High-frequency hindrance lifting and possibility of multiphoton amplification in active molecular media

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It is shown that a radiation flux can be amplified with a gain proportional to I^k using vibrational-rotational transitions in active molecular media. Such analogs of two-to five-photon ($k = 1$–4) amplifiers can be designed on the basis of high-frequency lifting of the hindrance in strong light fields. The gain of short pulses in the media of CO and CO_2 lasers is calculated. The results show that the processes mentioned might be used for nonlinear amplification and compression of powerful pulses.

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1. The problem of developing amplifiers and lasers based on stimulated two-photon emission (TPE) has been under discussion in the literature for nearly 15 years.[1-10] One of the main advantages of the use of multiphoton stimulated emission is the possibility of amplifying pulses whose peak power is so high that ordinary laser—"one-photon"—amplification becomes ineffective ("saturation" of the amplification).

No multiphoton amplifiers, however, have been developed so far, mainly because there is no suitable working medium that can satisfy the extremely stringent requirements, namely a high density of the inverted medium and a large TPE cross section.[2,3,6,8] In addition, since realization of TPE calls for a high-intensity input pulse, an important characteristic of the active medium is high optical breakdown strength. In particular, the hopes of attaining TPE from excited atomary iodine,[6,8] the medium of the photodissociation iodine laser, were not fulfilled precisely for the last mentioned reason. Taking all the foregoing factors into account, even the suggestion that metal vapor be used as the active medium, made in one of the latest papers on two-photon amplification of pulses,[10] seems unrealistic.

We plan to show in this paper that it is nevertheless possible to develop amplifiers with gains proportional to $|E|^4$, $|E|^6$, $|E|^8$, and even $|E|^{10}$ (E is the electric field intensity), i.e., analogs of multiphoton (up to five-photon) amplifiers, but of an entirely different type than heretofore considered. The effect that might hopefully realize the multiphoton amplification is the lifting of the hindrances[1)] on vibrational-rotational transitions in strong optical fields,[11] and the suitable active medium is that of the well-investigated CO_2 or CO lasers.

2. We consider the propagation, in an active molecular medium, of a laser pulse whose duration τ_p is longer than the transverse relaxation time T_2. Assume that the laser frequency ω is at resonance with the frequency of the vibrational rotational transition $\omega_{v_1,j_1}^{v_2,j_2}$ ($\omega=\omega_{v_1,j_1}^{v_2,j_2}+\Delta$, $\Delta \ll \omega$ are the vibrational and $j_{1,2}$ the rotational quantum numbers). The amplitude of the polarization of the medium at the frequency ω includes in the general case nonlinear terms due to resonant multiphoton processes of the type $2\omega-\omega=\omega_{v_1,j_1}^{v_2,j_2}+\Delta$, $3\omega-2\omega=\omega_{v_1,j_1}^{v_2,j_2}$, etc., which in analogy with the corresponding absorption processes can be called one-photon emission of third, fifth, etc. order (OPE-3, OPE-5, ...).[11]

In the considered quasistationary approximation, the form of the polarization produced at the frequency ω can be obtained by the method of the generalized two-level system[12]:

$$\mathscr{P}(\omega)=\frac{i}{\hbar}\frac{N_0\eta}{T_2^{-1}+i\Delta}[d^2+2d\varkappa_3|E|^2+(\varkappa_3^2+2d\varkappa_5)|E|^4$$
$$+2(\varkappa_3\varkappa_5+d\varkappa_7)|E|^6+(\varkappa_5^2+2d\varkappa_9+2\varkappa_3\varkappa_7)|E|^8]E, \tag{1}$$

where N_0 is the particle density, η is the difference between the populations of the excited and ground states of the working transitions, $d=\langle v_1, j_1|\hat{d}|v_2, j_2\rangle$ is its dipole moment, $\varkappa_n$ is the n-th order polarizability responsible for the OPE-n in the working transition.[2)]

We call particular attention to the crossing terms of the type $d\varkappa_n|E|^{n-1}E$, $\varkappa_n\varkappa_m|E|^{m+n-2}E$, which are due to interference between processes of different orders. At different field frequencies, these terms lead to parametric interaction of these fields.[12] In our case, however, they do not depend on the phase of the field and play exactly the same role as the incoherent processes. For example, the second term in the polarization (1), which is proportional to $d\varkappa_3$, has exactly the same field dependence as the polarization in two-photon emission and absorption.

