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Optical pumping of the $N_2O - He$ and $N_2O - CO_2 - He$ mixtures by a pulsed multifrequency HF laser for producing active media to amplify 10-μm high-power ultrashort pulses

G.K. Vasiliev, E.F. Makarov, Yu.A. Chernyshev

***Abstract*. Optical pumping of the $N_2O - He$ and $N_2O - CO_2 - He$ active media (at a total pressure of 1 atm) by $\sim$ 1-μs pulses from a multifrequency HF laser is studied with a view to use these media for amplifying a 10-μm, 10^{-12}-s ultrashort pulse (USP). Transitions forming the USP amplification band are determined, the energy stored in them is found, and the gains corresponding to maximum inversion in the medium are calculated.**

***Keywords*:** *optical pumping, $N_2O - He$ and $N_2O - CO_2 - He$ active media, HF laser, amplification of picosecond pulses*

1. Introduction

We studied experimentally [1] the optical pumping of N_2O molecules in mixtures with Kr and He by 4-μs pulses from a multifrequency HF laser operating on the $H_2 + F_2$ chain reaction. The laser spectrum contained approximately 30 lines in the wavenumber range $k = 3000 - 3700$ cm^{-1}. In [2], we proposed to use N_2O molecules in mixtures with inert gases pumped by a pulsed HF chemical laser for amplifying ultra-power ultrashort pulses (USPs) in the mid-IR range. Pumping produces population inversion at the $00^01 - 10^00$ vibrational transition in N_2O molecules; a radiation pulse of duration $\sim 10^{-12}$ s with a wavenumber $k = 927.74$ cm^{-1} (or 948.59 cm^{-1}) is supplied from a master oscillator to the system at the instant of maximum population inversion and is amplified in several sections. It was noted [2] that the efficiency of optical pumping by a multifrequency HF laser can be increased by using CO_2 molecules along with the N_2O molecules (which absorb radiation lines lying in the range 3220 – 3490 cm^{-1}). The absorption spectrum of HF laser radiation by CO_2 molecules is shifted relative to the spectrum of N_2O molecules to the blue and corresponds to the lines that are not involved in the N_2O pumping in the frequency range 3530 – 3690 cm^{-1}. At the same time, the emission spectra of the N_2O and CO_2 molecules at the $00^01 - 10^00$ transitions overlap (the band centres are located at 939 and 961 cm^{-1}); therefore, a common amplification band of USPs can be formed in principle.

An increase in the pump efficiency was demonstrated experimentally in the study of lasing in the $N_2O - He$, $CO_2 - He$, and $N_2O - CO_2 - He$ active media under a high pressure ($p \sim 10$ atm) [3]. However, a special analysis is required to explore the prospects of employment of $N_2O - CO_2$ mixtures for generating short pulses. A concept of designing an ultra-power laser system generating USPs under optical pumping (scientific substantiation, expected parameters, technical implementation and prospects of modifications) was proposed in [4]. In this concept, the main stage is saturated amplification in an active medium with a discrete spectrum (non-overlapping upon an increase in pressure). In main sections with saturated amplification, the active medium pressure is assumed to be equal to 1 atm.

Here, we carry out a detailed analysis of the N_2O and $N_2O - CO_2$ active media (at a total pressure of 1 atm), which are obtained upon pumping by pulses from a multifrequency HF laser.

2. Description of the optical pump model

Absorption of the HF laser lines induces vibrational transitions in N_2O (and CO_2) molecules, accompanied by excitation of either a symmetric and an asymmetric modes $[v_1v_2v_3 - (v_1 + 1)v_2(v_3 + 1)]$ or a deformation and an asymmetric modes $[(v_1v_2v_3 - v_1(v_2 + 2)(v_3 + 1)]$. Here, v_1, v_2, v_3 are the quantum numbers of three fundamental vibrational modes of a linear molecule: symmetric, deformation, and asymmetric. An analysis reveals that the contribution to the absorption of HF laser radiation by N_2O (and CO_2) molecules comes from the following states: 00^00, 01^10, 02^l0 ($l = 0, 2$), 10^00, 03^l0 ($l = 1, 3$), 00^01, 01^11, 02^l1 ($l = 0, 2$), 00^02, 01^12 (written in the ascending order of energies).

We introduce the intensities I_i of the HF laser lines (in quantum cm^{-2} s^{-1}) ($i = 1 - 22$), the relative populations n_{jm} of the absorbing vibrational states [$j = 1 - 10$, $m = 1 \div (l + 1)$], the relative populations f_k of the rotational states ($k = 0 - 50$), and the absorption cross section σ_{ijkm} for laser lines by individual states of the molecules. In this notation, the pump rate is $\sum_{ijkm} \sigma_{ijkm} I_i n_{jm} f_k [M]$, where $M = N_2O, CO_2$. The absorption cross sections were calculated by using the vibrational band intensities from [5 – 7], the data on collisional broadening of N_2O and CO_2 lines from [5, 8, 9], and wavenumbers of laser lines from [10]. All possible transitions in N_2O and CO_2 lying within ±1.5 cm^{-1} from the centre of the laser pump line were taken into

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account while calculating the laser radiation absorption. The absorption line profile was assumed to be Lorentzian. A total of ~ 500 and ~ 200 individual vibration–rotation transitions in N_2O and CO_2 molecules respectively were considered. The spectroscopic data for these transitions were borrowed from [5, 11, 12] for N_2O and [6, 7] for CO_2.

