NOTATION

p, v, T, pressure, volume, and temperature; R, universal gas constant; $\mu_{N_2O_4}$, molecular mass; C_v, C_p, isochoric and isobaric specific heats; Z, compressibility coefficient; $C_{p_{ef}}$ effective specific heat at constant pressure; Z_{ef}, effective coefficient considering effects of dissociation and nonideality; $K_{p_{10}}$, $K_{p_{20}}$, temperature dependent equilibrium constants for first and second reaction stages for an ideal gas mixture; α_{10}, α_{20}, degree of dissociation of first and second reaction stages in ideal gas state.

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CHARACTERISTICS OF A GAS-DISCHARGE CO-LASER IN GENERATION ON AN OVERTONE

1. STEADY-STATE REGIME

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The work presents the parametric dependences of the energy and spectral characteristics of the gas-discharge CO-laser in steady-state regime in generation on the first overtone on the gas temperature, the pumping power, and the cavity Q-factor of the resonator. The article examines questions of the selection of the individual vibrational and rotational lines. Comparisons with experimental results are made.

At present there is considerable interest in lasers on vibrational overtone of the carbon monoxide molecule [1-4]; this is due to the potentially high effectiveness of such a laser close to the IR range and the possibility of using it for problems of laser chemistry, isotope separation, etc. We will also demonstrate that the investigation of this laser makes it possible to elaborate on the kinetics of CO molecules, especially at high vibrational levels. Knowledge of the theoretical parametric dependences obtained on the basis of numerical modeling makes it possible to predict the expected characteristics of such a laser, and it stimulates and facilitates its experimental investigation. The present work consists in the theoretical investigation of the characteristics of the gas-discharge CO-laser operating in steady-state regime of generation on the first overtone. We analyze the parametric dependences of the energy and spectral characteristics on the gas temperature, the pumping power, and the cavity Q-factor of the resonator. We also examine questions of the selection of individual vibrational and rotational lines.

Previously, we substantiated the possibility of obtaining generation on vibrational overtones of carbon monoxide molecules with pumping in a stationary and pulsed electric discharge [1]. An analysis of the population density of vibrational levels of CO molecules indicated that it is possible to effect partial inversion at the transitions J—1, $v \to J$,

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v—k not only for $k=1$ but also for $k\geq 2$, and at the first overtone the gain according to the calculation exceeded 0.1 m^{-1} with parameters of the discharge and of the gaseous medium close to those obtained in the experimental work [5]. It also followed from the results of the calculations that the gain of the small laser signal at the first overtone is maximal at high vibrational transitions. In this the laser on a overtone differs markedly from a laser on the fundamental frequency where the gain of the small signal rapidly increases with increasing number of the vibrational level, after which the maximum is attained and a drop begins. This result is basically due to the different nature of the dependences of the Einstein coefficient vs the number of vibrational levels on the fundamental frequency and on overtones. The authors of [1] also investigated the maximum values of the gain on the fundamental frequency and on the first two overtones vs the gas temperature and the pumping power. These dependences are analogous for all three types of transition, but on each subsequent overtone the gain decreases by more than one order of magnitude.

The conclusions arrived at in [1] also began to receive experimental confirmation. At the same time as [1], [2] was also published where generation on the first overtone in a supersonic stream with previous pumping of the vibrational levels in an electric discharge was experimentally obtained. Later on, generation on an overtone was also obtained in a pulsed discharge [3]. We will show that the calculation of the generation spectrum on the first overtone in a pulsed electric discharge was carried out in [4] on the assumption that there is no generation on the fundamental transitions. In the mentioned experimental works the authors did not succeed in suppressing generation on the fundamental frequency, and it occurred together with generation on the first overtone. Such an operating regime is investigated in detail below.

