

Application of a distributed gold catalyst to increase the output power and efficiency of a sealed-off CO₂ laser excited by a transverse rf discharge in tubes

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Abstract. It is shown experimentally that a transverse rf discharge in tubes has a higher plasmachemical activity than a dc discharge, which deteriorates the parameters of rf discharge-excited CO₂ lasers. The output power and efficiency of a sealed-off CO₂ laser were substantially increased by using a distributed gold catalyst facilitating CO₂ regeneration. The output power of 0.77 W cm⁻¹ per unit length of an active medium with efficiency of 18.5% was obtained in an experimental CO₂ laser with tubes with a distributed gold catalyst excited by a 27.1 MHz rf discharge.

Keywords: sealed-off CO₂ laser, rf discharge in tubes, CO₂ dissociation, distributed gold catalyst.

1. Introduction

Along with the use of increasingly high rf frequencies (exceeding 100 MHz) for pumping compact slab CO₂ lasers with diffusion cooling of the active medium [1–3], there also exists the opposite trend [4–6] relating to the prospects of developing relatively inexpensive high-power rf semiconductor oscillators operating at frequencies up to 40 MHz.

To obtain an acceptable lasing efficiency at comparatively low rf excitation frequencies f , it is necessary to increase the interelectrode distance d [3]. Otherwise, near-electrode layers that are unfavourable for lasing, whose thickness d_s can be estimated from the relation $fd_s \approx 40$ MHz mm, will occupy a considerable part of the active volume. An increase in the interelectrode distance deteriorates the conditions of diffusion cooling of the active volume, and the slab geometry loses its advantages. Indeed, gas cooling in the interelectrode gap occurs only in the direction of the water-cooled electrodes, while cooling in a tube or a channel of square cross section is radial, i.e., about 1.5 times more efficient.

Note also that increasing the electrode spacing or the tube diameter d renders the use of a waveguide cavity

inexpedient. To provide the efficient suppression of higher-order transverse modes in a waveguide cavity of length L , the Fresnel number $N_F = d^2/(4\lambda L)$ (where λ is the wavelength) should satisfy the condition $N_F < 1/\pi$. To obtain this condition, the length L should increase proportionally to d^2 with increasing d , which complicates the obtaining of a homogeneous rf discharge along the resonator [1–3, 7]. In this connection, it is of interest to study the parameters of a CO₂ laser with diffusion cooling and a transverse rf discharge in tubes with an open cavity.

Transverse rf discharges in tubes of various diameters for pumping CO₂ lasers have long been studied, but the results have been ambiguous [1, 7–11]. Obviously, this is due to the fact that an rf discharge may exist under these conditions in two forms: the so-called low-current or α form and the high-current or γ form. These two forms of the discharge have substantially different structures and parameters of the near-electrode layers. This difference becomes decisive in lasers with diffusion cooling, in which the interelectrode distance is comparable with the total thickness of the near-electrode layers, and the layers themselves cover a considerable part of the surface confining the discharge volume. For a number of reasons, the high-current γ form is unsuitable for lasers with diffusion cooling, although it is used quite successfully in lasers with a convectively cooled active medium [1–3].

Another important feature of the rf discharge, first noticed by the authors of [12–15], is its enhanced activity with respect to the decomposition of CO₂, which may be the main reason behind the decrease in the laser power. This can be due to the processes occurring in the near-electrode regions, which are distributed over the entire length of the discharge volume in the transverse rf discharge. This problem can be solved by using a catalyst facilitating the CO₂ regeneration in the near-electrode regions of the discharge. The action of a gold catalyst distributed over the surface of the tube in a dc discharge was analysed in detail in [16]. However, conditions in the near-electrode regions of the transverse rf discharge differ from those in the dc discharge because the near-electrode layer plasma is not quasi-neutral and is characterised by high electron and ion energies.

The aim of this paper is to study the possibilities of developing an efficient sealed-off CO₂ laser with diffusion cooling excited by a transverse 27.1-MHz rf discharge in a dielectric tube. We paid special attention to increasing the output power and efficiency of the laser by stabilising the chemical composition of the laser mixture with the help of a

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gold catalyst distributed over the surface of the discharge tube for the CO₂ regeneration.