An analysis of all possible vibrational relaxation processes in the systems studied here shows that equilibrium in individual modes and between symmetric and deformation modes is observed during optical pumping under conditions being simulated; in addition, equilibrium is also observed between the deformation and asymmetric modes in mixtures of N_2O and CO_2 molecules. This makes it possible to consider separately the energies stored in the symmetric, deformation and asymmetric modes and to introduce the rate constants describing V–V energy transfer from the asymmetric mode to a combination of the other two modes upon collisions with unexcited N_2O and CO_2 molecules and atoms of the diluent gas, as well as the rate constants describing the energy transfer from a combination of symmetric and deformation modes to translational degrees of freedom (which coincide with the rate constants of the V–T relaxation of the deformation mode at a moderate temperature).

Thus, the main energy relaxation channels are associated with the following processes:

$$(00^01) + \mathrm{M} \overset{k_{3\mathrm{M}}}{\longleftrightarrow} (11^10),\ (03^10) + \mathrm{M}, \tag{1}$$

$$(01^10) + \mathrm{M} \overset{k_{2\mathrm{M}}}{\longleftrightarrow} (00^00) + \mathrm{M}, \tag{2}$$

where the vibrational states of N_2O and/or CO_2 molecules are given in the parentheses; $\mathrm{M} = \mathrm{N_2O}, \mathrm{CO_2}, \mathrm{He}$; $k_{2\mathrm{M}}$ and $k_{3\mathrm{M}}$ are the rate constants of the V–T and V–V energy transfer, which are connected with the time $\tau_{i\mathrm{M}}$ of vibrational relaxation via these channels through the relation $k_{i\mathrm{M}} = (p\tau_{i\mathrm{M}})^{-1}$ $(i = 2,\ 3)$.

We introduce the average numbers ε_1 and ε_3 of quanta in the symmetric and asymmetric modes. For the sake of convenience in writing the equations, we assume that ε_2 is equal to half the average number of quanta in the deformation mode. Using the superscripts N and C for the rate constants and variables pertaining to the N_2O and CO_2 molecules and introducing the functions W, W_{VV}, W_{VT} describing the pump and the V–V and V–T energy transfer, we obtain the following expressions for $\varepsilon_2^{\mathrm{N}}$ and $\varepsilon_3^{\mathrm{N}}$:

$$\frac{\mathrm{d}\varepsilon_2^{\mathrm{N}}}{\mathrm{d}t} = \frac{W + 1.5W_{\mathrm{VV}} - W_{\mathrm{VT}}}{F_1}, \tag{3}$$

$$\frac{\mathrm{d}\varepsilon_3^{\mathrm{N}}}{\mathrm{d}t} = \frac{W - W_{\mathrm{VV}}}{F_2}. \tag{4}$$

These equations should be supplemented with the equation for running temperature and the expressions connecting the missing average number of quanta with $\varepsilon_2^{\mathrm{N}}$ and $\varepsilon_3^{\mathrm{N}}$ through the equilibrium conditions. For a two-component system, e.g., N_2O – He, we have

$$\begin{aligned}
T &= 300 + \frac{3340[\mathrm{N_2O}]/[\mathrm{M}]}{1 + 1.43[\mathrm{N_2O}]/[\mathrm{M}]}\left\{\int_0^t \sum_{ijkm} \sigma_{ijkm}^{\mathrm{N}} I_i\, n_{jm} f_k \mathrm{d}t - 0.37\right.\\
&\quad \left.\times\left[\varepsilon_1^{\mathrm{N}} + 0.92\left(\varepsilon_2^{\mathrm{N}} - 0.06\right) + 1.73\varepsilon_3^{\mathrm{N}}\right]\right\},\\
W &= \sum_{ijkm} \sigma_{ijkm}^{\mathrm{N}} I_i\, n_{jm} f_k,\\
W_{\mathrm{VV}} &= \sum_{\mathrm{M}} k_{3\mathrm{M}}^{\mathrm{N}}[\mathrm{M}]\left(1 + \varepsilon_1^{\mathrm{N}}\right)\left(1 + \varepsilon_2^{\mathrm{N}}\right)\varepsilon_3^{\mathrm{N}},\\
W_{\mathrm{VT}} &= \sum_{\mathrm{M}} k_{2\mathrm{M}}^{\mathrm{N}}[\mathrm{M}]\left(\varepsilon_2^{\mathrm{N}} - \varepsilon_{02}^{\mathrm{N}}\right),\\
F_1 &= 1 + 2\left(1 + \frac{1}{\varepsilon_2^{\mathrm{N}}}\right)\left(\frac{\varepsilon_1^{\mathrm{N}}}{\varepsilon_2^{\mathrm{N}}}\right)^2 \exp\left(\frac{154}{T}\right),\\
F_2 &= 1,\\
\varepsilon_1^{\mathrm{N}} &= \left[\left(1 + \frac{1}{\varepsilon_2^{\mathrm{N}}}\right)^2 \exp\left(\frac{154}{T}\right) - 1\right]^{-1}.
\end{aligned} \tag{5}$$