To find the distribution function of the CO molecules according to vibrational levels and the generation spectrum on the vibrational and rotational transitions, here we carried out the numerical solution of a system of kinetic equations for the population densities of the levels by a method described in [6]. The kinetic equations have the form

$$\frac{dn_v}{dt} = R^v_{eV} + R^v_{VV} + R^v_{VT} + R^v_A + g_2^{v+2,v} I_2^{v+2,v} - g_2^{v,v-2} I_2^{v,v-2} = 0, \tag{1}$$

where R^V_{eV}, R^V_{VV}, R^V_{VT}, and R^V_A are terms describing the change in the population densities of the vibrational levels due to excitation and deexcitation by electron shock, VV of the exchange, VT of relaxation and spontaneous emission. Equations (1) take into account forced emission on the first overtone, $I_2^{V,V-2}$ is the flux density of the radiation quanta at the transition $v\to v-2$; $g_2^{V,V-2}$ is the gain on this same transition. The gain is calculated by the formula

$$g_2^{v,v-2} = \frac{\lambda^2}{8\pi} A_{v,v-2} J \left[n_v \frac{B_v}{T} e^{-\frac{B_v (J-1) J}{T}} - n_{v-2} \frac{B_{v-2}}{T} e^{-\frac{B_{v-2} J (J+1)}{T}} \right] G(\nu), \tag{2}$$

where λ is the wavelength of the corresponding transition; $G(\nu)$, normalized profile of the line; J, rotational quantum number of the lower level. The energy passing into the molecular vibrations was determined by the numerical solution of the Boltzmann equation for the energy distribution function of electrons [7]. The Einstein coefficients for the corresponding radiational transitions were taken in accordance with the analysis carried out in [8]. To find the distribution functions of the CO molecules according to vibrational levels where generation occurs on the transitions between them, the following condition was used:

$$g_2^{v,v-2} = \Gamma, \tag{3}$$

where Γ is the threshold gain determined by the losses in the resonator. To find the radiation intensity at these transitions, Eq. (1) was used. In the process of solution, the gain $g^{v,v-v_2}$ was chosen maximal with respect to J, the number of the rotational transition $J-1\to J$. In the kinetic equations, temperature figures as a parameter, and it is determined by the pumping power and the removal of heat. In our calculations we confined ourselves to the examination of conditions close to those obtained in [5].

Figure 1 shows the distribution functions of carbon monoxide molecules according to vibrational levels in the gain regime, in generation on the first overtone and on the fundamental frequency. The conditions of the calculation are as follows: density of the carbon monoxide molecules $n_{CO} = 8.5\cdot 10^{16}$ cm^{-3}, of the helium molecules $n_{He} = 8.8\cdot 10^{7}$ cm^{-3}, gas temperature $T = 100°K$, density of the pumping power $W = 1.5$ W/cm^3, reduced electric field intensity $E/N = 1.3\cdot 10^{-16}$ $W\cdot cm^2$.

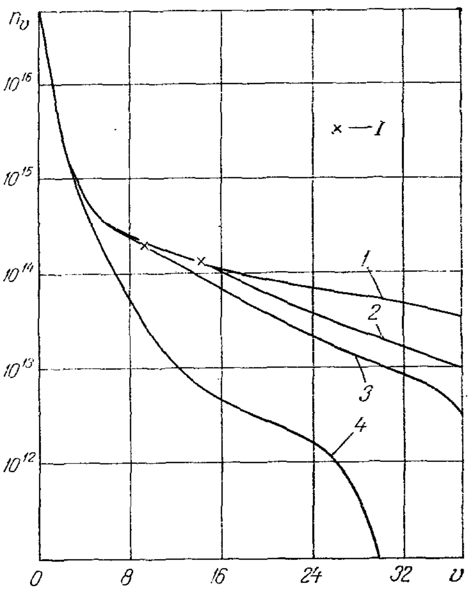

Fig. 1

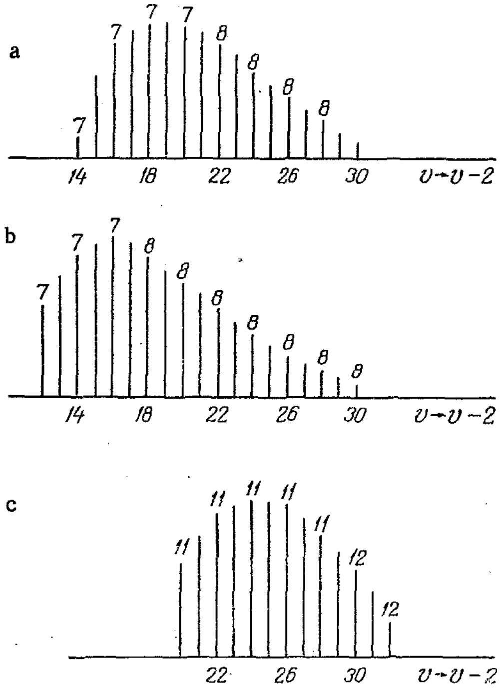

Fig. 2

Fig. 1. Distribution functions of CO molecules according to vibrational levels: 1) in gain regime with generation on the first overtone; 2) $\Gamma = 10^{-3}$ cm^{-1}; 3) $5 \cdot 10^{-4}$ cm^{-1}; 4) in generation on the fundamental frequency, $\Gamma = 10^{-3}$ cm^{-1}; I) position of the beginning of the generation spectrum n_v, cm^{-3}.

