-1}$ , the development of spontaneous-emission lasing should be greatly hindered in such lasers. Practical interest attaches therefore to amplification regimes in which a sufficiently high intensity at the frequency of the induced emission is produced by some external source (usually a laser).

We consider below in the main two-photon laser amplifiers, since these are the easiest to realize. The main results are qualitatively valid also for amplifiers of higher phototransition order.

## 1. STANDING WAVE LASER AMPLIFIER (SWA)

It is simplest to eliminate effectively the inversion of two-photon transitions by induced emission with the aid of a resonator laser amplifier; one such amplifier is illustrated in Fig. 1. A cell with inversely-excited (relative to the transition with  $\hbar\omega_2 = 2\hbar\omega_1$ ) medium 2 is placed in the resonator of an ordinary laser 1 (operating at the frequency  $\omega_1$ ). Without loss of generality, we can assume that the phototransition in the medium 2 is sufficiently forbidden and we can neglect all the inversion-relaxation processes ( $\Delta N_2, \text{ cm}^{-3}$ ) during the lasing times. If the medium 2 is "single-photon" transparent to radiation of frequency  $\omega_1$ , then initially the generation of the laser 1 develops in accordance with the same laws as in the absence of cell 2. However, starting with a certain field intensity  $I$ , an effective induced radiation of the energy inversely stored in the medium 2 can take place in the resonator (in view of the linear dependence of the gain  $\alpha$  of the two-photon transition on  $I$ ). To find the critical conditions for the onset of effective amplification and to determine the efficiency, the peak power, and the temporal characteristics of the radiation amplification, we shall analyze the two-photon SWA on the basis of the balance equations usually employed for the simplified laser model (i. e., assuming a small change of the traveling-light-wave intensity per path).

It is of interest in this case to consider two operating regimes of the exciting laser 1—the  $Q$ -switched regime and the free-running regime.

A.  $Q$ -switched regime. In this case we can regard

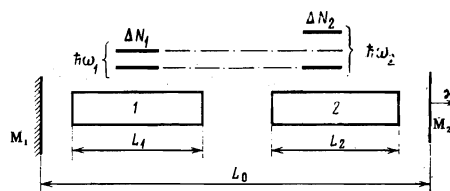


FIG. 1. Schematic diagram of two-photon laser with coherent excitation: 1—exciting laser with emission frequency  $\omega_1$  (its length is  $L_1$ ); 2—medium (of length  $L_2$ ) with population inversion on a single-photon forbidden transition with frequency  $\omega_2 = 2\omega_1$ ;  $M_1$  and  $M_2$ —resonator mirrors;  $x = 1 - R$  is the coefficient of the active losses of the resonator;  $L_0$  is the resonator base.

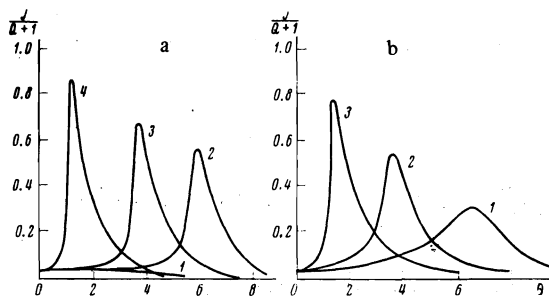


FIG. 2. Time evolution of the lasing in a two-photon laser with coherent excitation ( $j(\theta)/(Q+1)$ —relative intensity of the radiation,  $\theta$ —dimensionless time,  $Q^{-1}=0.03$ ): a)  $Q$ -switched regime of exciting lasers, curves 1, 2, 3, and 4 correspond to  $\Gamma_2 = 1.0, 1.05, 1.11,$  and  $1.2$ ; b) free-running regime of exciting laser, curves 1, 2, and 3 correspond to  $\Gamma_2 = 0.4, 0.5,$  and  $1.0$ .

the initial inversion  $\Delta N_1^0$  ( $\text{cm}^{-3}$ ) as given for medium 1, and the balance equations take the form

$$\frac{dj}{d\theta} = \Gamma_1 n j - j + \Gamma_2 m j^2, \quad \frac{dn}{d\theta} = -\Gamma_1 n j, \quad \frac{dm}{d\theta} = -\frac{\Gamma_2}{Q} m j^2 \quad (1)$$

with the initial conditions at  $\theta = 0$  (the  $Q$ -switching instant)

$$j(0) \ll 1, \quad n(0) = 1, \quad m(0) = 1.$$

Here  $j$ ,  $n$ , and  $m$  are the dimensionless emission intensity and the inverted populations in media 1 and 2, respectively

$$j = I \left[ \frac{L_1 \Delta N_1^0 c}{L_0} \right]^{-1}, \quad n = \frac{\Delta N_1}{\Delta N_1^0}, \quad m = \frac{\Delta N_2}{\Delta N_2^0},$$

$\theta = t/\tau_c$  is the dimensionless time, where  $\tau_c = 2L/(1-R)c$  is the characteristic time of the optical resonator. The parameters  $\Gamma_1$ ,  $\Gamma_2$  and  $Q$  in the system (1) are equal to

$$\Gamma_1 = \sigma c \tau_c \Delta N_1^0 \frac{L_1}{L_0}, \quad \Gamma_2 = 2\beta_2 c \tau_c \Delta N_2^0 \frac{L_2 \Delta N_1^0 c}{L_0} \frac{L_1}{L_0}, \quad Q = 2 \frac{\Delta N_2^0 L_2}{\Delta N_1^0 L_1}$$

and have the following physical meaning.