For the three component system N_2O – CO_2 – He, we have

$$\begin{aligned}
T &= 300 + \frac{3340[\mathrm{N_2O}]/[\mathrm{M}]}{1 + 1.43([\mathrm{N_2O}] + [\mathrm{CO_2}])/[\mathrm{M}]}\left\{\int_0^t W\mathrm{d}t - 0.37\right.\\
&\quad \times\left[\varepsilon_1^{\mathrm{N}} + 0.92\left(\varepsilon_2^{\mathrm{N}} - 0.06\right) + 1.73\varepsilon_3^{\mathrm{N}}\right] - 0.4\frac{[\mathrm{CO_2}]}{[\mathrm{N_2O}]}\\
&\quad \left.\times\left[\varepsilon_1^{\mathrm{C}} + 0.96\left(\varepsilon_2^{\mathrm{C}} - 0.06\right) + 1.69\varepsilon_3^{\mathrm{C}}\right]\right\},\\
W &= \sum_{ijkm} \sigma_{ijkm}^{\mathrm{N}} I_i\, n_{jm} f_k + \sum_{ijkm} \sigma_{ijkm}^{\mathrm{C}} I_i\, n_{jm} f_k \frac{[\mathrm{CO_2}]}{[\mathrm{N_2O}]},\\
W_{\mathrm{VV}} &= \left[k_{3\mathrm{N}}^{\mathrm{N}}[\mathrm{N_2O}] + k_{3\mathrm{He}}\,\mathrm{N}[\mathrm{He}] + k_{\mathrm{eff}}[\mathrm{CO_2}]\frac{\varepsilon_2^{\mathrm{C}}}{\varepsilon_2^{\mathrm{N}}}\exp\left(-\frac{67}{T}\right)\right]\\
&\quad \times\left(1 + \varepsilon_1^{\mathrm{N}}\right)\left(1 + \varepsilon_2^{\mathrm{N}}\right)\varepsilon_3^{\mathrm{N}} + \left(k_{3\mathrm{C}}^{\mathrm{C}}[\mathrm{CO_2}] + k_{3\mathrm{He}}^{\mathrm{C}}[\mathrm{He}]\right)\frac{[\mathrm{CO_2}]}{[\mathrm{N_2O}]}\\
&\quad \times\left(1 + \varepsilon_1^{\mathrm{C}}\right)\left(1 + \varepsilon_2^{\mathrm{C}}\right)\varepsilon_3^{\mathrm{C}},\\
W_{\mathrm{VT}} &= \sum_{\mathrm{M}} k_{2\mathrm{M}}^{\mathrm{N}}[\mathrm{M}]\left(\varepsilon_2^{\mathrm{N}} - \varepsilon_{02}^{\mathrm{N}}\right) + \sum_{\mathrm{M}} k_{2\mathrm{M}}^{\mathrm{C}}[\mathrm{M}]\left(\varepsilon_2^{\mathrm{C}} - \varepsilon_{02}^{\mathrm{C}}\right),\\
F_1 &= 1 + 2\left(1 + \frac{1}{\varepsilon_2^{\mathrm{N}}}\right)\left(\frac{\varepsilon_1^{\mathrm{N}}}{\varepsilon_2^{\mathrm{N}}}\right)^2 \exp\left(\frac{154}{T}\right)\\
&\quad + \frac{[\mathrm{CO_2}]}{[\mathrm{N_2O}]}\left(\frac{\varepsilon_2^{\mathrm{C}}}{\varepsilon_2^{\mathrm{N}}}\right)^2 \exp\left(\frac{113}{T}\right)\\
&\quad \times\left[1 + 2\left(\frac{\varepsilon_1^{\mathrm{C}}}{\varepsilon_2^{\mathrm{C}}}\right)^2\left(1 + \frac{1}{\varepsilon_2^{\mathrm{N}}}\right)\exp\left(\frac{189}{T}\right)\right],\\
F_2 &= 1 + \left(\frac{\varepsilon_3^{\mathrm{C}}}{\varepsilon_3^{\mathrm{N}}}\right)^2 \frac{[\mathrm{CO_2}]}{[\mathrm{N_2O}]}\exp\left(\frac{180}{T}\right),\\
\varepsilon_1^{\mathrm{C}} &= \left[\left(1 + \frac{1}{\varepsilon_2^{\mathrm{N}}}\right)^2 \exp\left(\frac{302}{T}\right) - 1\right]^{-1},\\
\varepsilon_2^{\mathrm{C}} &= \left[\left(1 + \frac{1}{\varepsilon_2^{\mathrm{N}}}\right) \exp\left(\frac{113}{T}\right) - 1\right]^{-1},\\
\varepsilon_3^{\mathrm{C}} &= \left[\left(1 + \frac{1}{\varepsilon_3^{\mathrm{N}}}\right) \exp\left(\frac{180}{T}\right) - 1\right]^{-1}.
\end{aligned} \tag{6}$$

Here, $2\varepsilon_{02}^{\rm N}$ and $2\varepsilon_{02}^{\rm C}$ are the equilibrium average numbers of deformation mode quanta at temperature T (in kelvins) and $k_{\rm eff} = k_{3\rm N}^{\rm C} + k_{3\rm C}^{\rm N}\exp(180/T)$. The numbers in the equations and expressions have the following meaning: 300 is the room temperature (in kelvins); 154 and 76 are the resonance energy defects (in kelvins) of the intramodal exchange $\upsilon_1^{\rm N} \leftrightarrow 2\upsilon_2^{\rm N}$ and $\upsilon_1^{\rm C} \leftrightarrow 2\upsilon_2^{\rm C}$; 113 and 180 are the resonance energy defects (in kelvins) of the intramodal exchange $\upsilon_2^{\rm C} \leftrightarrow \upsilon_2^{\rm N}$ and $\upsilon_3^{\rm C} \leftrightarrow \upsilon_3^{\rm N}$; $189 = 76 + 113$; $302 = 76 + 2\times 113$; $67 = 180 - 113$; 3340 is the ratio of the radiation energy quantum for the highest intensity line of the HF laser to the heat capacity of the monatomic gas (in kelvins); 0.06 is the value of $\varepsilon_2^{\rm N}$ ($\varepsilon_2^{\rm C}$) at room temperature; 0.37 and 0.4 are the ratios of the symmetric mode quantum energy to the energy of the HF laser radiation quantum for N_2O and CO_2 molecules; 0.92 and 0.96 are the ratios of double the deformation mode quantum energy to the symmetric mode quantum energy for N_2O and CO_2; 1.73 and 1.69 are the ratios of the asymmetric mode quantum energy to the symmetric mode quantum energy for N_2O and CO_2; and $1.43 = c_V/c_V^{\rm He} - 1$ (where c_V and $c_V^{\rm He}$ are the heat capacities of N_2O (CO_2) and helium). The reciprocal process in Eqn (1) was disregarded during the derivation of the equations since its inclusion leads only to an insignificant correction (less than 0.5%) in the results of calculations. The quantitative data on the relaxation of N_2O in collisions with N_2O and He molecules were borrowed from [13], the data on the relaxation of CO_2 in collisions with CO_2 and He molecules were borrowed from [14, 15], and the data on $k_{\rm eff}$ were taken from [14].