Interference terms appear only when the working transition has both a dipole moment and polarizabilities $\varkappa_n$, or else simultaneously polarizabilities $\varkappa_n$ of different orders. This situation occurs, for example in the CO molecule (see Sec. 4 below). In transitions forbidden to one-photon absorption and emission (e.g., $|v, j\rangle \to |v+1, j\pm 3\rangle$), the polarization contains terms starting with the fifth power in the field ($\sim\varkappa_3^2|E|^4E$). Examples are transitions with $\Delta j \pm 3$ in CO_2, for which there are no interference terms at all[3] and only the term $\sim\varkappa_3^2|E|^4E$, which describes the OPE-3 process, is essential. In such transitions, purely nonlinear amplification of laser radiation is possible.

A very essential fact that determines the possibility of using the processes under consideration for amplification and compression of light pulses in molecular media is the large value of the polarizabilities for vibrational-rotational transitions. As shown below, for the processes $j=6\to j=8$ and $j=4\to j=8$ between the states $v=7$ and $v=6$ in the CO molecule, the values of $\varkappa_3$ and $\varkappa_5$ are equal to $\sim 10^{27}$ and 10^{35} cgs esu, respectively (see also Ref. 11).

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3. Taking (1) into account, we easily obtain the following equations for the change of the laser-pulse intensity and for the population difference η in the plane-wave approximation:

$$\frac{\partial I}{\partial z}+\frac{1}{V}\frac{\partial I}{\partial t}=\alpha\eta f(I), \qquad \frac{\partial\eta}{\partial t}+\frac{\eta-\eta_0}{T_1}=-\beta\eta f(I); \tag{2}$$

$$\alpha=\frac{4\pi\omega^2 N_0 T_2^{-1}}{kc^2\hbar(T_2^{-2}+\Delta^2)} \qquad \beta=\frac{4}{\hbar^2}\frac{T_2^{-1}}{T_2^{-2}+\Delta^2}, \tag{3}$$

where $I=|E|^2$, V is the wave propagation velocity, η_0 is the equilibrium population difference, T_1 is the longitudinal relaxation time, and

$$f(I)=d^2I+2d\varkappa_3 I^2+(\varkappa_3^2+2d\varkappa_5)I^3+2(\varkappa_3\varkappa_5+d\varkappa_7)I^4+(\varkappa_5^2+2d\varkappa_9+2\varkappa_3\varkappa_7)I^5+\ldots \tag{4}$$

At $T_2 < \tau_p \ll T_1$, neglecting the terms $(\eta-\eta_0)/T_1$ in the second equation of (2), and introducing the variables $u_1=\alpha z$ and $u_2=\beta(t-z/V)$, we write Eqs. (2) in the form

$$I_{u_1}=\eta f(I), \quad \eta_{u_2}=-\eta f(I), \tag{5}$$

which reduces to an ordinary differential equation of the form

$$dI/du_2=f(I)[(I_0)_{u_2}/f(I_0)-I+I_0], \tag{6}$$

where

$$I_0=I(u_1,u_2)|_{u_1=0}, \quad (I_0)_{u_2}=\partial I_0/\partial u_2. \tag{7}$$

Unfortunately, it is impossible to solve this equation for $f(I)$ in general form. An approximate investigation of the amplification process, however, is possible if the following considerations are taken into account.

The effective gain of a pulse on account of quadratic, cubic, etc. terms of $f(I)$ occurs at substantially different intensities of the light pulse (see Secs. 5 and 6), so that we can consider separately the gain due to each of these terms. In addition, as noted above, there exist transitions for which the principal role in the polarization (1) is played by one of the terms. We must therefore first discuss pulse propagation in a nonlinear medium whose polarization is described by one particular term of (1). In such cases Eq. (7) can be represented in the form

$$\frac{dI}{d\Theta}=I^n\left[-\frac{1}{n-1}\frac{dI_0^{-n+1}}{d\Theta}-I+I_0\right], \tag{8}$$

$$\Theta=\mathscr{K}u_2, \tag{9}$$

$\mathscr{K}$ is the coefficient of I^n in (4). We rewrite the first equation of (5) by introducing the variable $\zeta=\mathscr{K}u_1$ in the form

$$I_\zeta=\eta I^n. \tag{10}$$

An approximate solution of (8) and (10) can be obtained by the method proposed in Ref. 10 for the investigation of two-photon amplification of pulses. We shall apply it to our problem.

