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Regenerative amplification of picosecond 10-μm pulses in a high-pressure optically pumped CO_2 laser

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***Abstract*. A model is developed and numerical calculations are performed for the regenerative amplification of seed picosecond 10-μm radiation pulses in a high-pressure optically pumped CO_2 laser. It is established that by varying the cavity parameters, the conditions of efficient regenerative amplification may be optimised for the case of a relatively short-duration (50 ns) solid-state Cr : Er : YSGG laser and 'long-duration' (250 ns) electrodischarge chemical HF laser, used for pumping. It is shown that the schemes of a CO_2 amplifier with optical pumping presented allow obtaining the pulses with the duration of 3 ps, output energy of 1 – 5 mJ, and peak power of ~ 1 GW at the pump energy of 150 – 200 mJ.**

Keywords: *high-pressure CO_2-laser, optical pumping, regenerative amplification of picosecond pulses.*

1. Introduction

In recent years, the works on generation of superpower picosecond laser pulses in the range of 10 μm are in progress [1 – 3]. Such pulses are usually amplified by high-pressure CO_2 amplifiers [4, 5]. Obtaining picosecond pulses with energies of ~ 1 J and higher requires gains that are well above 10^6, which is a difficult task in using a linear circuit of amplifiers. Employment of a seed pulse at the first stage of the regenerative amplification scheme [4, 5] substantially helps solving the problem and provides obtaining a pulse energy on the order of a millijoule at gains above 10^5.

An electric discharge, widely used for pumping active media of high-pressure CO_2 lasers, has drawbacks related, in particular, to molecule dissociation and instability at high pressures. Stabilisation of the discharge by an electron beam enhances its stability, but does not solve the problem of fast degradation of an active mixture.

Transfer to optical pumping of an active medium seems promising. In a high-pressure optically pumped CO_2 laser, it is important that the lasing energy level could be excited selectively, which provides a high-energy deposition (up to 10^3 J L^{-1}) as well as high efficiency [6]. There are schemes for optical pumping high-pressure CO_2-lasers, mainly, by a radiation of chemical lasers: HBr ($\lambda = 4.3$ μm) [7], HF ($\lambda = 2.74$ μm) [8], and DF lasers ($\lambda = 3.64$ μm) [9]. The conversion efficiency in the case of pumping by the radiation with $\lambda = 4.3$ μm reached 40 % by energy at almost 100 % quantum efficiency. Note that in recent years, a substantial progress is observed in our country in the creation of nonchain chemical self-sustained-discharge HF(DF) lasers, generating pulses with a duration of ~ 200 ns and energy of dozens and hundreds of joules [10, 11]. In pumping a high-pressure CO_2 laser by the radiation of a nonchain chemical laser at the wavelength $\lambda = 2.78$ μm (the pulse energy and duration are, respectively, 40 J and 2.5 μs) the laser pulses with the energy of ~ 7 J and duration of ~ 50 ns were obtained [12] at the wavelength around 10 μm.

Unfortunately, high-power chemical lasers are not appropriate for optical pumping in laboratory conditions. In addition, at high output energy it is difficult to obtain a pulse-periodic operation regime. Thus, the schemes suggested in [13] for pumping by a radiation of solid-state lasers may be promising. The most efficient among these schemes is, probably, excitation of compound vibrations (the levels 10^01, 02^01) by radiation of the lasers operating in the range 2.7 – 2.8 μm with the active medium doped with erbium.

An important advantage of solid-state lasers as compared to chemical lasers is a shorter pulse duration (50 – 100 ns) obtained by *Q*-switching and the repetition rate of 5 – 10 Hz without a noticeable fall in the output energy. Making the pumping pulse duration shorter is important because relaxation processes are faster at an elevated pressure of an active medium. In [14], a scheme of the high-pressure CO_2 amplifier, optically pumped by a Cr : Er : YSGG-laser, was suggested, realised, and exhibited high efficiency. The wavelength of the main pump laser line is $\lambda = 2.796$ μm, which is almost optimal from the viewpoint of conditions for absorption by carbon dioxide molecule and further amplification.