The system of equations (3) – (6) using these spectroscopic and relaxation parameters satisfactorily explains the experimental results [1] on the vibrational pumping of N_2O [2]. This allows us to use this system for predicting the parameters of the N_2O and N_2O – CO_2 active media in the ultra-high-power lasers. In this study, we have calculated the values of ε_2 and ε_3, the specific energy $E_{\rm t}$ (in J L^{-1}) stored at all possible inverse transitions between asymmetric and symmetric modes, the specific energy E (in J L^{-1}) stored at transitions forming the amplification band of the USP radiation near the frequency of the master oscillator, as well as the gains of the active medium depending on the radiation energy density F (J cm^{-2}) of the HF laser. These quantities were calculated at the instant of attainment of maximum population inversion in the active medium.

In our calculations, we took into account the following lines of the HF laser:

$$\rm P_{1-0}\,(6)\,(1.5\,\%),\ P_{1-0}\,(7)\,(5.5\,\%),\ P_{1-0}\,(8)\,(6\,\%),$$

$$\rm P_{1-0}\,(9)\,(5\,\%),\ P_{1-0}\,(10)\,(2.5\,\%),\ P_{1-0}\,(11)\,(1\,\%),$$

$$\rm P_{2-1}\,(5)\,(2\,\%),\ P_{2-1}\,(6)\,(5.5\,\%),\ P_{2-1}\,(7)\,(11.5\,\%),$$

$$\rm P_{2-1}\,(8)\,(7\,\%),\ P_{2-1}\,(9)\,(5\,\%),\ P_{2-1}\,(10)\,(5\,\%),$$

$$\rm P_{2-1}\,(11)\,(2\,\%),\ P_{2-1}\,(12)\,(2\,\%),$$

$$\rm P_{3-2}\,(4)\,(1\,\%),\ P_{3-2}\,(5)\,(2\,\%),\ P_{3-2}\,(6)\,(3.5\,\%),$$

$$\rm P_{3-2}\,(7)\,(5\,\%),\ P_{3-2}\,(8)\,(5\,\%),\ P_{3-2}\,(9)\,(2.5\,\%),$$

$$\rm P_{4-3}\,(5)\,(2\,\%),\ P_{4-3}\,(6)\,(3\,\%),$$

where the fractions of the laser radiation energy per line are given in percent [16]. The main contribution to the optical pumping of N_2O molecules comes from the lines $\rm P_{1-0}\,(10)$, $\rm P_{1-0}\,(11)$, $\rm P_{2-1}\,(7)$, $\rm P_{2-1}\,(8)$, $\rm P_{2-1}\,(9)$, $\rm P_{2-1}\,(10)$, $\rm P_{3-2}\,(4)$, $\rm P_{3-2}\,(5)$, $\rm P_{3-2}\,(6)$, $\rm P_{3-2}\,(7)$ (43.5 % of the laser energy), while the main contribution to the pumping of CO_2 molecules comes from the lines $\rm P_{1-0}\,(6)$, $\rm P_{1-0}\,(7)$, $\rm P_{1-0}\,(8)$, $\rm P_{1-0}\,(9)$, $\rm P_{2-1}\,(5)$, $\rm P_{2-1}\,(6)$ (25.5 % of the laser energy). Note that distribution of the radiation energy over the HF laser lines can be modified by introducing admixtures (e.g., SF_6 or C_2F_5) accelerating the rotational relaxation of HF molecules [17] and/or using a selective resonator separating the individual spectral lines [3]. The laser pulse was assumed to be triangular in shape with rise and fall times of 0.7 and 0.3 μs, respectively (the latter value is approximately the same as in [16]).