The parameter  $\Gamma_1 = \alpha_1^0/(1-R)$  is the ratio of the initial gain  $\alpha_1^0$  per round trip to the loss coefficient  $(1-R)$  referred to one mirror (including the passive losses). The lasing condition for laser 1 is  $\Gamma_1 > 1$ ; in the case of interest to us, that of a high-intensity exciting laser, we have  $\Gamma_1 \gg 1$ .

The parameter  $\Gamma_2 = \alpha_2^0/(1-R)$  is the ratio of the gain  $\alpha_2^0$  of the medium 2 (corresponding to the peak value of the intensity  $I_1^0 = \Delta N_1^0 c / 2\tau_c$  of laser 1 at  $\Gamma_1 \gg 1$ ) to the loss factor  $(1-R)$ . In order for the radiation to effectively remove the inversion on the two-photon transition it is necessary to satisfy the condition  $\Gamma_2 > 1$ .

The parameter  $Q$  is the ratio of the inversely stored energies in the media 2 and 1. As follows from (1), in this problem  $Q$  is the quality factor of the system; the

maximum SWA gain, for example, is equal to  $Q+1$ . In the effective-amplification cases of practical interest we have  $Q \gg 1$ .

If the condition  $\Gamma_1 \gg 1$  is satisfied, i.e., if the laser 1 operates high above the threshold, the lasing sets in so rapidly (the rise time is  $\sim \tau_c/\Gamma_1$ ) that the intensity of the coherent radiation reaches its maximum value (in the absence of medium 2) before the nonlinear interaction<sup>1)</sup> of the laser light with the medium can manifest itself to any considerable degree. For that instant of time, the system (1) takes the form (if the dimensionless time  $\theta$  is reckoned from the instant when the laser 1 reaches peak power).

$$\frac{dj}{d\theta} = -j + \Gamma_2 m j^2, \quad \frac{dm}{d\theta} = -\frac{\Gamma_2}{Q} m j^2 \quad (2)$$

with initial conditions

$$m(0) = 1, \quad j(0) = 1.$$

It follows from the system (2) that gain can arise only in the case when  $dj/d\theta|_{\theta=0} > 0$ , i.e., at  $\Gamma_2 > 1$ . Analysis shows that in view of the nonlinear character of the amplification of the light, small variations of  $\Gamma_2$  ( $\delta\Gamma_2 \ll Q^{-1}$ ) near  $\Gamma_2 = 1$ , or more accurately

$$\Gamma_2^{\text{thr}} = 1 + Q^{-1}, \quad (3)$$

lead to appreciable changes of the attained peak intensity  $j_{\text{max}}$  of the radiation, of the SWA gain. In other words, the amplification has a threshold, the condition for which is given by (3). In analogy with ordinary (linear) lasers, we can call the proposed SWA a two-photon laser with coherent excitation (in contrast to the two-photon laser with self-excitation considered in<sup>[9]</sup>, the threshold of which is reached naturally at exceedingly large values of  $\Gamma_2$ ).

Figures 2a and 3a show by way of example the results of a numerical solution of the system (2) at the following values of the parameters:  $Q^{-1} = 0.03$  (threshold value  $\Gamma_2^{\text{thr}} = 1.03$ ),  $\Gamma_2 = 1.00, 1.05, 1.11,$  and  $2.00$ . Figure 2a shows the time dependences of the coherent-radiation intensity, normalized to the maximum possible intensity  $I_{\text{max}}$  (corresponding to the instantaneous transition and emission of all the energy inversely

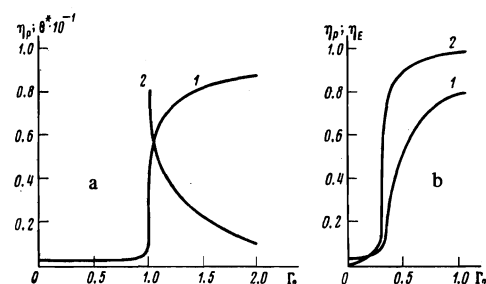


FIG. 3. Dependence of the relative power gain  $\eta_P \equiv j_{\text{max}}/(Q+1)$  of a two-photon SWA (curves 1), of the time  $\theta^*$  needed to reach peak power (curve 2), and of the energy efficiency  $\eta_E$  (curve 2 of Fig. 2b) on the critical parameter  $\Gamma_2$  at  $Q^{-1} = 0.03$ . a— $Q$ -switched regime, b—free-running regime.