The expression for the leading front of the amplified pulse can be obtained from (10) by putting $\eta\approx 1$:

$$I=[I_0^{1-n}-(n-1)\zeta]^{1/(1-n)}. \tag{11}$$

The trailing edge of the pulse (large Θ) can be described by the series $I=I^{(1)}+I^{(2)}+\ldots$, $(I^{(n)}\ll I^{(n-1)})$, where $I^{(n)}$ is obtained from (8) in which we assume in succession $\partial I^{(1)}/\partial\Theta=0$, $\partial I^{(2)}/\partial\Theta=0$, etc. From this we get

$$I^{(1)}=I_0^{1/(1-n)}+\frac{1}{1-n}\frac{\partial I_0}{\partial\Theta}, \qquad I^{(2)}=-\frac{\partial I^{(1)}}{\partial\Theta}[I^{(1)}]^{-n},\ldots \tag{12}$$

It is convenient to describe multiphoton amplification by approximating the laser pulse by a linear function of the time[10]

$$I_0(\Theta)=\gamma\Theta, \quad \gamma=\Delta I_0/\Delta\Theta. \tag{13}$$

Introducing next the dimensionless variables

$$x=\gamma^{n/(n+1)}\Theta, \quad y=\gamma^{-1/(n+1)}I, \tag{14}$$

we can represent the expression for the shape of the pulse after it passes through the length, in first-order approximation, in the form

$$y=\begin{cases} x(1-lx^{n-1})^{-1/(n-1)}, & x<x_m \\ x+x^{-n}, & x>x_m \end{cases}, \tag{15}$$

where the joining point of the solutions x_m is determined from the equation

$$x_m(1-lx_m^{n-1})^{-1/(n-1)}=x_m+x_m^{-n}. \tag{16}$$

The quantity l in (15) and (16) is given by

$$l=(n-1)\gamma^{(n-1)/(n+1)}\zeta=(n-1)\left[\left(\frac{\Delta I_0}{\Delta t}\right)\frac{1}{\beta\mathscr{K}}\right]^{(n-1)/(n+1)}\alpha\mathscr{K}z. \tag{17}$$

By comparing the solution (15) with the numerical solution of the initial equations (5) we can establish that these solutions agree approximately at $l^2\gg 1$. It follows from (15) and (16) at at such l we have $x_m\approx l^{-1/(n-1)}$ and effective amplification of the pulses takes place.

Figure 1 shows the shapes of the output pulse at $l=6$, plotted by using (15) and by numerically integrating Eqs. (5) at $n=3$ and $l=6$.

The maximum amplitude of the output pulse at $l^2\gg 1$ can be approximately estimated from (16):

$$y_m=x_m+x_m^{-n}\approx l^{-1/(n-1)}+l^{n/(n-1)}. \tag{18}$$

With the aid of (15) we can find the energy gain for the leading front of the input pulse:

$$K_e=\left[\int_0^{x_m}x(1-lx^{n-1})^{-1/(n-1)}dx+\int_{x_m}^{x}(x+x^{-n})\,dx\right]\left[\int_0^x x\,dx\right]^{-1}$$

$$=\frac{2}{x^2}\int_0^{x_m}x(1-lx^{n-1})^{-1/(n-1)}\,dx+\frac{x^2-x_m^2}{x_m^2}+\frac{2}{n-1}\frac{(x^{n-1}-x_m^{n-1})}{x^{n+1}x_m^{n-1}}. \tag{19}$$

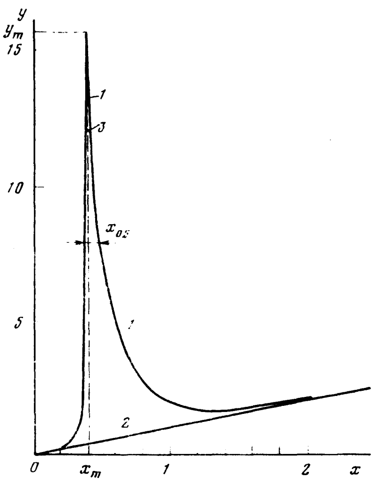

FIG. 1. Curve 1—shape of output pulse, plotted in accord with Eq. (15) for $l=6$; 2—leading front of the input pulse; 3—result of numerical integration of Eqs. (5).

If $x^2 \gg x_m^2$, then the first term of (19) can be neglected. The optical efficiency of such an amplifier, defined as the ratio of the energy drawn by the pulse from the medium to the energy stored in the active medium is

$$m(\Theta)=\frac{1}{\zeta}\int_0^{\zeta}[1-\eta(\zeta_1,\Theta)]d\zeta_1. \tag{20}$$

Recognizing that $\eta(\zeta, \Theta) = I_\zeta / I^n$ [see (10)], we get from (20)

$$m=1+\frac{I^{1-n}-I_0^{1-n}}{(n-1)\zeta}$$
$$\approx 1+\frac{1}{l}[(x+x^{-n})^{1-n}-x^{1-n}]. \tag{21}$$

The amplified pulse becomes compressed at $l^2 \gg 1$. The width x_r of the output pulse on the level r can be determined approximately by solving the equation $x_1 + x_1^{-n} = ry_m$, $x_r = x_1 - x_m$. For a rough estimate we can use the approximate formula

$$t_r=(r^{-1/n}-1)x_r$$
$$\times \gamma^{-n/(n+1)}\beta^{-1}\mathcal{K}^{-1}. \tag{22}$$