Fig. 2. Spectra of generation on the first overtone: a) T = 80°K, W = 1 W/cm³; b) T = 80°K, W = 2 W/cm³; c) T = 160°K, W = 2 W/cm³. $n_{CO} = 1.8 \cdot 10^{17}$ cm^{-3}, $n_{He} = 1.8 \cdot 10^{18}$ cm^{-3}, $\Gamma = 10^{-3}$ cm^{-1}.

The gain on the transition of the first overtone is substantially smaller than on the fundamental transitions. With the same losses of the resonator, the excess of the generation threshold is also considerably smaller. In consequence, the effectiveness of transformation of the pumping energy into radiation is low. As a result, the distribution function of the molecules according to levels in generation on an overtone is considerably closer to the distribution function without generation than to the distribution function in generation on the fundamental transitions. When the losses in the resonator are reduced, the effectiveness of energy transformation increases, and the distribution function decreases more at high levels.

The spectra of generation on the first overtone are shown in Fig. 2. The relative magnitude of the generation power on each vibrational transition is proportional to the height of the corresponding line, and above each line the rotational quantum number of the lower level of the corresponding transition is indicated. At low gas temperature (T = 80°K) the origin of the generation spectrum lies at the level v = 12–14. This fact fits in well with the experimental data of [2] although they were obtained with generation in a resonator mounted transversely to a stream of previously excited gas; this does not correspond to the conditions of our calculations. However, the origin of the generation spectrum with steady-state distribution function of the population density of the levels is determined by the margin of vibrational energy of the molecules, the gas temperature, and the magnitude of the threshold gain, and not by the actual pumping conditions and the direction of motion of the active medium in the resonator. The mentioned correlation of the origin of the generation spectrum with the experimental results must therefore be considered satisfactory.

When the pumping power is increased, the origin of the generation spectrum, and largely also its maximum, are shifted to lower levels. Under the conditions of pumping in a stationary discharge that are close to those experimentally realized in [5], it is impossible to

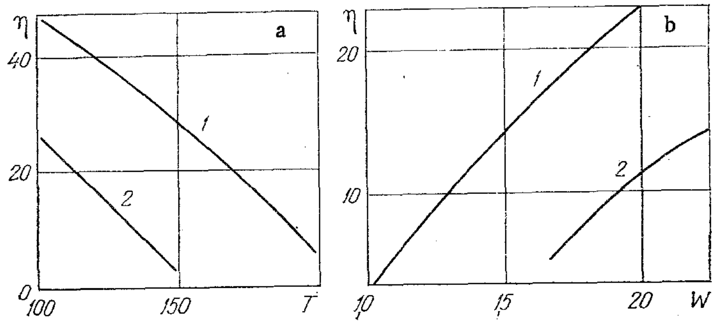

Fig. 3. Dependence of the generation efficiency on the first overtone on the gas temperature (a) (1) $\Gamma = 5 \cdot 10^{-4}$ cm^{-1}; 2) 10^{-3} cm^{-1}, $W = 1$ W/cm^3) and on the pumping power (b) (1) $T = 150$°K; 2) 175°K, $\Gamma = 10^{-3}$ cm^{-1}). The other conditions are the same as in Fig. 2. η, %.

lower the gas temperature to 80°K. Under these conditions the gas temperature is 140-180°K. In this case the generation spectrum originates at much higher levels: $v = 20$-22, and its maximum occurs at the level $v = 24$-26. At higher temperatures the rotational quantum numbers of the generation lines are larger. It should be noted that the longwave boundary of the generation spectrum depends weakly on the temperature and the pumping power, and that it lies at the level $v = 30$-32. We also point out that the generation spectrum on the first overtone, in consequence of the proximity to the threshold, proves to be very broad. In the calculations it was assumed that the magnitude of the losses of the resonator is the same at all transitions. Yet in the frequency band of generation there may be changes in the transparency of the windows and in the reflection factor of the outlet mirror, of the absorption in air or in the active medium; this may undoubtedly affect the generation spectrum. Some of these factors may exert a particularly great influence in view of the fact that, in order to suppress generation on the fundamental frequencies, it is necessary to place frequency-selective elements in the resonator.