Presently, there exists a technical possibility for realising a high-pressure regenerative CO_2 amplifier with optical pumping, which operates in a pulse-periodic regime. However, there were no numerical modelling investigations for regenerative amplification of ultrashort pulses in such an amplifier until the present time.

This study is aimed at discussing an optical scheme, calculation model, and numerical simulation results for amplification of picosecond pulses in a regenerative high-pressure optically pumped CO_2 amplifier.

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2. Exciting scheme

In the general form, a regenerative amplifier comprises an active medium placed into the cavity formed by two mirrors, one of which is dichroic and transmits the pump radiation (see Fig. 1). The active medium is limited by Brewster windows (NaCl) and the cavity includes an optical element (a germanium plate with a thickness of ~ 100 μm), which provides injection of a seed pulse and output of the amplified single pulse. The optical elements of the cavity should introduce no noticeable losses. Note that modern deposition technologies are capable of producing mirrors for the middle IR band with a reflection coefficient of 99 % – 99.5 %. The time of regenerative amplification should not substantially differ from the pump pulse duration, which imposes certain limitations (not strong) on the cavity length. Optical damage of the elements limits the density of energy flux in the pump beam. Gain saturation (depletion of the inversion population for the laser transition) and nonlinear phenomena in optical elements (mainly in the germanium plate) limit the density of the energy flux and power of the amplified pulse.

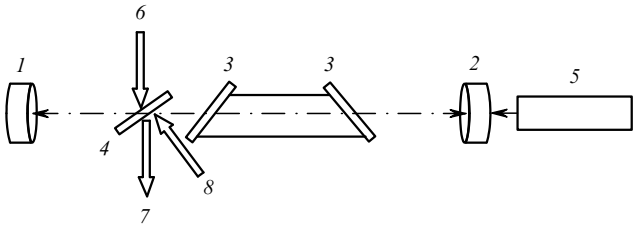

Figure 1. Schematic diagram of a high-pressure regenerative CO_2 amplifier with optical pumping: (*1*) spherical mirror; (*2*) spherical dichroic mirror; (*3*) Brewster windows; (*4*) germanium plate; (*5*) pump laser; (*6*) injected ultrashort pulse; (*7*) amplified radiation; (*8*) laser pulse producing a plasma mirror.

In what follows, we assume that the high-pressure CO_2 laser is pumped by a laser radiation with the frequency resonant to those of the transitions $000 - 02^01$. Figure 2 shows the line shape of this transition calculated for the mixture comprising carbon dioxide at the pressure of 1 atm and at room temperature, dissolved in helium at the pressure of 15 atm. The calculation was performed by the formula

$$F(\omega) = \sum_j n_j \left[\frac{j}{2j+1} f(\omega - \Omega_P(j)) + \frac{j+1}{2j+1} f(\omega - \Omega_R(j)) \right]. \quad (1)$$

Here, $f(x) = \delta/[\pi(x^2 + \delta^2)]$; δ is the collision broadening of the line; n_j is the equilibrium population for the jth rotational sublevel of the fundamental vibration level; $\Omega_P(j)$ and $\Omega_R(j)$ are the frequencies of P- and R-branches for the transitions $000 - 02^01$. The spectroscopic data needed for calculating these frequencies are given in [15]. The Einstein coefficient for the transition under study is close to 11.2 s^{-1}. In the calculations, the absorption coefficient at the wavelength of optical pumping was ~ 0.1 cm^{-1}. Such a coefficient limits the length of the active medium to 15 – 20 cm. The pump radiation intensity was well below