2. Model experiment

First we constructed a small model laser for studying the operation of various configurations of electrodes and selecting the best among these. This laser was also used to refine the process of catalyst deposition by determining the required density of the coating and its stability under rf discharge conditions.

Figure 1 shows the scheme this laser. It consists of a quartz tube with an inner diameter of 13 mm. The discharge part is 60 cm long. Transformer oil flowing through the jacket on the outer side of the tube was used for cooling. The resonator consisted of a rear spherical copper mirror with the radius of curvature 4 m and a reflectivity of 98 %, and a plane ZnSe output mirror with a reflectivity of 90 %. The discharge was fed by a 2-kW rf generator operating at a frequency of 27.1 MHz.

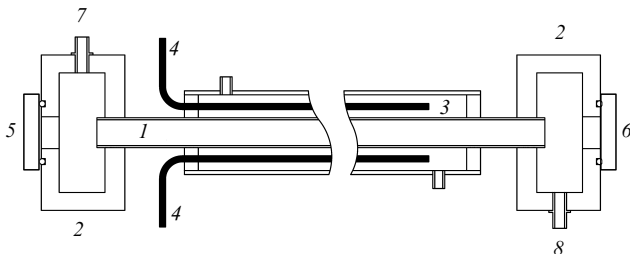


Figure 1. Scheme of the model laser: (1) discharge volume; (2) adjusting blocks; (3) cooling jacket; (4) buses for supplying rf current to electrodes; (5, 6) optical resonator mirrors; (7, 8) connecting pipes for supplying and pumping out the laser mixture.

We selected the electrode configuration based on considerations presented in monograph [1], where it was suggested that upon pumping a diffusely cooled laser by an rf discharge having the γ type near-electrode layers, it is desirable to use electrodes with the smallest possible area to suppress the so-called thermal screening of the tube volume by the γ type near-electrode layer characterised by a considerable heat release directly at the inner wall of the tube adjoining the layer. It was assumed that, despite a local input of current into the tube volume, diffusion processes may result in a more or less uniform filling of the tube cross section by the discharge plasma. A laser with such a configuration of electrodes was also described in [11].

We tested several configurations of electrodes. Three of them and the corresponding shapes of the discharge emission are shown in Figs 2a–c. The electrodes are made of copper tubes of diameter 2.5 mm pressed tightly to the discharge tube from the outer side. One can see that none of these configurations provided a sufficiently uniform filling of the tube cross section by the discharge, which contradicts to the data presented in [1, 11]. Such a discrepancy is probably due to a smaller diameter of the discharge tubes used in our experiments. In all cases, the radiation power did not exceed 10 W. The best results were obtained with wide electrodes, whose construction is shown in Fig. 2d. In this case, a greater part of the tube was uniformly filled by the positive discharge column. The V-

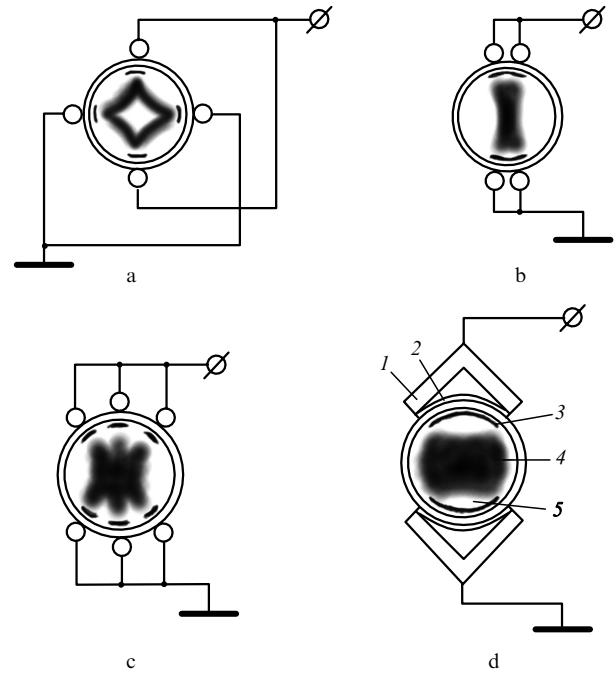


Figure 2. Various schemes of arrangements of rf electrodes: (a) electrode system formed by four tubes, (b) pairwise arrangement of tubes, (c) triple tubes, and (d) system with electrode made of foil; (1) V-shaped duralumin elements; (2) aluminium foil; (3) glowing near-electrode regions; (4) positive column plasma; (5) dark near-electrode regions.