3. Amplification bands of USPs

The following peculiarities of the vibrational energy distribution under optical pumping of N_2O and CO_2 molecules should be taken into account in an analysis of amplification bands. It was mentioned above that symmetric and deformation modes are excited simultaneously with the asymmetric mode during pumping. Equilibrium sets in rapidly between the former modes due to quasi-resonance exchange $10^00 - 02^l0$ ($l = 0,\ 2$). According to calculations, the population of the 020 state amounts to 1 % – 9 % of the ground state population depending on the pump energy density (1 – 10 J cm^{-2}). Since the state 020 is triply degenerate while the state 10^00 is not degenerate, the population of the latter state is no more than one third of that of the former state. Hence the states $\upsilon_1\upsilon_2\upsilon_3$ with a nonzero value of υ_1 are scarcely populated. If the average number of quanta ε_3 in the asymmetric mode is not too large, only the first few levels are found to be noticeably populated – $0\upsilon_2^l\upsilon_3$ (00^01, $00^02, \ldots$ and 01^11, $01^12, \ldots$). The shifts of bands corresponding to transitions from levels $00^0\upsilon_3$ and associated with anharmonism are insignificant. For example, the centres of the bands $00^01 - 10^00$, $00^02 - 10^01$ and $00^03 - 10^02$ in N_2O lie at $k = 938.85$, 936.56 and 934.12 cm^{-1}, respectively. Bands of transitions from level $01^1\upsilon_3$ are also characterised by a slight shift relative to one another (for example, the centres of the bands $01^11 - 11^10$ and $01^12 - 11^11$ lie at $k = 918.02$ and 915.72 cm^{-1}, respectively); however, these bands are shifted by ~ 20 cm^{-1} relative to transitions from levels $00^0\upsilon_3$. Hence, R-branches of the bands $01^1\upsilon_3 - 11^1(\upsilon_3 - 1)$ are superimposed on the P-branches of the bands $00\upsilon_3 - 10^0(\upsilon_3 - 1)$.

As a result, two amplification bands are formed (Fig. 1). The first of these bands corresponds to the sum of P-branches of the transition bands $00\upsilon_3 - 10^0(\upsilon_3 - 1)$ and R-branches of the transition bands $01^1\upsilon_3 - 11^1(\upsilon_3 - 1)$ and has a peak near 930 cm^{-1}, while the second band is equal to the sum of the R-branches of the transition bands $00\upsilon_3 - 10^0(\upsilon_3 - 1)$ and has a peak near 950 cm^{-1}. Transitions $10^0\upsilon_3 - 20^0(\upsilon_3 - 1)$ ($\upsilon_3 = 1, 2, \ldots$) whose upper states are populated in processes $00\upsilon_3 - 10^0\,(\upsilon_3 - 1)$ ($\upsilon_3 = 2, 3, \ldots$) contribute towards the amplification of USP radiation in the absence of collisions. Centres of the first few bands in the system $[10^0\upsilon_3 - 20^0(\upsilon_3 - 1)]$ lie at $k = 917.49$ ($10^01 - 20^00$), 915.91 ($10^02 - 20^01$) and 914.11 cm^{-1} ($10^03 - 20^02$). In other words, the R-branches of this system of bands contribute to the first amplification band.

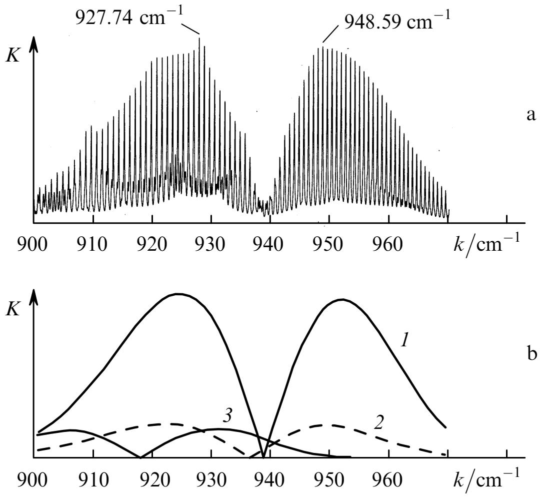

Figure 1. (a) Calculated gains K of the N_2O active medium and (b) their envelopes in the 00^01-10^00 bands [curve (*1*)], 00^02-10^01 [curve (*2*)] and 01^11-11^10 [curve (*3*)] making the main contribution to the formation of the USP amplification band. The total pressure of the mixture is 1 atm, the pressure $p_{N_2O}=0.2$ atm, the diluent gas is He, and $F=4$ J cm^{-2}.

Centres of the bands in the system $20^0\upsilon_3-30^0(\upsilon_3-1)$, $11^1\upsilon_3-21^1(\upsilon_3-1)$ $(\upsilon_3=1,2,\ldots)$ lie in the range of wavenumbers $k<896$ cm^{-1}, and these bands make zero contribution to the amplification of USPs: the spectral width of the input pulse of duration 10^{-12} s is 20 cm^{-1}. In the subsequent process of saturated amplification, the pulse duration decreases to $(0.7-0.8)\times10^{-12}$ s [4], which corresponds to the amplification band broadening to $\sim25-28$ cm^{-1}. The band centres in the system $0\upsilon_2\upsilon_3-1\upsilon_2(\upsilon_3-1)$ $(\upsilon_2>1,\ \upsilon_3=1,2,\ldots)$ lie at $k<902$ cm^{-1}; the contribution of these bands to the amplification of USPs can also be neglected, especially if we take into account the relatively low population of the states $0\upsilon_2\upsilon_3$ $(\upsilon_2>1)$.