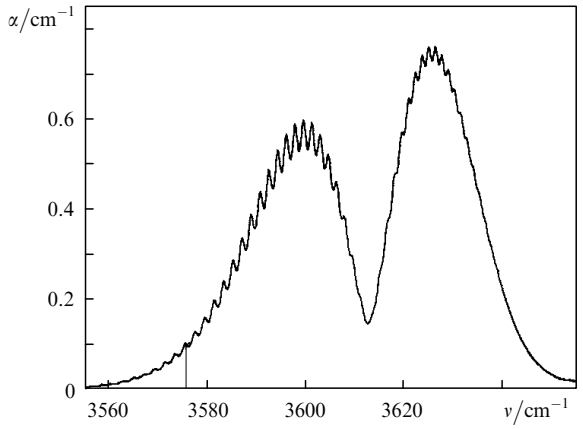

Figure 2. A line shape of the $00^00 - 02^01$ transition. The pump frequency is marked by a vertical line.

the saturation value and varied with a distance according to the Bouguer – Beer law.

The absorption of a single pump quantum increases the energy stored in the asymmetrical mode by one quantum $\hbar\omega_3$ of this mode and the total energy stored in the symmetrical and deformation modes is increased by two quanta $\hbar\omega_2$ of the deformation mode. In this case, as soon as the intramode (VV) relaxation is completed and the equilibrium between symmetrical and deformation modes is established, the population of the 00^01 level is well above that of the 10^00 level. The population difference $N_{00^01} - N_{10^00}$ on the lasing transition also slightly increases due to VT-relaxation processes, which cool the symmetrical and deformation modes faster than the asymmetrical mode.

The populations N_{00^01} and N_{10^00} can be expressed in terms of numbers e_2 and e_3, where e_j is the number of quanta in the jth mode per molecule. We calculated the dynamics of a e_j variation under the assumption of the equilibrium mode distribution over levels and equal temperatures for the symmetrical and deformation modes, which was slightly violated only when the amplified ultrashort pulse passed. In addition to the transitions induced by radiation, we also took into account deactivation of the deformation vibrations and the decay of a quantum of the asymmetric vibrations into three quanta of the deformation mode in collisions. In the notations used in [16], on time intervals where the intensity of a short amplified pulse is negligible, the numbers of quanta e_2 and e_3 are given by the equations

$$\frac{de_3}{dt} = p_{e_3} - r_3 f_3, \quad (2a)$$

$$\frac{de_2}{dt} = f_2[p_{e_2} + 3f_3 e_3 - r_2(e_2 - e_{2T})], \quad (2b)$$

where $p_{e_3} = \sigma I_p/(\hbar\omega_p)$; I_p, ω_p, and σ are the intensity, frequency, and absorption cross section for pump quanta; $p_{e_2} = 2p_{e_3}$; e_{2T} is the equilibrium value of e_2 at a temperature T.

The kinetic coefficient needed for calculating the rates r_3 and r_2, the formulae expressing the numbers f_3, f_2 and level populations N_{00^01} and N_{10^00} in terms of the numbers e_2 and e_3 are given in [16]. Temperature was calculated by the equation

$$\frac{\mathrm{d}T}{\mathrm{d}t} = \frac{r_2\hbar\omega_2(e_2 - e_{2T}) + r_3 f_3 \hbar(\omega_3 - 3\omega_2)}{c_V}, \qquad (2c)$$

where c_V is the heat capacity.

In calculations, the deactivation time for the deformation vibrations was hundred nanoseconds and deactivation time for the asymmetric mode was on the order of a microsecond. Under the action of an amplified ultrashort pulse, the populations of the vibration levels 00^01 and 10^00 actually spasmodically change, and the equilibrium distribution slightly breaks. However, the equilibrium is re-established at the instant of the next pulse passing and such breaks actually have no effect.