shaped duralumin element in this construction simultaneously plays the role of an rf current conductor and an element providing the rigidity of the structure in the longitudinal direction, while the electrodes are aluminium foil strips tightly abutting the discharge tube wall.

Experiments on generation of laser radiation were performed for a model with the electrode system shown in Fig. 2d, both in the mixture exchange and in sealed-off regimes. The same composition of the gas mixture CO₂ : N₂ : He : Xe = 1 : 3 : 16 : 1 was used in both cases. The relatively high content of xenon enabled the reduction of power losses in the near-electrode layers. The radiation power was measured with a PW-250 Synrad power meter. The power supplied to the discharge was measured by the method of cooling liquid calorimetry taking the output laser power into account.

Figure 3 shows the dependence of the radiation power on the rate of exchange of the mixture. The highest radiation power was 42 W and the maximum efficiency was 10 %. The dependences of the radiation power and efficiency on the rf power supplied in the mixture exchange and sealed-off regimes are shown in Fig. 4. In both cases, the output power achieved the maximum at the input power of about 400 W. However, in the sealed-off regime, the output power did not exceed 22 W and the efficiency was below 7 %. Note that the reflectivity of the output mirror used in the experiments (90 %) is not optimal for the small discharge length (60 cm) mentioned above. This explains the low lasing efficiency and the considerable difference between the maximum powers obtained in the sealed-off and mixture exchange regimes.

The decrease in the output power on passing to the sealed-off regime suggests that the gain decreases due to the decomposition of CO₂. The change in the mixture compo-

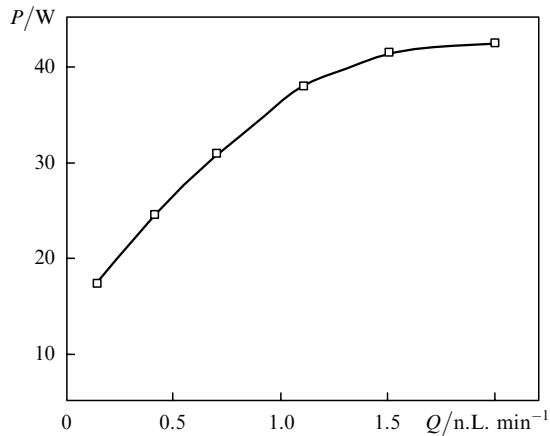


Figure 3. Dependence of the radiation power P on the rate of exchange Q of the mixture in a model laser with a discharge length 60 cm (n.L. stands for normal litres).