stored in the medium 2;  $I_{\max} = \Delta N_2^0 c L_2 / L_0$ , i. e., of the function  $j(\theta)/(Q+1)$ . The plots of  $\eta_P \equiv j_{\max}/(Q+1)$  and  $\theta^*$  (the time required to reach peak power) against the parameter  $\Gamma_2$  are shown in Fig. 3a. It is seen that even at a small excess of the threshold, the SWA gain  $j_{\max}$  is close to its asymptotic value  $Q+1$  ( $\eta_P = 1$ ); an even faster increase occurs in the energy efficiency

$$\eta_E = \frac{1}{Q} \int_0^\infty j d\theta$$

on going through the threshold value, for all three curves of Fig. 2a corresponding to  $\Gamma_2 > 1$  and  $\eta_E \approx 1.0$ .

**B. Free-running regime.** In this case we can assume the rate  $q$  of the pumping (of the inversion) of laser 1 to be specified. If we put  $\Delta N_1^0 = q\tau_c$ , then, in the same notation as before, we arrive at the system of the balance equations

$$\frac{dj}{d\theta} = \Gamma_1 n j - j + \Gamma_2 m j^2, \quad \frac{dn}{d\theta} = 1 - \Gamma_1 n j, \quad \frac{dm}{d\theta} = -\frac{\Gamma_2}{Q} m j^2 \quad (4)$$

with initial conditions

$$n(0) = 0, \quad m(0) = 1, \quad j(0) \ll 1.$$

At large values of  $\Gamma_1$ , after a time  $\sim \tau_c/\Gamma_1$ , the population inversion of laser 1 becomes quasistationary,  $dn/d\theta \approx 0$ , i. e.,  $n j \approx \Gamma_1^{-1}$ . Starting with this instant of time, the system (4) can be written in the form

$$\frac{dj}{d\theta} = 1 - j + \Gamma_2 m j^2, \quad \frac{dm}{d\theta} = -\frac{\Gamma_2}{\theta} m j^2 \quad (5)$$

with initial conditions

$$m(0) = 1, \quad j(0) \ll 1.$$

As follows from the system (5), in the free-running regime of laser 1, at  $Q \gg 1$ , an excess of the parameter  $\Gamma_2$  above a certain critical value  $\Gamma_2^{\text{thr}}$  also leads to a sharp increase of the intensity of the induced radiation. The value  $\Gamma_2^{\text{thr}}(Q \rightarrow \infty)$  can be obtained from the condition that the right-hand side of the first equation of (5) be a minimum at  $m = 1$ ; this yields  $\Gamma_2^{\text{thr}} = \frac{1}{4}$ .

It should be noted that in this case  $\Gamma_2^{\text{thr}}$  depends more strongly on  $Q$  (for example,  $\Gamma_2^{\text{thr}} = 0.33$  at  $Q = 0.03$ ), and the transition region is broader than for the  $Q$ -switching regime. Fig. 2b shows the dependence of the relative intensity  $j(\theta)/(Q+1)$  as a function of the dimensionless time  $\theta$  for parameter values  $\Gamma_2 = 0.4, 0.5$ , and  $1.0$ , while Fig. 3b shows the dependence of the relative power gain  $\eta_P \equiv j_{\max}/(Q+1)$  and of the maximum amplifier efficiency  $\eta_E \equiv [(1-m)/Q]_{\max}$  on  $\Gamma_2$ .

The foregoing calculation was performed under the assumption that the laser 1 is continuously pumped. It is of interest to assess the instant of pump-interruption time at which the SWA efficiency is still not substantially decreased. From a comparison of the systems (2) and (5) we see that at  $\Gamma_2 \gtrsim 1$  the pumping in the free-running regime can be turned off at the instant when  $j \approx 1$  (see Fig. 2b); the subsequent evolution of the radi-

ation will be described in this case by the curves of Fig. 2a at the corresponding values of the parameters. On the other hand, if  $\frac{1}{4} < \Gamma_2 < 1$ , then to attain the effective-amplification regime the pump must continue until much higher  $j$  are reached (practically to  $j_{\max}$ ). Another important question is to ascertain in which of the regimes of laser 1 it is easiest to obtain two-photon generation (effective amplification). At the same value of the pump we have for the two regimes of laser 1:

$$\Delta N_1^0 = \xi q \tau_r, \quad Q\text{-switching,}$$

$$\Delta N_1^0 = q \tau_c, \quad \text{free running,}$$

where  $\tau_r$  is the characteristic relaxation time of the inversion in the medium 1, and  $\xi$  is the efficiency of energy conversion in the case of  $Q$  switching (usually  $\xi = 3.3-0.1$ ). It follows therefore that at  $Q \gg 1$  the  $Q$ -switching regime is more effective if the condition  $\tau_r/\tau_c > 4/\xi$  is satisfied.