Thus, the gains at the instant of the maximum population inversion were calculated by considering the $00^0\upsilon_3-10^0(\upsilon_3-1)$, $01^1\upsilon_3-11^1(\upsilon_3-1)$ $(\upsilon_3=1,2,\ldots)$ transitions, while the stored energies E_{930} and E_{950} (for $k\sim930$ and 950 cm^{-1}) were calculated by considering the 00^01-10^00 transition, the $00^0\upsilon_3-10^0(\upsilon_3-1)-20^0(\upsilon_3-2)$ $(\upsilon_3=2,3,\ldots)$ cascade transition, and the $01^1\upsilon_3-11^1$ (υ_3-1) $(\upsilon_3=1,2,\ldots)$ transitions. Transitions in the CO_2 molecule analogous to the above transitions in the N_2O molecule were used for the N_2O-CO_2 active medium for calculating the gains and the energies stored in the states contributing to the amplification bands. Centres of the first few bands in the systems $00^0\upsilon_3-10^0(\upsilon_3-1)$, $10^0\upsilon_3-20^0$ (υ_3-1) and $01^1\upsilon_3-11^1(\upsilon_3-1)$ for CO_2 lie at $k=960.96$ (00^01-10^00), 958.55 (00^02-10^01), 955.89 (00^03-10^02), 917.64 (10^01-20^01), 917.82 (10^02-20^01), 916.26 (10^03-20^02), 927.16 (01^11-11^10), and 925.08 cm^{-1} (01^12-11^11). Note that the bands $02^0\upsilon_3-12^0(\upsilon_3-1)$ $(\upsilon_3=1,2,\ldots)$ with centres at $k=941.7$ cm^{-1} (02^01-12^00), 937.38 cm^{-1} $(02^02-12^01),\ldots$ virtually coincide with the bands $00^0\upsilon_3-10^0(\upsilon_3-1)$ $(\upsilon_3=1,2,\ldots)$ for N_2O. The R-branches of the bands $03^1\upsilon_3-13^1(\upsilon_3-1)$ $(\upsilon_3=1,2)$ with centres at $k=908.37$ (03^11-13^10) and 905.2 cm^{-1} (03^12-13^11), as well as the bands $04^0\upsilon_3-14^0(\upsilon_3-1)$ $(\upsilon_3=1,2)$ with centres at $k=911.14$ (04^01-14^00) and 905.9 cm^{-1} (04^02-14^01) partially overlap with P-branches of the bands $00^0\upsilon_3-10^0$ (υ_3-1) $(\upsilon_3=1,2,\ldots)$. However, if we consider the fact that the populations of the levels $02^0\upsilon_3$, $03^1\upsilon_3$ and $04^0\upsilon_3$ are lower than the population of the levels $02\upsilon_3$, $03\upsilon_3$ and $04\upsilon_3$ by factors of 3, 4 and 5, respectively, the contributions of these bands to the amplification of the USP radiation can be neglected.

4. Results of calculations and discussion

Figures 1 and 2 show the calculated gains of the N_2O and N_2O-CO_2 active media at the instant of the maximum inversion (under the conditions of our calculations, this approximately corresponds to the instant of the pump pulse termination). The highest gain in the medium N_2O falls at the wavenumber 927.74 cm^{-1} and is associated with the lines P(13) $(00^01-10^00$, 927.74 cm$^{-1})$ and R(11) $(01^11-11^10$, 927.78 cm$^{-1})$, the linewidth at half-amplitude being ~0.08 cm^{-1} under a pressure of 1 atm. The highest gain in the N_2O-CO_2 medium is associated mainly with the lines R(10) $(00^01-10^00$, 947.8 cm$^{-1})$, R(13) $(00^02-10^01$, 947.78 cm$^{-1})$ in N_2O, P(16) $(00^01-10^00$, 947.74 cm$^{-1})$ in CO_2 and corresponds to 947.76 cm^{-1}. The highest gain is larger in the N_2O-CO_2 medium due to a contribution from the high-intensity line P(16) (00^01-10^00).

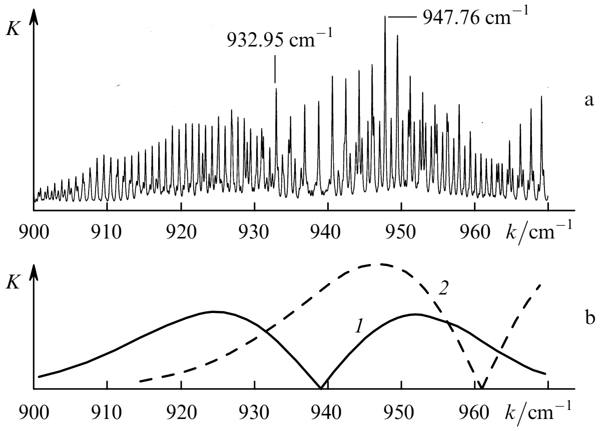

Figure 2. (a) Calculated gains K of the N_2O-CO_2 active medium and (b) their envelopes in the 00^01-10^00 bands for N_2O [curve (*1*)] and CO_2 [curve (*2*)], making the main contribution to the formation of the USP amplification band. The total pressure of the mixture is 1 atm, the pressure $p_{N_2O}=0.2$ atm, $p_{CO_2}=0.2$ atm, the diluent gas is He, and $F=4$ J cm^{-2}.

The energy stored in the amplification section of cross-sectional area S, length L and specific energy E is equal to ESL. If $K_{\max}$ is the maximum gain of the active medium due to pumping and $K_{\max}L$ is the admissible gain factor in the section ($K_{\max}L\leqslant10$ [4] and is determined by stability to the amplified spontaneous radiation and uncontrollable feedbacks), the limiting length of the section L is $\sim1/K_{\max}$ and the stored energy is $\sim E/K_{\max}$. In addition to E, the quantity $E/K_{\max}$ is an important parameter of the amplification system since it defines the limiting radiation energy in the saturated amplification mode. The factor of the HF laser radiation energy conversion into the energy stored in the amplification band of USP radiation is proportional to E/F.