The evolution of a short-duration pulse in the cavity was described by the equation in a moving coordinate system:

$$2\mathrm{i}k\frac{\partial E}{\partial z} = -\nabla_\perp^2 E - 4\pi\frac{\omega^2 P}{c^2}, \qquad (3)$$

where E and P are the amplitudes of the field $\boldsymbol{E} = \mathrm{Re}\{E\exp[\mathrm{i}(kz - \omega t)]\}$ and polarisation $\boldsymbol{P} = \mathrm{Re}\{P\exp[\mathrm{i}(kz - \omega t)]\}$.

Equations (2) were solved by the Runge–Kutt method; Eqn (3), which describes evolution of the transversal structure of a laser beam, was solved by the Crank–Nicolson method. The process of regenerative amplification was calculated by stages. For initial conditions at each successive stage, we used the results obtained at the previous stage.

In the course of regenerative amplification of the ultrashort seed pulses with an increase in their energy and, hence, in the intensity, a phase self-modulation effect (PSM) may arise on the Kerr-type nonlinearity. Polarisation in through optical elements of the cavity (the plate for input and output of the amplified pulse, the windows of a gas cell) was calculated with allowance made for the nonlinear summand $n_2 I$ in the expression for a refractive index (here, I is the intensity, $n_2^{\mathrm{Ge}} \approx 3\times10^{-13}$ cm^2 W^{-1}, $n_2^{\mathrm{NaCl}} \approx 3\times10^{-16}$ cm^2 W^{-1} [17], and the thickness of a window from NaCl is 5 mm). Polarisation in the amplifying medium was presented as the sum $\boldsymbol{P} = N_{\mathrm{CO_2}}\sum_j p_j$, where p_j is the polarisation on the vibration–rotational transition $|10^00, j-1\rangle \leftrightarrow |00^01, j\rangle$ for carbon dioxide molecules with the concentration $N_{\mathrm{CO_2}}$ (the frequency ω matches that of the R-branch of the transition $10^00 - 00^01$). Omitting vibration subscripts, we may present the summand p_j in the form $p_j = d_{j-1\,j}\rho_{j\,j-1} + d_{j\,j-1}\rho_{j-1\,j}$, where d is the dipole moment, ρ is the density matrix for a two-level system, which evolves according to the equation (in the interaction representation)

$$\mathrm{i}\hbar\dot{\rho} = -\boldsymbol{E}(d\rho - \rho d) - \mathrm{i}\hbar R, \qquad (4)$$

where R are the relaxation summands. It was assumed that off-diagonal elements of the density matrix attenuate in the time lapse $1/\delta$ and redistribution over rotational sublevels of a vibration level occurs in the time lapse $2/\delta$, that is,

$$R_{jj} = (\rho_{jj} - n_j^0 N_{00^01})\frac{\delta}{2}, \quad R_{j-1\,j-1} = (\rho_{j-1\,j-1} - n_{j-1}^0 N_{10^00})\frac{\delta}{2},$$

where n_j is the normalised equilibrium population of a rotational sublevel. For calculating a variation of population for the vibration levels N_{00^01} and N_{10^00} under the action of the pulse amplified, we also used system of equations (4).

3. Calculation results

In using radiation of a solid-state laser for optical pumping (a 50-ns short pulse), the length of a cell with the active medium was 15 cm, whereas the cavity length was 30 cm (see Fig. 1). The cell was placed symmetrically relative to the mirrors with the radius of curvature 6.5 m. A cross section of the fundamental transversal mode of the cavity ($S = \pi r_0^2/2$, where r_0 is the beam radius at the intensity level of e^{-2}) was 0.05 cm^2. The distribution of the pump radiation intensity was the super-Gaussian profile $I = I_0\exp(-r^6/a^6)$. The cross section of the pump beam ($S_{\mathrm{p}} \simeq \pi a^2$) in the cell was 0.05 cm^2 as well. Such a beam profile allows realising an efficient excitation of an amplifier working mixture and serves as a spatial filter for selecting the lowest mode of the cavity. The energy absorbed in the active medium was 150 mJ and the density of the pump energy flux was approximately 3 J cm^{-2}. The calculations were mainly performed for the mixture $\mathrm{CO_2}:\mathrm{He} = 1:15$ at a total pressure of 16 atm. Note that an increase in the active medium temperature was at most 40°.