In conclusion, let us dwell on the practical realization of two-photon-transition SWA. Effective amplification at  $\Gamma_2 > 1$  and  $Q \gtrsim 3$  leads, in the case of relatively easily realized parameters of the apparatus,  $L_2/L_0 \approx 0.75$  and  $\tau_c = 3 \times 10^{-8}$  sec, to the inequalities

$$\beta_2 \Delta N_2^0 I_1^0 > 10^{-3} \text{ cm}^{-1}, \\ \Delta N_2^0 / I_1^0 > 10^{-10} \text{ cm}^{-1} \cdot \text{sec}.$$

There have been very few studies so far of the two-photon transition cross sections  $\beta_2$ . For strong two-photon transitions, for example, for the Doppler-broadened Na (3S-4D) line we have  $\beta_2 \approx 10^{-41} \text{ cm}^4 \text{ sec}^{[10]}$ ; for the weakest of the investigated transitions in broad spectra of aromatic compounds, the cross sections  $\beta_2$  are of the order of  $10^{-49}-10^{-50} \text{ cm}^4 \text{ sec}^{[11]}$ .

Figure 4 shows schematically the regions of two-photon generation plotted in the coordinates  $\beta_2$  and  $I_1^0$  for different values of the inversion in the medium 2. It is seen that were we to find substances with two-photon transition cross sections  $10^{-46}-10^{-47} \text{ cm}^4 \text{ sec}$ , then two-photon lasing with coherent excitation would be perfectly feasible. Under these conditions, the intensity of the driving (single-photon) laser does not exceed  $10^8 \text{ W/cm}^2$ , and the intensity of the light pulse amplified by the two-photon medium (at  $Q = 33$ , see Fig. 2) is  $\lesssim 3 \times 10^9 \text{ W/cm}^2$ , which is much less than the self-focusing limit.

The following two remarks are in order here:

1. In the case of broad molecular spectra, characterized by small values of  $\beta_2$ , it is possible to employ

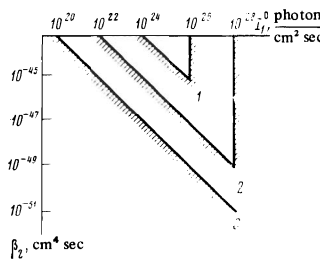


FIG. 4. Regions of intense two-photon generation under initial inversion:  $10^{16}$  (1)  $10^{18}$  (2), and  $10^{20} \text{ cm}^{-3}$  (3).

a laser of fixed frequency (for example, a neodymium or ruby laser) as the exciting source.

2. For atoms and molecules in the gas phase, in two-photon absorption in a standing wave (and consequently in the corresponding radiative transition), the Doppler broadened line of the transition narrows down to a value determined by the collisions, meaning a proportional increase of the transition cross section at the maximum of the line. This phenomenon has been under intensive study of late.<sup>[10,12,13]</sup> In other words, up to a total concentration  $\sim 10^{18}$  cm<sup>-3</sup> (at which the Doppler and collision broadenings become comparable) we can use for estimates  $\tilde{\beta}_2^{s.w} \Delta N_2^0 \approx \text{const}$ , where  $\tilde{\beta}_2^{s.w}$  is the cross section of the transition for the standing wave ( $\beta_2^{s.w} > \beta_2$ ). Therefore in the gas phase (when the condition  $Q \gg 1$  is satisfied), two-photon lasing can be obtained in principle at low pressures in medium 2 and at low radiation intensities of laser 1.

## 2. TWO-PHOTON TRAVELING-WAVE AMPLIFIER (TWA)

Just as for a single-photon amplifying medium, large stimulated-emission intensities can be obtained also with amplification of a traveling wave of frequency  $\omega = \omega_2/2$ . In this case the multiphoton character of the transition leads, however, to a number of singularities, unusual for linear amplifiers, in the propagation of the light pulse through the amplifying medium.

For the purpose of comparison, let us list briefly the results of the analysis of the amplification regime in the single-quantum case on the basis of the kinetic equations.<sup>2)</sup> The motion of the light pulse through a medium with specified initial inversion  $\Delta N_1^0$  and with a transition cross section  $\sigma$  is described, in the absence of losses, by the system of equations<sup>[15,16]</sup>.

$$\frac{\partial I}{\partial x} + \frac{1}{c} \frac{\partial I}{\partial t} = \sigma \Delta N_1 I, \quad \frac{\partial \Delta N_1}{\partial t} = -2\sigma \Delta N_1 I. \quad (6)$$

The system (6) can be solved analytically<sup>[15]</sup>; in the particular case of propagation of an initially rectangular light pulse the intensity of the coherent radiation at the point  $x$  and at the instant of time  $t > x/c$  takes the form

$$I = I_0 \exp\{\sigma \Delta N_1^0 x\} [\exp\{\sigma \Delta N_1^0 x\} + (1 - \exp\{\sigma \Delta N_1^0 x\}) \exp\{2\sigma I_0(x - ct)\}]^{-1}.$$

It follows therefore that the peak power of the radiation is reached at the initial section of the pulse  $t = x/c$ , and increases exponentially with increasing length of the amplifier. The subsequent part of the pulse propagates in the depleted medium and is less amplified, so that the pulse leaving the amplifier is shorter if the amplifier length is increased.