Figure 3a shows the change in efficiency of transformation of the pumping energy into radiation in dependence on the gas temperature, Fig. 3b shows the dependence on the density of the pumping power. The magnitude of the transformation efficiency is the maximum generation efficiency if there are no internal losses in the resonator. In a real case, when the loss coefficient upon passage through the resonator in two directions is A, and the coupling coefficient of the outlet mirror is θ, so that $\Gamma = (A + \theta)/2L$, L is the length of the resonator, the generation efficiency is $\theta/2L\Gamma$ times smaller than the transformation efficiency. In view of the small gain of the laser on an overtone in steady-state regime and the possible losses in suppressing generation on the fundamental transitions, this ratio may be fairly small.

The gas temperature, the density of the pumping power, and the magnitude of the threshold gain have a very strong effect on the effectiveness of transformation. Independent checking of the gas temperature and of the pumping power is possible only within fairly narrow limits, and the threshold gain will not be very small. We can therefore barely expect that the effectiveness of transformation will exceed 10%.

With regard to the practical application of the laser, of interest is its operation in the regime of selecting a certain generation line. Figure 4a shows the generation spectrum without selection of lines. For the conditions of the calculation we have: $n_{CO} = 1.8 \cdot 10^{17}$ cm^{-3}, $n_{He} = 1.8 \cdot 10^{18}$ cm^{-3}, $T = 150$°K, $W = 1$ W/cm^3, $\Gamma = 5 \cdot 10^{-4}$ cm^{-1}, the transformation efficiency is 28%. Figure 4 shows for the same conditions how the transformation efficiency changes in dependence on the vibrational transition of the first overtone on which the generation line separates, the generation line corresponding to the maximum gain according to the rotational quantum number. The transformation efficiency in the selection regime decreases but the power emitted on one line increases noticeably in comparison with free generation, of course, when the threshold gain is the same. This increase is particularly great in the longwave region of the generation spectrum. The curves of Fig. 4b correspond well to the distribution of the gain according to levels, and not to the generation spectrum

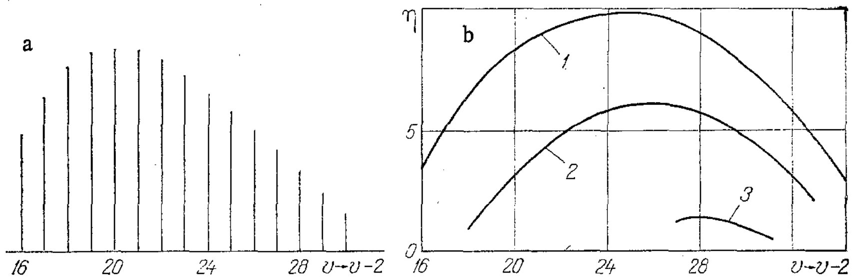

Fig. 4. Generation characteristics: a) spectrum without selection of lines, b) dependence of the generation efficiency on the position of the selected line (1) $\Gamma = 5\cdot10^{-4}$ cm^{-1}; 2) $7\cdot10^{-4}$ cm^{-1}; 3) 10^{-3} cm^{-1}).

without selection. The short-wave boundary of the spectrum coincides in both cases but the maximum and the longwave boundary of the spectrum in selection correspond to higher transitions. However, since with selection, additional losses are inevitably introduced into the laser resonator, the effectiveness of transformation may be considerably reduced and the range of separated lines become smaller.

On the whole, the calculation results presented above indicate that a stationarily acting convective CO laser with pumping in gas-discharge tubes may operate on the transitions of the first overtone of carbon monoxide molecules with an efficiency that is of practical interest. To improve the efficiency, it is necessary to work with lower temperatures which are attainable only with adiabatic cooling in a widening supersonic gas stream. Laser operation in such a regime requires a special investigation.

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