4. We consider now the conditions for realization of nonlinear amplification in the active medium of an electro-ionization CO laser at atmospheric pressure.[13] We investigate first the transitions forbidden to one-photon emission. For the transition $v=6$, $j=8 \to v=7$, $j=6$ the only nonzero polarization terms begin with the fifth power in the field [see (1)], and are proportional to $\varkappa_3^2$. The schemes of the transitions that make the main contribution to $\varkappa_3$ are shown in Fig. 2 [see also Eq. (4) of Ref. 11]. Recognizing that the dipole moments of the transitions $d_{v_1,j_1}^{v_1+1,j_2} \approx \sqrt{v_1}\cdot 10^{-19}$ cgs esu, we can easily find that $\varkappa_3$ for the laser frequency $\nu = \nu_{6,8}^{7,6} = 1930.2\ \text{cm}^{-1}$ is equal to $\varkappa_3 \approx 1.3\cdot 10^{-27}$ cgs esu.[4)] From the experimental data on the energy input and efficiency of CO lasers at atmospheric pressure[13] we can easily estimate the density of the inverted particles in the transitions $v \to v-1$ of the active medium: at 5 atm it is easy to ensure $N_0 = 5\cdot 10^{17}$. In the nanosecond pulse regime ($\tau_p \lesssim 2-3$ nsec · atm) we can neglect the rotational-translational and vibrational relaxations and assume that only the rotational relaxation is important.[15]

Rigorous calculations can be made only with allowance for the kinetics of the rotational relaxation. Rough estimates are frequently made in an approximation wherein it is assumed that at an amplified-pulse duration $\tau_p > \chi\tau_{\text{rot}}$ (χ is a coefficient larger than unity and characterizes the rate of migration over the rotational spectrum, τ_{rot} is the rotational relaxation time) the energy is drawn from the entire spectrum of the rotational components.[13] This assumption is valid at least for pulses with duration $\tau_p \gtrsim 5\tau_{\text{rot}}$, amplification of which in Ref. 14 (see also Ref. 13) resulted in an energy pickoff of ~70% of the energy stored on the upper laser level of the active medium of an atmospheric-pressure CO_2 laser. In the estimates below the duration of the input pulse satisfies the condition $\tau_p > 5\tau_{\text{rot}}$. In the course of multiphoton amplification, however, the pulse is compressed. As a result, not all the rotational levels of the upper vibrational state contribute to the amplification of these pulses. To simplify the estimates we shall assume nevertheless that if the duration of the exit laser pulse $\tau_p \gtrsim \tau_{\text{rot}}$, then the entire energy stored on the upper level is picked off, i.e., we shall use in the estimates the total density of the inverted particles N_0. This leads, of course, to some underestimate of the threshold values of the inversion density N_0 and of the active-medium length needed for effective pulse amplification, and to an overestimate of the optical efficiency (in the same ratio as in a one-photon amplifier). To obtain the coefficients cited below it is actually necessary to have a somewhat larger amplification length and inversion density N_0. We note that the value $N_0 \sim 5\cdot 10^{17}$ assumed in the estimates is not the limit for electro-ionization CO and CO_2 lasers.[13,15] On the other hand an increase of N_0 by two or three times can compensate for the decrease of the number of rotational levels that participate in the amplification process.

We proceed now to the estimates. Since no measurements were made of the rotational relaxation time in the active medium of a CO laser, we use the value $\tau_{\text{rot}} = T_2 = 2.8\cdot 10^{-10}$ sec · atm obtained from data on collision-broadened absorption lines of CO.[16] For the presented parameters, CGSE ($\mathcal{K} = \varkappa_3^2$). Next, assuming that the slope of the leading front of the input pulse $\Delta I_0/\Delta t = 2\cdot 10^{15}$ CGSE ($\gamma = 5.9\cdot 10^{24}$ CGSE), $l = 6$ ($z = 190$ cm), we obtain from (16) the instant of time at which the maximum peak intensity is reached: $t_m = x_m\gamma^{-3/4}\beta^{-1}\mathcal{K}^{-1} \approx 5\cdot 10^{-10}$ sec. Since the duration t_1 of the leading front should exceed t_m, we find from (18), (19), (21) and (22), by putting $t_1 = 10^{-9}$ (the peak intensity of the input pulse is $S_0 = cI_0/2\pi = 10^9$ W/cm), that $I_m = y_m\gamma^{1/4} = 2.3\cdot 10^{17}$ CGSE ($S_m = 1.1\cdot 10^{10}$ W/cm^2), $K_e = 4.75$, $m = 0.9$, $t_{1/3} = 1.6\cdot 10^{-11}$ sec[5)]. If the trailing edge is also of duration ~1 nsec, i.e., the duration of the input pulse at the 0.5 level is 1 nsec (the energy flux of the input pulse is $W_0 = t_1 S_0 = 1$ J/cm^2), then the energy gain for the entire pulse is $\tilde{K}_e \lesssim K_e/2 = 2.87$. The specific energy pickoff can be calculated from the formula $\Phi = (K_e - 1)W_0/2z = 9\cdot 10^{-3}$ J/cm^3.