Figure 3 shows the dependences of K_{max} as well as ε_2 and ε_3 for the N_2O active medium on the pump energy density F. Figure 4 shows the dependences of the quantities E_t, E_{930} and E_{950} (while calculating the two latter quantities, the spectral width of the amplification band was assumed to be equal to 30 cm^{-1}), E/K_{max} and E/F on F for the active medium N_2O; here, E was set as the highest value of E_{930} and E_{950} for a given radiation energy density of the HF laser. One can see from Figs 3 and 4 that an increase in the pump energy density leads to the following effects: the values of K_{max}, E_t, E_{930}, E_{950} and E/K_{max} increase; the increase in E_{930} prevails over the increase in E_{950}, and the dependence of E/F on F has a peak. The band with a peak at $k \sim 930$ cm^{-1} is preferred for amplifying a short pulse in the N_2O active medium. A sharper increase in the energy stored in this band as compared to the band with a peak at $k \sim 950$ cm^{-1} is due to contribution from transitions $01^1v_3 - 11^1(v_3 - 1)$ $(v_3 = 1, 2, \ldots)$. The frequency corresponding to $k = 927.74$ cm^{-1} should be used for amplification. A considerable part (60 % – 80 %) of the entire stored specific energy E_t is concentrated in the amplification band.

An important factor in the explanation of the nature of the dependence of E/K_{max} on the pump energy density F is that the coefficient K_{max} is determined by random resonance of two individual vibration – rotation transitions from different vibrational levels [P(13) $(00^01 - 10^00)$ and R(11) $(01^11 - 11^10)$]. The number of states contributing to the specific energy E but making zero contribution to K_{max} increases with F. Hence the ratio E/K_{max} must increase with F (in the absence of resonance, the increase would have been more pronounced). This is the main reason behind an increase in E/K_{max}. An increase in the translation – rotation temperature with F is a less significant reason behind the variation of E/K_{max}. The value of E/K_{max} increases slightly due to transformation of the rotational distribution.

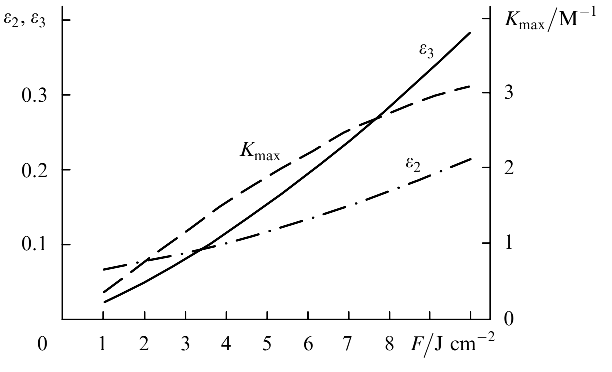

Figure 3. Dependence of the average numbers of quanta in the deformation (ε_2) and asymmetric (ε_3) modes, as well as of the maximum gain K_{max} in the N_2O active medium, on the pump energy density. The total pressure of the mixture is 1 atm, the pressure $p_{N_2O} = 0.2$ atm, the diluent gas is He.

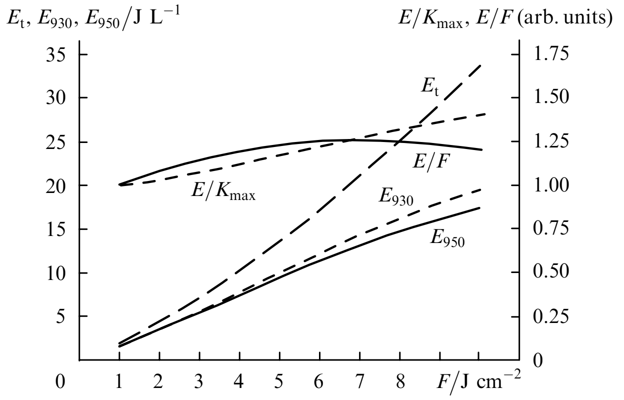

Figure 4. Dependences of E_t, E_{930}, E_{950} (the spectral width of the amplification band is 30 cm^{-1}), E/K_{max} and E/F on the pump energy density. The total pressure of the mixture is 1 atm, the pressure $p_{N_2O} = 0.2$ atm, and the diluent gas is He.

It was found in calculations that upon an increase in the pump energy density from 1 to 10 J cm^{-2}, the temperature increases from 304 to 367 K. The variation of E/K_{max} due to maximum heating under calculation conditions does not exceed 5 %. The optimal efficiency of HF laser radiation energy conversion into the energy stored in the amplification band of USP radiation is attained for $F \sim 7$ J cm^{-2}. A decrease in efficiency upon a further increase in F is due to a continuous increase in the population density of the states not participating in the amplification of the USP radiation. For typical pulsed chemical HF lasers, $F = 3 - 5$ J cm^{-2}. One can see from Fig. 4 that the value of E/F for such values of F does not differ much from the optimal value. In other words, this range of F can be used for pumping. In this range, each HF laser radiation quantum absorbed in the active medium corresponds to 0.65 quantum of energy that can be converted into the energy of USP radiation.

Figure 5 shows the dependence of the quantities E_t, E_{930} and E_{950} (while calculating the two latter quantities, the spectral width of the amplification band was assumed to be equal to 30 cm^{-1}) and E/K_{max} on the partial pressure of CO_2 in the N_2O – CO_2 – He mixture; as in the case of the N_2O medium, E is the highest among the values of E_{930} and E_{950} for a given F. It can be seen that in the medium N_2O – CO_2, all specific energies increase with CO_2 pressure in the mixture. At the same time, the values of E/K_{max} decrease with increasing CO_2 pressure. The quantity K_{max} is mainly determined by the random resonance in vibration – rotation transitions R(10) $(00^01 - 10^00)$ in N_2O and P(16) $(00^01 - 10^00)$ in CO_2.