Under the assumptions made above, the propagation of a light pulse through a two-photon-amplifying medium is described by the following system of equations for the relative intensity  $j = I/\Delta N_2^0 c$  and the relative inversion  $n \equiv \Delta N_2/\Delta N_2^0$ :

$$c \frac{\partial j}{\partial x} + \frac{\partial j}{\partial t} = \frac{1}{\tau} n j^2, \quad \frac{\partial n}{\partial t} = -\frac{1}{\tau} n j^2, \quad (7)$$

where the characteristic time is  $\tau = [2\beta_2(\Delta N_2^0 c)^2]^{-1}$ .

The system (7) can in this case not be solved by the method of<sup>[15]</sup>, since this method makes use in essence the linearity of the equations in  $I$ . Nonetheless, as will be shown below, the linear problem can also be reduced to a solution of an ordinary differential equation.

All the physical processes in the medium start with the instant of the arrival of the light pulse at the considered point  $x$  ( $t = x/c$ ). It is also convenient to change over to dimensionless distance and dimensionless time, defined by the relations

$$s = \frac{x}{c\tau}, \quad \theta = \frac{t - x/c}{\tau}.$$

In terms of the indicated coordinates, the system (7) can be transformed into

$$\frac{\partial j}{\partial s} = \frac{\partial^2}{\partial s \partial \theta} (s \eta_E), \quad \frac{\partial j}{\partial s} = j^2 \frac{\partial}{\partial s} (1 - \eta_E) s. \quad (8)$$

where

$$\eta_E = \frac{1}{s} \int_0^s [1 - n(s', \theta)] ds'$$

is the energy efficiency of the amplifier, i.e., the fraction of the total energy  $E = \frac{1}{2} \Delta N_2^0 \hbar \omega_2$ , of the induced radiation by the instant of time  $\theta$ .

From the system (8) it follows that

$$\frac{\partial \eta_E}{\partial \theta} = \frac{1 - \eta_E}{Q[Q - s(1 - \eta_E)]}, \quad (9)$$

where  $Q(\theta) \equiv \Delta N_2^0 c / I_0(\theta\tau)$ ;  $I_0(t)$  is the intensity of the initiating pulse at the point  $x = 0$ .

An analysis of Eq. (9) in the case of a rectangular pump pulse ( $Q = \text{const}$ ) shows that in an amplifier of "length"  $s < Q$  the initial condition  $\eta_E(s, 0) = 0$  is valid, and the solution constitutes a white pulse that increases in amplitude and shortens in duration, determined by the expression

$$\theta = Q^2 \left[ -\ln(1 - \eta_E) - \frac{s}{Q} \eta_E \right].$$

However, in view of the nonlinear character of the amplification in the two-photon system, the idealized rectangular wave form of the initial leads, starting with the critical TWA length  $s_{cr} = Q$ , to a physically incorrect solution,<sup>3)</sup> when the leading part of the pulse has the form of a  $\delta$  function (this is equivalent to the initial condition  $\eta_E(s, 0) = 1 - Q/s$ ,  $s \geq Q$ ). Thus, in contrast to linear quantum amplifiers, for which the rectangular input pulse approximation is valid up to large incident-signal gains, for two-photon amplifiers this approximation becomes invalid already at an energy gain value

$$K_E = \eta_E \frac{Q(\theta_{\text{put}})}{\theta_{\text{put}}} s \approx \frac{2}{\eta_E}.$$

In other words, for the case of large gains, which is of practical interest, the approximation in which the

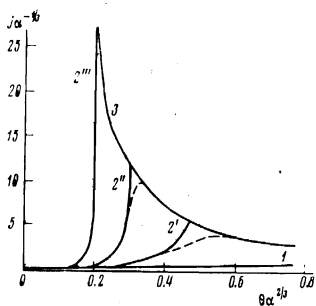


FIG. 5. Evolution of a radiation pulse moving through a two-photon amplifying medium: line 1—pulse at the amplifier ( $l = 0$ ), curves 2', 2'', 2'''—leading front of the pulse (Eq. (12)) at the relative lengths  $l = 2.0, 3.33$ , and  $5.0$ ; curve 3—trailing edge of the pulse (Eq. (13), second approximation). The dashed lines are the exact numerical solution of Eq. (11) at the indicated values of the parameter  $l$ .

initial pulse is rectangular is valid only far from saturation of the amplifying medium,  $\eta_E \ll 1$ . In the case of the "saturated" amplifier operating regime, on the other hand, when an appreciable fraction of the energy goes over into stimulated emission ( $\eta_E \sim 1$ ), the finite slope of the exciting pulse must be taken into account.