Figure 3 presents a typical dependence of the amplified pulse energy versus the number of cavity passes. It was calculated for the injected pulse energy of 3 nJ and duration of 1.2 ps (the carrier frequency is 976 cm^{-1}). The loss per cavity pass was 4 %. The pulse was injected into the cavity at the instant $t_{\mathrm{inj}} \approx 50$ ns delayed by 50 ns from the onset of the pump pulse, whose shape is also presented in Fig. 3. The maximal pulse energy of ~ 2.5 mJ is reached in ~ 110 ns after the injection (after 110 cavity passes). This value slightly increases (by approximately 20 %) if the moment of injection is optimised (at $t_{\mathrm{inj}} \approx 60$ ns) and reduces approximately twice if the loss per cavity pass rises by 8 %. The energy $E_{\max}$ rather weakly depends on the injected energy E_{inj} (in the range of energies $E_{\mathrm{inj}} = 3 - 100$ nJ).

Evolutions of the shape and spectrum of the amplified pulse with the same parameters as in Fig. 3 are given in Figs 4 and 5. On a linear stage, the pulse duration slowly rises and the spectrum contracts whereas the pulse remains transform limited after 152 passes across the cavity. The

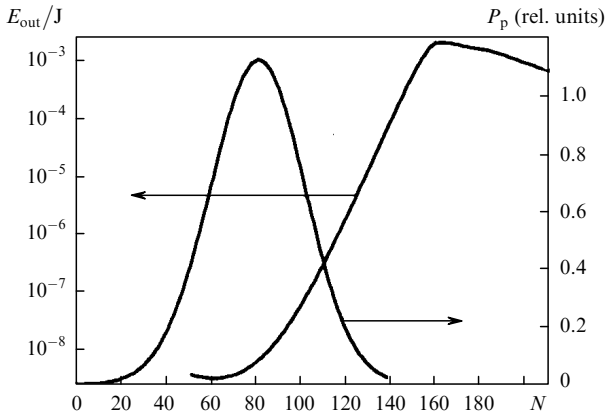

Figure 3. Output energy E_{out} versus the number of passes N through a short cavity and the shape of the pump pulse (P_{p}) with the half-width of 50 ns. The time of passing the cavity is $2L/c = 1$ ns, the instant of injection corresponds to $N = 51$.

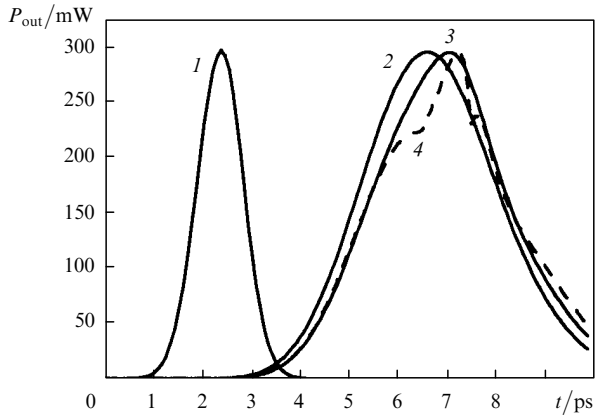

Figure 4. Evolution of the shape of the seed pulse amplified in a short cavity of a regenerative scheme (the value $N = 1$ corresponds to 1 ns).