The population density of vibrational states in the $00^01 - 10^00$ band in CO_2 is twice as large as in N_2O. Consequently, the population densities of states with $j = 15$

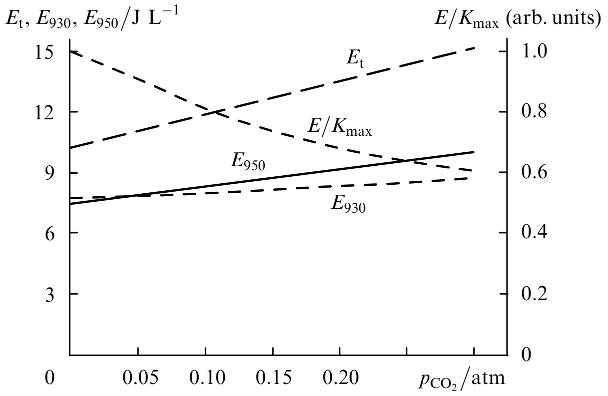

Figure 5. Dependences of E_t, E_{930}, E_{950} (the spectral width of the amplification band is 30 cm^{-1}) and E/K_{max} on the CO_2 pressure. The total pressure of the mixture is 1 atm, the pressure $p_{N_2O} = 0.2$ atm, the diluent gas is He, and $F = 4$ J cm^{-2}.

(CO_2) and $j = 11$ (N_2O) differ by a factor of about three at a temperature close to 320 K (calculations lead to a value close to 320 K for all mixtures with CO_2). Moreover, the cross section of the P(16) transition in CO_2 is larger than the cross section of the R(10) transition in N_2O by a factor of 1.36. This results in a strong dependence of $K_{\max}$ on the CO_2 concentration. Consequently, in spite of the fact that the addition of CO_2 increases the specific energy stored in the medium, the real energy obtained in the section with saturated amplification decreases due to a decrease in its admissible length. The use of the $N_2O - CO_2$ mixtures is expedient in the case when the maximum increase in the factor of conversion of the HF laser energy into the energy of USP radiation is required.

Note that it is possible to increase the energy extracted from the active medium N_2O. For two carrier frequencies corresponding to the wavenumbers 927.74 and ~ 683 cm^{-1} (or ~ 709 cm^{-1}), the following scheme of transitions can be realised:

$$00^01 - 10^00 - 01^10,$$

$$00^02 - 10^01 - 20^00 - 11^10 - 02^20,$$

$$00^03 - 10^02 - 20^01 - 11^11 - 02^21,$$

$$01^11 - 11^10 - 02^20,$$

$$01^12 - 11^11 - 02^21.$$

The transitions $10^00 - 01^10$ (694.14 cm^{-1}), $20^00 - 11^10$ (683.07 cm^{-1}), $20^01 - 11^11$ (668.85 cm^{-1}), $11^10 - 02^20$ (702.52 cm^{-1}) and $11^11 - 02^21$ (688.84 cm^{-1}) (the band positions are shown in the parentheses) form an additional amplification band with maxima at ~ 683 and ~ 709 cm^{-1}. In the absence of collisions, the population densities of the upper states of these transitions are determined by the population densities of the initial states $00^0\upsilon_3$ and $01^1\upsilon_3$ ($\upsilon_3 = 1, 2, \ldots$) at the instant of attainment of the maximum inversion. It can be shown that the fraction of the additional energy is $\sim(694/939)[1 - \varepsilon_2(1 + \varepsilon_3)/(1 + \varepsilon_2)\varepsilon_3]$. One can see from Fig. 3 that this fraction is equal to zero for $F \approx 3.5$ J cm^{-2} (since $\varepsilon_2 = \varepsilon_3$) and increases with the pump energy density. For $F = 10$ J cm^{-2}, this fraction amounts to ~ 30 %.

5. Conclusions

We have calculated the pumping of the $N_2O - He$ and $N_2O - CO_2 - He$ mixtures at a total pressure of 1 atm by pulses from a multifrequency HF laser. Two amplification bands of USPs at ~ 930 and ~ 950 cm^{-1} are formed as a result of pumping of N_2O molecules. The specific energy stored during pumping is higher in the 930-cm^{-1} band. For an HF laser radiation energy density of ~ 5 J cm^{-2}, the specific energy amounts to ~ 10 J L^{-1}. Each HF laser radiation quantum absorbed in the active medium corresponds to 0.65 quantum of energy that can be converted into the USP energy. The output energy of the section with saturated amplification increases with the pump energy density. However, starting from a certain pump level (~ 7 J cm^{-2}), the efficiency of the section decreases due to a continuous increase in the population of the states making zero contribution to the amplification of the USP radiation. From the practical point of view, the range of pump energy densities 3 – 5 J cm^{-2} is admissible. For the $N_2O - CO_2$ mixture, the 950-cm^{-1} band is preferable for amplification of USPs. The specific energy stored in this mixture is higher than in the N_2O mixture, but the energy stored in the amplification section is lower on account of a smaller admissible length of the section. The use of the $N_2O - CO_2$ mixtures is expedient when the maximum conversion coefficient of the HF laser radiation energy into the USP energy is required.

It is shown that the energy extracted from the N_2O active medium can be increased by using additional amplification bands at ~ 683 and ~ 709 cm^{-1} related to transitions from the intermediate states appearing due to transitions forming the main amplification bands.

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