A linear approximation of the leading front is not quite correct and, as will be shown below, exaggerates the estimates somewhat (see Fig. 3b). Closer to experiment is an approximation of the input pulse by a "trimmed" Gaussian curve:

$$I_0(t)=I_0\frac{\exp[-(t/\tau_p-1)^2]-e^{-1}}{1-e^{-1}} \quad (0 \leq t \leq 2\tau_p). \tag{23}$$

For this purpose, a numerical integration was carried out of the initial equations (5), in the second of which

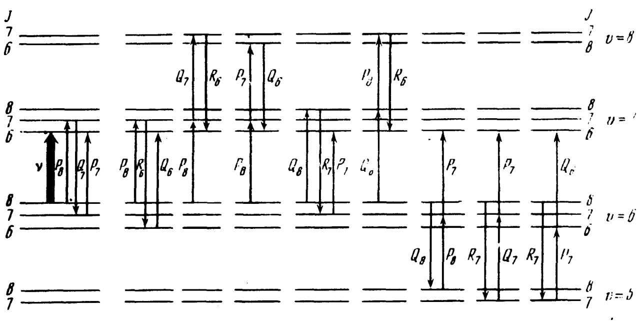

FIG. 2. Diagrams of transitions that contribute to the polarizability for the transition $v=6$, $j=8 \to v=7$, $j=6$ in CO. Thick arrow—frequency of amplified pulse.

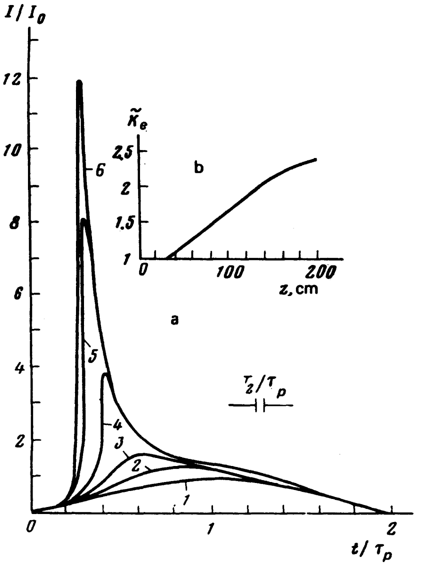

FIG. 3. a) Shapes of amplified pulse at various length of CO-laser active medium. Curves 1–6 correspond to the lengths z = 20, 40, 60, 100, 160, and 200 cm. b) Corresponding dependence of the energy gain $\tilde{K}_e$ on the length ($S_0 = 10^9$ W/cm^2, $\tau_p = 10^{-9}$ sec).

account was taken of the relaxation term $(\eta - \eta_0)/T_1 cT_1 = 5 \cdot 10^{-8}$ sec · atm.[15] All the remaining parameters of the active medium of the CO laser are given in the table. The results are given in Fig. 3a, which shows the shapes of the pulses for various lengths of the active medium. It is seen that a substantial amplification of the peak intensity and of the energy begins at an amplifier length $z \gtrsim 40$ cm. With further increase of the length, the pulse is compressed, its intensity and energy increase, and the intensity maximum shifts to the start of the pulse. In contrast to the case of one-photon amplification, the maximum does not shift directly to the start of the pulse; since the nonlinear amplification has a threshold, the maximum is located at a certain distance from the start of the pulse. The dependence of the energy gain on z is shown in Fig. 3b. It follows from Fig. 3 that the approximate calculations agrees well enough with the results of the numerical integration.

The results of the approximate analysis show that the amplification efficiency depends on the slope of the

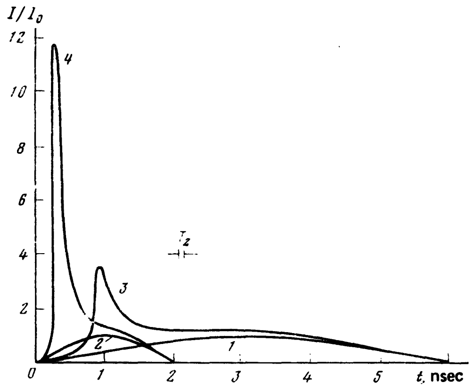

FIG. 4. Shapes of output pulses from the active medium of a CO laser (z = 200 cm) at identical peak intensities $S_0 = 10^9$ W/cm^2 and at various durations of the input pulse: 1, 2—input pulses, 3, 4—output pulses.

leading front. To check this conclusion, Eqs. (2) were numerically integrated for two pulse duration at the same input peak power. The results are shown in Fig. 4, from which it is seen that at a given peak intensity the shorter pulses are amplified more.