For the calculation it would be possible to use as before Eq. (9) for  $\eta_E$ , assuming  $Q$  to be a given function of  $\theta$ . In the saturation regime, however, it is of greatest interest to investigate the wave form of the amplified signal and its intensity, i. e., to solve the problem for the function  $j(\theta)$ .

As follows from (8)

$$j^{-1} - Q = (1 - \eta_E)s \quad (10)$$

and the equation for the reduced intensity  $j$  takes the form

$$\frac{\partial j}{\partial \theta} = j^2 \left[ Q^{-1} - \frac{dQ}{d\theta} - j \right]. \quad (11)$$

Eq. (11) has a singularity at the origin ( $\theta = 0$ ); the behavior of the function  $j(\theta)$  in the vicinity of this point is described by expression (10) with  $\eta_E = 0$ , i. e.,

$$j^{-1}(\theta) - Q(\theta) = -s, \quad \theta \rightarrow 0. \quad (12)$$

For large values of  $\theta$ , Eq. (11) leads to an asymptotic expression for the intensity

$$j = j^{(1)} + j^{(2)} + \dots, \quad j^{(1)} = Q^{-1} - \frac{dQ}{d\theta}, \quad (13)$$

$$j^{(2)} = [j^{(1)}]^{-2} \frac{dj^{(1)}}{d\theta}, \dots$$

It is important that the asymptotic expression (13) for the trailing edge of the pulse does not contain the distance  $s$  that the pulse has traversed in the medium.

In first-order approximation, the waveform of the pulse is determined by expressions (12) ( $\theta < \theta^*$ , leading front), and (13) ( $\theta > \theta^*$ , trailing edge), joined together at the point  $\theta^*$  obtained from the condition

$$\frac{1}{Q(\theta^*) - s} = Q^{-1}(\theta^*) - \frac{dQ(\theta^*)}{d\theta}.$$

Figure 5 shows the approximate expressions (12) and (13) as well as the exact numerical solutions of (11) for an exciting pulse that increases linearly with time

$$Q^{-1} = \alpha\theta, \quad \alpha = \tau \frac{dI_0}{dt} / \Delta N_2^0 c.$$

It can be shown (see Fig. 5) the condition for effective operation of the amplifier is  $l = s\alpha^{1/3} > 1$ ; with increasing relative length  $l$  of the amplifier, the accuracy of the approximations (12) and (13) increases, and at  $l \approx 5$  the approximate solution practically coincides with the exact one. At large  $l$ , simple analytic expressions are obtained for the maximum intensity of the stimulated emission  $j_{\max} \approx l^2 \alpha^{1/3}$ , the time  $\theta^* \approx l^{-1} \alpha^{-2/3}$  required to reach maximum intensity, the amplifier efficiency  $\eta_E \approx 1 - \theta^*/\theta_{pu1}$  at an exciting-pulse duration  $\theta_{pu1}$  and at an energy gain

$$K_E \approx \eta_E \frac{2Q(\theta_{pu1})s}{\theta_{pu1}} \approx 2\eta_E(1 - \eta_E)^2 l^2.$$

The relative length  $l$  of the amplifier, in analogy with the single-photon amplifier, can be represented in the form

$$l = \sigma_{\text{eff}} \Delta N_2^0 L_{\text{ampl}}, \quad (14)$$

where

$$\sigma_{\text{eff}} = \frac{dI_0}{dt} \left[ 2\beta_2 / \frac{dI_0}{dt} \right]^{1/2}.$$

For a giant laser pulse with typical spike duration  $3 \times 10^{-8}$  sec and a pulse energy density  $10 \text{ J/cm}^2$ , the slope is  $dI_0/dt \approx 10^{35}$  photons/cm<sup>2</sup> sec<sup>2</sup>. In this case, for values  $\beta_2 = 10^{-47}$  and  $10^{-4} \text{ cm}^4/\text{sec}$  we have  $\sigma_{\text{eff}}$  equal respectively to  $3 \times 10^{-20}$  and  $3 \times 10^{-18} \text{ cm}^2$ . For an amplifier with  $L_{\text{ampl}} \sim 1 \text{ m}$ , the necessary inversion densities are<sup>4)</sup> (at  $l = 3$ )  $\Delta N_2^0 = 10^{18}$  and  $10^{16} \text{ cm}^{-3}$ . These estimates show that the conditions for attaining lasing in a SWA and effective amplification in a traveling wave amplifier are close to each other.