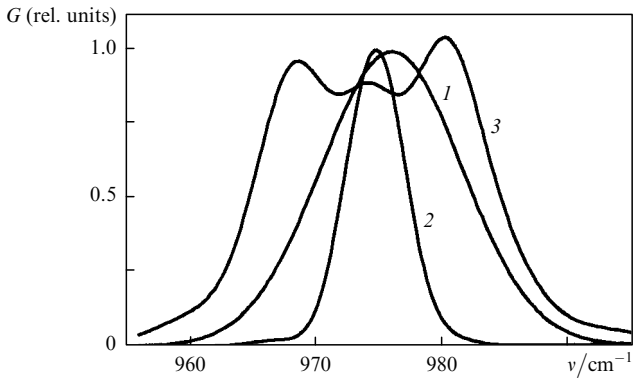

Figure 5. Evolution of the pulse spectrum in amplifying the seed pulse with the energy of 3 nJ in a short cavity: the instant of injection, corresponding to the number of passes $N = 51$ (*1*); and the spectrum at $N = 152$ (*2*) and 158 (*3*).

pulse energy is approximately 1.1 mJ and duration is about 3.1 ps. These parameters weakly change as the duration of the injected pulse varies in the limits 0.6–1.5 ps.

Nevertheless, after 158 passes across the cavity, the pulse shape is slightly distorted and its width corresponds to the duration of a transform-limited pulse from the subpicosecond range. In this case, the pulse energy is 2 mJ and its duration remains close to 3.1 ps.

Finally, after 161 passes across the cavity, the pulse energy increases to 2.25 mJ, its shape is substantially distorted, and the spectrum (not shown in Fig. 5) 'overlaps' not only the R-branch but also the P-branch of the transition.

Simultaneously with the distortion of the tempral structure of the pulse, the transverse structure of the light packet changes. After 152 transits across the cavity, the distortions of this structure are virtually absent. After 158 passes, the packet structure is still close to Gaussian, but its diameter is approximately 2.5 times smaller than the initial and the intensity at the axis is above 7×10^{10} W cm^{-2}. After 161 passes, the structure of the light packet is strongly distorted.

In the calculations performed for the pump pulse with the duration of 250 ns (such a duration is typical of an electrodischarge chemical HF laser), the cavity length was taken 1 m. The cavity was formed by a concave and a convex mirrors with the radii of curvature 170 and −100 cm, respectively. Near the convex mirror, the cross section of the fundamental mode was ~ 0.05 cm^{-2}. The cell 15 cm in length was 'pressed' to the convex mirror. A germanium plate was placed near the concave mirror, where the cross section of the fundamental mode was ~ 0.25 cm^2 (it is almost five times greater than in the cell). Thereby, nonlinear effects in germanium should have played a smaller role. The pump beam had the same shape [$I \sim \exp(-r^6/a^6)$] and cross section (0.05 cm^2) as in the short cavity. The loss per cavity pass was 4 %. A seed pulse with the energy of 10–30 nJ passed to a cavity of a high-pressure CO_2 laser with the delay relative to the pump pulse onset of approximately 240 ns ($N \approx 50$).

For the sake of brevity, we will discuss only the results of calculations performed for the pump energy of 300 mJ (the density of the energy flux is 6 J cm^{-2}). A principal distinction from the previous calculation results for a short cavity is absence of noticeable distortions of the pulse shape and structure at the pulse energy of 5–7 mJ. Note a 'smoother' temporal behaviour of the energy of an amplified pulse (in a range close to its maximal value) in the case of a long cavity. It is worth noting that with a short cavity, a 'smooth' behaviour is observed when the nonlinear element is absent. Such a behaviour of the amplified pulse energy is related to the fact that the onset of PSM at an increased pulse intensity leads to more rapid limitation of the output energy as compared to the conditioned depletion of the population inversion. In a long cavity, the role of PSM is weak and the limitation is caused by the depletion of the population inversion.

With the variant of the cavity considered, the amplified pulse intensity on a semiconductor plate is limited by a value less than 10^{10} W cm^{-2}. The energy of the pulse subjected to negligible influence of PSM E_{max} reached the maximum of 7.5 mJ in approximately 480 ns after the injection (80 passes through the cavity). Note that the energy E_{max} rather weakly depends on the injected energy E_{inj} (in the energy range $E_{inj} = 10 - 100$ nJ).