The shapes of the output pulses at a fixed length z = 200 cm, as functions of the peak intensities of the input pulses, are shown in Fig. 5. It is seen from this figure that amplification of the peak intensity of the input pulse by only 2.75 times leads to an extremely strong compression of the amplified pulse, to a value $t_r < \tau_{\text{rot}}$. Under these conditions the initial equations are no longer valid. It is clear from physical considerations, however, that after the amplified pulse is compressed to a value $\sim \tau_{\text{rot}}$ any further amplification of the pulse is ineffective, since only a small part then in the amplification; in addition, it is known[17] that pulses with $\tau_p < T_2 \approx \tau_{\text{rot}}$ can not be noticeably amplified at all. Thus, if the compression of the pulse to a value $\tau_p \approx T_2$ takes place over a length much less than that of the active medium, then amplification due to the polarization terms $\sim \varkappa_2^3 |E|^4 E$ [see (1)] can be neglected and it is necessary to take into account the next polarization terms $(\sim 2\varkappa_3\varkappa_5 |E|^6 E,\ \sim(\varkappa_5^2 + 2\varkappa_5\varkappa_7)|E|^8 E$ etc.). To ensure high

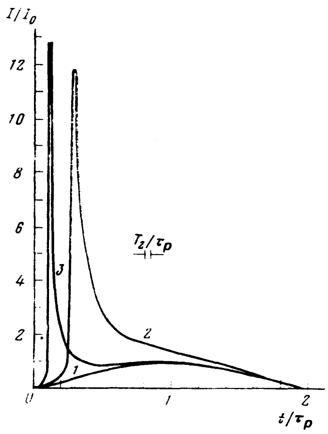

FIG. 5. Shapes of output (2, 3) pulses in the active medium of a CO laser (z = 200 cm) at identical durations ($\tau_p = 10^{-9}$ sec) and various peak intensities ($S_0 = 10^9$ W/cm^2—curve 2, $S_0 = 2.75 \cdot 10^9$ W/cm^2—curve 3) of the input pulse (curve 1).

TABLE I.

	N			
	1	2	3	4
n	3	5	2	3
active medium	CO	CO	CO	CO_2
transition	$j=8(v=6) \to j=6(v=7)$	$j=8(v=6) \to j=4(v=7)$	$P_{10}(v=6 \to v=7)$	$j=1(00^01) \to j=4(10^00)$
mixture pressure, atm	5	5	5	5
N_0, cm^{-3}	$5 \cdot 10^{17}$	$5 \cdot 10^{17}$	$5 \cdot 10^{17}$	$2 \cdot 10^{18}$
τ_{rot}, sec	$5.6 \cdot 10^{-11}$	$5.6 \cdot 10^{-11}$	$5.6 \cdot 10^{-11}$	$4 \cdot 10^{-11}$
$\alpha\mathcal{K}$, cgs esu	$6.5 \cdot 10^{-15}$	$1.6 \cdot 10^{-31}$	$6.5 \cdot 10^{-6}$	$1.4 \cdot 10^{-17}$
$\beta\mathcal{K}$, cgs esu	$3.4 \cdot 10^{-10}$	$8.2 \cdot 10^{-27}$	0.34	$3.7 \cdot 10^{-13}$
τ_p, sec	10^{-9}	$4 \cdot 10^{-10}$	10^{-9}	$3 \cdot 10^{-10}$
l	6	8	4	6
γ, cgs esu	$5.9 \cdot 10^{24}$	$3 \cdot 10^{42}$	$6 \cdot 10^{13}$	$9 \cdot 10^{28}$
S_0, W/cm^2	10^9	$5 \cdot 10^9$	10^7	$5 \cdot 10^9$
W_0, J/cm^2	1	2	10^{-2}	1.5
z, cm	190	600	15	720
S_m, W/cm^2	$1.1 \cdot 10^{10}$	$8 \cdot 10^{10}$	$3.1 \cdot 10^8$	$1.4 \cdot 10^{11}$
W, J/cm^2	2.87	6.7	$8.6 \cdot 10^{-2}$	8.25
K_e	4.75	5.7	16.2	10
Φ, J/cm^3	$9 \cdot 10^{-3}$	$7.8 \cdot 10^{-3}$	$5 \cdot 10^{-3}$	$9 \cdot 10^{-3}$
m	0.9	0.7	0.6	0.55
$t_{1/2}$, sec	$1.6 \cdot 10^{-10}$	$8 \cdot 10^{-11}$	$3.8 \cdot 10^{-10}$	10^{-10}