### 3. LASER "IGNITION" OF TWO-PHOTON MEDIUM WITH INVERSION

The development of lasing on multiphoton transitions from spontaneous emission (in view of the time dependence of the gain on the emission intensity) is hardly realizable; at the same time, coherent excitation calls for high intensities of the resonant radiation ( $I > I^{\text{thr}}$ , see Sec. 1), which are difficult to achieve over appreciable areas. Great interest attaches therefore to the problem of "ignition" of an extended two-photon medium by a narrow powerful-laser beam with frequency  $\omega = \omega_2/2$  via the diffraction divergence of the developing stimulated emission or under conditions of an optical resonator with a weakly diverging standing wave. This approach is particularly attractive under conditions when  $I$  does not exceed  $I^{\text{thr}}$  greatly, i. e.,  $Q \sim 1$ . In this case a simple SWA would lead to an in-

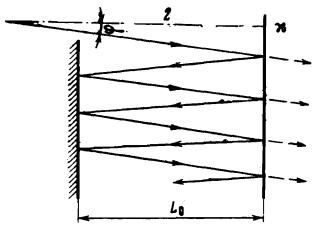


FIG. 6.

significant enhancement of the intensity of the incident light, while "ignition" makes it possible to obtain an intensity to the intensity of the driving laser, but with much higher energy (by a factor equal to the ratio of the cross section areas of medium 2 and the exciting laser beam). A detailed analysis of the problem of the broadening of a laser beam in a nonlinear medium, especially with allowance for its diffraction divergence entails considerable difficulty of both fundamental and mathematical character. We therefore confine ourselves here only to qualitative estimates for the simple model shown schematically in Fig. 6. Let the medium 2 (inversely populated on a transition with  $\Delta E = \hbar\omega_2$ ) be placed in a planar optical resonator with transmission  $\kappa = (1 - R)$ ; a laser beam of frequency  $\omega = \omega_2/2$  (with characteristic transverse dimension  $\Delta$ ) is incident on the medium through an opening in the resonator at an angle  $\varphi$  to its axis. If the intensity of the incident beam exceeds  $I_{thr}$ , then at  $\varphi = 0$  the lasing on the two-photon transition will cease after a time  $\sim \tau_c$ . It is obvious that if the parallel displacement of the beam in the resonator during this time is shorter than its transverse dimensions, then as the lasing advances it will subtend over the entire volume of the resonator. An estimate of the angle  $\varphi$  yields  $\varphi \approx (1 - R)\Delta/4L$ , which corresponds to  $\varphi \approx 10^{-4}$  at  $\Delta \approx 1$  cm,  $(1 - R) \approx (3-5)\%$ , and  $L_0 = 1$  m. On the other hand, at  $\lambda \approx 1 \mu$  the diffraction divergence of the laser beam is  $\varphi_{difr} \approx \lambda/\Delta \approx 10^{-4}$ . This means that in practice the "ignition" of the medium can be realized as a result of the natural divergence of a normally-incident laser beam.

#### 4. CONCLUSION

We have considered here different excitation regimes of intense coherent radiation on two-photon transitions with population inversion with the aid of an external laser source of light of frequency  $\omega_1 = \omega_2/2$ . Our calculations have shown the following:

1. The amplification process in a two-photon SWA has a threshold. The threshold intensity of the exciting laser is proportional to the two-photon absorption cross section  $\beta_2$ , to the per-unit population inversion in the two-photon medium  $\Delta N_2^0$ , and to the intensity of the exciting laser. The threshold behavior is most clearly pronounced when the exciting laser is Q-switched, and an excess of 2% above threshold ensures complete elimination of the inversion  $\Delta N_2^0$  by the induced radiation.

2. The essentially nonlinear character of the amplification of the stimulated emission on the two-photon transition leads to a number of distinguishing features of the propagation of the pulse through the amplifying

medium in the SWA. Thus, for example, the idealization wherein the input pulse is rectangular, which is frequently used for ordinary SWA, leads in this case to physically incorrect results even during the initial stages of the amplification. An approximate analytic solution is obtained for the propagation of a pulse in a two-photon SWA. Using as an example an input pulse that increases linearly with time, expressions are obtained for the criterion of effective amplification, for the gain, for the efficiency, and for the evolution of the pulse shape as it moves through the medium.

An extended medium with inversely populated two-photon transition levels can be "ignited" by a narrow laser pulse of frequency  $\omega = \omega_2/2$ . The entire medium can go over into the regime of intense induced transition already at relatively small excesses of the light intensity of the exciting laser above threshold. The considered regime is particularly attractive at low gains, when the increase of the total energy of the coherent radiation is due mainly to the increase in the cross section area of the emerging laser beam.

The performed numerical estimates show that the development of high-efficiency two-photon-transition quantum amplifiers is perfectly realistic. The active media for such amplifiers can be organic glasses with impurity molecules whose lower triplet level energy is close to  $\hbar\omega_2$ . In this case, owing to the transfer of the light energy absorbed by the matrix to the impurity, inversions up to  $10^{19}$  cm $^{-3}$  can be attained, with a relaxation time  $\sim 1$  sec.