Having passed 80 transits across the cavity, the pulse approaches a duration of 3.1 ps, its shape is slightly distorted, and the spectrum is weakly broadened due to PSM. Finally, after 83 passes across the cavity (it takes a time of ~ 500 ns), the pulse energy reaches ~ 8 mJ, its shape is substantially distorted, and the spectrum 'covers' not only the R-branch of the transition, but P-branch also. However, the transversal structure of the light packet is distorted insignificantly.

4. Discussion of the results

The results obtained show that without a nonlinear element, the energy of the ultrashort pulse reached in the course of regenerative amplification, relatively weakly depends on its initial energy and duration (in the range 0.5–2 ps) and may be controlled by varying pump and cavity parameters. An increase in the duration of the optical pump pulse from 50 to 250 ns is not principal; however, it requires a longer cavity.

Presence of a nonlinear element inside the cavity limits the energy (or, at least, intensity) of the pulse, which can be reached without distortions of spectral-temporal and spatial structure of the light packet. Hence, the amplified pulse should leave the cavity before PSM causes undesirable

distortions in its structure. Thus, the limitation of the output energy is determined not only by the depletion of the population inversion on the lasing transition, but also by PSM in the nonlinear element.

An important characteristic of the amplified radiation is a signal/noise ratio at the output of the amplifier. We assume that this parameter is independent of the number of passes across the cavity, because amplification occurs in a linear regime. The ratio is determined by the parameters of injected radiation and the energy of noise radiation, which is acquired in the fundamental mode of the cavity in the frequency band $\delta\omega$ to the moment of injection $t_{\rm inj}$. By neglecting the amplification and absorption, we may estimate the energy acquired in the fundamental mode by the formula

$$E^{\rm r}_{\rm noise} \approx AF(\omega)\delta\omega\frac{S}{4\pi L^2}\frac{\omega}{\omega_{\rm p}}\int_0^{t_{\rm inj}} P_{\rm p}(t)(t_{\rm inj}-t){\rm d}t, \qquad (5)$$

where $A \approx 0.21\ {\rm s}^{-1}$ is the Einstein coefficient; L is the cavity length; and $P_{\rm p}$ is the pump power. In the conditions corresponding to Fig. 4, the energy of noise radiation (5) is $\sim 10^{-16}$ J.

In the case of parametric generation of the injected pulse [18], the signal/noise ratio $E_{\rm inj}/E_{\rm noise} = K$ is $10^5 - 10^6$. In other words, already at $E_{\rm inj} = 1$ nJ the energy of the noise injected with a signal $E_{\rm inj}/K$ is well above an energy of the noise $E^{\rm r}_{\rm noise}$ (5) produced by the regenerative amplifier itself. Thus, in a typical situation where the energy of the injected pulse is 0.1 – 1 μJ, the main factor determining a contrast of the amplified pulses is the signal/noise ratio for the injected pulse.

5. Conclusions

Thus, a model is developed and numerical calculations are performed, which ultimately correspond to the regenerative amplification conditions for seed picosecond 10-μm laser pulses in an optically pumped CO_2 amplifier. It is established that by varying cavity parameters one can optimise the conditions of efficient regenerative amplification as with a relatively short-duration (50 ns) solid-state Cr : Er : YSGG pump laser so and of with a 'long-duration' (250 ns) electrodischarge chemical HF-laser. It is shown that the schemes of a presented CO_2 amplifier with optical pumping allow obtaining the pulses with the duration of 3 ps, output energy of 1 – 5 mJ, and power of ~ 1 GW at the pump energy of 150 – 200 mJ. Such an amplifier may be employed as the first stage in a chain of amplifying cascades of a super-power laser system in the 10-μm range and its radiation may be used for carrying out independent fundamental and applied investigations.

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