amplification efficiency of such pulses, we can use also transitions with $\Delta j=\pm 2$, ± 3 at larger values of j_1 (with smaller $\varkappa_3$) or transitions with $\Delta j=\pm 4$, ± 5, for which the first term in the polarization (1) is proportional to $\varkappa_5^2|E|^8E$. The results of a preliminary calculation of the amplification of pulses of frequency $\nu=1890.7$ cm^{-1} on the transition $v=6$, $j=8\to v=7$, $j=4$ in CO ($\varkappa_5=6\cdot 10^{-36}$ cgs esu) are given in the table (column 2). It follows from the table that on this transition one can obtain effective amplification of pulses with intensity $S_0=5\cdot 10^9$ W/cm^2. Pulses of this intensity are not amplified in practice on the transition $j=8\to j=6$ considered above, on account of the polarization terms $\sim\varkappa_3^2|E|^4E$. The possibility of their amplification on the very same transition on account of higher nonlinearities $2\varkappa_3\varkappa_5$ or $\varkappa_5^2+2\varkappa_3\varkappa_7$ depends on the sign of these quantities (see Sec. 5 below).

5. We discuss now the feasibility of nonlinear amplification on a transition allowed in the dipole approximation, when the expression for the polarization (1) contains in addition to the term $\sim d^2E$ also terms $\sim d\varkappa_3|E|^2E$, $\varkappa_3^2|E|^4E$, etc. This situation is possible, in particular, in a CO laser, where the laser transitions in the P branches have both dipole moments and polarizabilities $\varkappa_n$ ($n=3, 5, 7\ldots$). For example, one of the terms $\varkappa_3$ for the line P_k in the band $v\to v+1$ is proportional to the dipole moments of the transition Q_k, P_{k-1}, Q_{k-1} in the same band. In the band $v=6\to v=7$ the value calculated for the P_7 transition in $\varkappa_3=-2\cdot 10^{-26}$ cgs esu while for the transition P_{10} in the same band is $\varkappa_3\approx 3.2\cdot 10^{-27}$ cgs esu.

Assume that the slope of the leading front and the length of the medium are such that only the first two terms of (4) are significant, the linear ($\sim I$) and the quadratic ($\sim I^2$). Then two fundamentally different cases are possible.

a) If $\varkappa_3<0$, then obviously the nonlinear term in (5) causes absorption of high-power pulses.

b) If $\varkappa_3>0$, then the nonlinear term proportional to $d\varkappa_3I^2$, in (4) can lead to effective amplification of pulses whose gains on account of the one-photon process is exceedingly small.[6)] For example, it is easy to estimate pulses with peak intensity $S_0=10^7$ W/cm^2 are not amplified in practice for the P_{10} transition in the band $v=6\to v=7$. For an approximate estimate of the gain we can leave in (4) only the term quadratic in intensity. The results of the calculations are given in the table (column 3). It is obvious that at $\varkappa_3^2+2d\varkappa_5>0$ amplification of more intense pulses is also possible on this transition, on account of the terms of (4) proportional to I^3, and at $\varkappa_3\varkappa_5+d\varkappa_7>0$ on account of the terms proportional to I^4.

6. We can estimate now the possibility of nonlinear amplification of pulses in an active medium of an electro-ionization CO_2 laser.[13,15] If the frequency of the input pulse is tuned to resonance with the transition $j=1(00^\circ1)\to j=4(10^\circ0)$, then the only significant terms in the expression (1) for the polarization are $\sim\varkappa_3^2|E|^2E$, where is determined by the dipole moments of the transitions P_4, R_2 and P_2:

$$\varkappa_3\approx 2d(P_4)d(R_2)d(P_2)/\hbar^2(\omega-\omega(P_4))(\omega(P_4)-\omega(R_2)). \qquad (24)$$

Recognizing that $d(P_i)\approx d(R_i)\approx 2\cdot 10^{-20}$ cgs esu, we obtain from (24) $\varkappa_3=4.8\cdot 10^{-29}$ cgs esu. According to Ref. 13 the rotational-relaxation time is $\tau_{\text{rot}}=2\times 10^{-10}$ sec $\cdot$ atm. At a mixture pressure 5 atm it is easy to obtain $N=2\cdot 10^{18}$ cm^{-3}. The results of the calculation of the gain of pulses of duration 3×10^{-10} sec with a peak intensity $S_0=5\cdot 10^9$ W/cm^3 ($W_0=1.5$ J) are given in the table (line 4). The results attest to the possibility of effective ($m=0.5$) amplification of pulses with energy on the order of several J/cm^2 in the active medium of a CO_2 laser. Such pulses are hardly amplified by the one-photon process.