Triplet phosphorescence bands of a large number of organic molecules lie near 530 nm (for example, triphenylene, hexaheliken, diphenyl, acenaphthene) or 345 nm, a situation that can be conveniently used for exciting neodymium-glass or ruby lasers. The largest known two-photon transition cross section is possessed by spiropyrene,  $\beta \approx 5 \times 10^{-49}$  cm $^4$  sec.<sup>[17]</sup> Estimates show that it is easy to reach the two-photon amplification threshold of hypothetical impurity molecules with cross sections larger by two orders of magnitude than the indicated value.

Other promising media are metal vapors. Here the two-photon absorption cross sections are quite large:  $\beta_2 \approx 10^{-41}$  cm $^4$  sec and the amplification threshold is reached at low inversions ( $\Delta N_2^0 \sim 10^{14}$  cm $^{-3}$ ). In addition, the effect of the narrowing of the Doppler-broadened two-photon absorption line in the standing wave makes it possible to realize, at low pressures, SWA with even smaller values of the inversion.

In conclusion, let us dwell briefly on the problem of multiphoton amplifiers with a higher phototransition order ( $k > 2$ ). The results obtained in the present paper for two-photon processes ( $k = 2$ ) can be generalized to include also  $k > 2$ . In particular, for a multiphoton TWA, with an exciting-light intensity increasing linearly with time, an expression can be obtained for the power gain

$$K_E \approx 2\eta_E (1 - \eta_E)^{2/(k-1)} I^{(k-1)/(k+1)},$$

$$l = \sigma_{eff} \Delta N_k^0 L_{amp}$$

where the effective cross section of the induced radiation is

$$\sigma_{\text{eff}}^{(k)} = \frac{dI_0}{dt} \left\{ \frac{\beta_k k(k-1)}{dI_0/dt} \right\}^{2/(k+1)} \quad (15)$$

It follows from (15) that the cross section  $\sigma_{\text{eff}}^{(k)}$  has an unexpectedly weak dependence on the order  $k$  of the radiation process. If, for example, we use the empirical relation  $\beta_k \approx 10^{10-30k} \text{ cm}^{2k} \text{-sec}$  and assume  $dI_0/dt = 10^{35} \text{ photon/cm}^2 \text{ sec}^2$ , then on going from the two-photon process to the three- and four-processes we obtain respectively

$$\sigma_{\text{eff}}^{(3)}/\sigma_{\text{eff}}^{(2)} \approx 0.3, \quad \sigma_{\text{eff}}^{(4)}/\sigma_{\text{eff}}^{(2)} \approx 0.1.$$

This circumstance offers more opportunities for finding systems that are promising for the excitation of coherent radiation on multiphoton transitions.

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<sup>1</sup>In this case it is possible also to neglect the linear loss, which is characterized by the time  $\tau_c$ .

<sup>2</sup>In the case of propagation of very short signals in amplifying media, account must be taken of the coherence of the interaction,<sup>[14]</sup> but the results of such an analysis are still qualitatively close to those obtained with the aid of the kinetic equation.

<sup>3</sup>This is equivalent to the condition  $2\beta_2 \Delta N_2^0 T_0 L_{\text{amp1}} = 1$ .

<sup>4</sup>The durations of a pulse with slope  $10^{35} \text{ photons/cm}^2 \text{ sec}^2$  needed for effective amplification, at the given parameters, are respectively  $10^{-8}$  and  $10^{-9}$  sec. The amplified-signal powers are then  $2 \times 10^9$  and  $2 \times 10^8 \text{ W/cm}^2$ .

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## Exchange spin polarization in a three-particle system

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A theory of spin polarization in a system of three particles, each of which has a spin 1/2, is developed. It is assumed that initially the whole system is in a given spin state. A redistribution of the initial polarization occurs during collisions between the particles as a result of exchange interaction. In particular, a beam of unpolarized electrons becomes polarized to a certain extent upon being scattered by oriented atoms. The obtained results allow a uniform computation of the polarization of the scattered beam and that of the target in elastic and inelastic collisions accompanied by particle exchange in the case of arbitrary initial polarization states. Results are presented of numerical calculations of the polarization of electrons scattered by oriented He(2<sup>3</sup>S) atoms and of the depolarization of electrons as a result of singlet-triplet transitions.

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Interest in spin-dependent processes, which has been displayed in recent years in the physics of atomic collisions, is stimulating the search for efficient sources of polarized electrons and the corresponding theoretical computations. To obtain oriented electron beams at present, the following physical processes are used: Mott scattering, the Fano effect, the photoionization of polarized atoms, photoemission from magnetic materials, and the Penning ionization of oriented atoms. In

the first two cases the polarization mechanism is connected with the relativistic spin-orbit interaction, which leads to the appearance of a preferred orientation in the electron beam. In the remaining processes the polarized beams are formed as a result of inelastic transitions leading to the emission of polarized atomic electrons.

Burke and Schey<sup>[1]</sup> first pointed out the feasibility of