7. It follows from the foregoing calculations that the considered nonlinear amplifiers can deliver pulses of duration $\sim 10^{-10}$ sec with energy density up to 6–8 J/cm^2 (see the table). An important question is whether the optical breakdown threshold of the active medium is exceeded in this case. There are unfortunately no published data on the optical breakdown of electro-ionization CO lasers at these pulse durations. Only experimental data on the optical breakdown of CO_2 lasers of high pressure are presently known[15] for nanosecond pulses. According to these data the breakdown energy density at a mixture pressure of several atmospheres varies in the range $W_{\text{br}}=3-7$ J/cm^2, depending on the degree of the preliminary ionization of the active medium. This gives grounds for hoping to attain in the considered examples pulse-parameter values close to those listed in the table. On the other hand, if the cited values of the output energy W turn out to be higher than W_{br}, it becomes necessary to decrease the length of the active medium to a value such that $W\lesssim W_{\text{br}}$ (see Fig. 3b).

8. In conclusion, we summarize the results of our calculations.

a) High-frequency lifting of the hindrances is a suitable effect with which to realize multiphoton amplification and compression of high-power pulses.

b) Suitable active media for this amplification are those of the now available CO and CO_2 lasers. It is possible also that the considered amplification method can be effected in active media of molecular lasers of the UV band (e.g., in N_2 or excimer lasers).

c) Since multiphoton amplification is accompanied by a substantial shortening of the pulses, the presented method can be used to produce ultrashort pulses with large peak power, which are needed, in particular, to obtain active media for x-ray lasers.

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[1)]The lifting of hindrances is in fact a multiphoton process that can take place also for some allowed orbital-rotational transitions (see Sec. 2 below).

[2)]In (1), d and $\varkappa_n$ are assumed to be real.

[3)]It is easy to show that this is due to the fact that CO_2 has no absorption lines in the Q branches.

[4)]Electro-ionization CO and CO_2 lasers of high pressure (~10 atm) can be continuously tuned within a sufficiently wide range.[13] The driving laser can therefore be a CO laser of higher pressure than an amplifier tuned to $\nu \approx 1930.2$ cm^{-1}.

[5)]We use as measure of the duration of the output pulse its duration at a level $r = 1/3$ of I_m, since the energy of the output pulse is concentrated in the base of the pulse (see Fig. 1).

[6)]The maximum intensity of the input pulses that are effectively amplified as a result of the one-photon process, can be easily estimated from the known formulas for the gain of a short pulse.[17] Its value for a CO laser at $\tau_p \sim 10^{-9}$ sec is 10^5-10^6 W/cm^2.

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Radiative collisions of atoms with excitation transfer at low resonance defects

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The effective cross section for the transfer of excitation from atom to atom in an external electromagnetic field with absorption and emission of a photon is calculated for small resonance defects. The radiative-collision line contour is determined in the strong- and weak-field approximations. It is shown that allowance for the angular dependence of the dipole-dipole interaction of the atoms affects significantly the line contour near resonance.

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Atomic collisions accompanied by emission or absorption of a photon (radiative collisions) are being intensively investigated both theoretically and experimental (see Yakovlenko's review[1]). We consider in this paper radiative collisions with excitation transfer

$$B^*(2)+A(1)+\hbar\omega \rightleftharpoons B(1)+A^*(2)$$

in an external electromagnetic field, when the photon energy $\hbar\omega$ is close to the energy difference $\hbar\omega_0$ between the initial and final states of the atomic systems (see Fig. 1):

$$E_2^{(A)}+E_1^{(B)}-E_1^{(A)}-E_2^{(B)}=E_{21}^{(A)}-E_{21}^{(B)}=\hbar\omega_0. \quad (1)$$

A resonant situation is produced here (the resonance defect is small) and one can expect relatively large effective cross sections if the electromagnetic field is strong enough. The recent advent of high-power tunable lasers has stimulated to a considerable degree the interest in these processes.

These collisions were first investigated theoretically for the resonant case by Gudzenko and Yakovlenko[2] and in later studies.[3–8] Ostroukhov, Smirnov, and Shlyapnikov[9] have considered the situation when the detuning from resonance is large enough and calculation of the cross section calls for knowledge of the quasimolecular terms of the atomic system. Excitation transfer in a

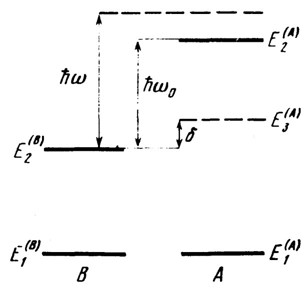

FIG. 1. Level scheme of atomic system.