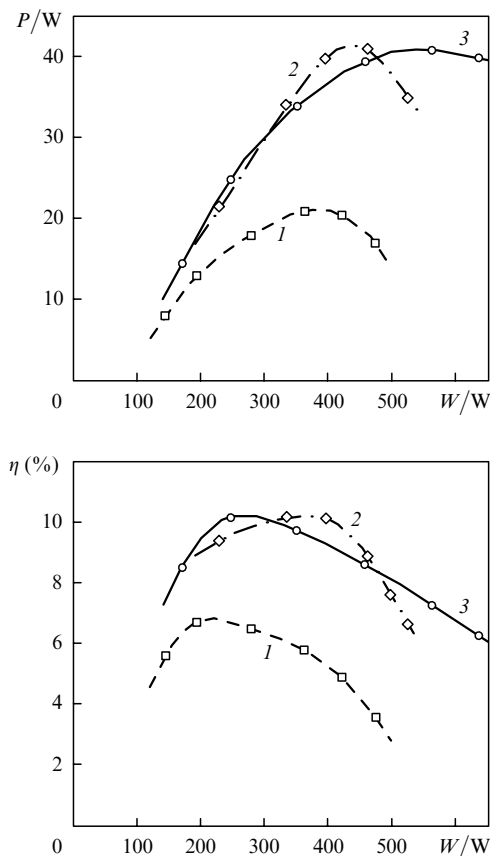


Figure 4. Dependences of the radiation power P and efficiency η on the input rf power W , obtained in model experiments for (1) operation without a catalyst in the sealed-off regime and (2) the gas exchange regime ($Q = 2$ n.L. min⁻¹), as well as (3) operation with a catalyst.

sition in the sealed-off regime is also confirmed by the change in the colour of the positive discharge column which was rosy in colour in the mixture exchange regime, especially near the entrance of the gas into the discharge. On passing to the sealed-off regime, the colour turned to bluish white almost instantaneously through the entire length of the discharge. These observations necessitated the use of a catalyst for the CO₂ regeneration.

Note that a considerably high rate of mixture exchange is required to stabilise the radiation power for such a short discharge (Fig. 3). The rate of exchange of 1.5 n.L. min⁻¹ (n.L. stands for normal litres, i.e., the amount of gas occupying a volume of one litre under normal conditions) corresponds to the velocity of more than 6 m s⁻¹ for the gas moving in the discharge region and to the gas average residence time in the discharge region of less than 0.1 s. For dc discharge lasers with the working mixture composition stabilised by slow circulation, the gas residence time in the discharge region (see, for example, [16, 17]) is about an order of magnitude longer, i.e., about 1 s. This is explained by the basic difference between the structures of the transverse rf discharge and dc longitudinal discharge. The near-electrode layers, in which the conditions strongly differ from those in the region of the positive column, are distributed over the entire length of the transverse rf discharge. It seems that it is the processes occurring in the near-electrode layers that determine the plasmachemical processes in the rf discharge.

In our opinion, the nature of the near-electrode layers in the rf discharge observed in model experiments plays a decisive role in understanding the operation of a laser in the investigated range of parameters. The specific density of the input power achieved 7.5 W cm⁻³, which corresponds to a discharge current density of more than 40 mA cm⁻² (the experimental and theoretical data on discharge parameters under identical conditions can be found, for example, in [18]). This exceeds the characteristic current density for the transition from the α to γ form in a discharge with metal electrodes, which amounts to about 20 mA cm⁻² in the case under study [1, 3]. The emission pattern in the discharge cross section (see Fig. 2d) is also reminiscent of a high-current discharge characterised by the presence of a dark space between emission near the electrodes and in the bulk.

Apparently, the transition from the α to γ form of discharge is observed in this case, when the secondary emission processes in the near-electrode layers are so strong that the emission structure typical of the γ form is observed, but a dielectric wall between the plasma and the metal electrode prevents the contraction of the near-electrode layer, and the discharge preserves its spatial homogeneity as a whole. The enhanced plasmachemical activity of the rf discharge also suggests that the near-electrode layers have properties similar to those of the γ -discharge layers. From the point of view of electrical engineering, the near-electrode layers of the α discharge are more like a capacitor and are characterised by a low dissipation of the rf power. Therefore, the plasmachemical properties of the α form of an rf discharge should not differ strongly from those of a dc discharge.

Thus, because the enhanced plasmachemical activity of the γ form is caused by near-electrode processes, the use of a distributed catalyst may be especially efficient for a discharge laser of the type considered here.

3. Catalyst

In the experiments with a catalyst, the coating was deposited on the inner wall of the discharge tube. The method of deposition by cathode sputtering of individual regions of length 20 cm is described in [16], but the authors of this work point to the possible inhomogeneity of the coating obtained in this way. In view of this, the coating in

our work was deposited directly on the entire length of the tube occupied by the discharge. The sputtering process was monitored visually. As a result, a semitransparent greyish blue layer was obtained, and the colour eventually changed to rose under the action of the discharge.

In [16], a quite complicated procedure for preliminary activation of the catalyst is described. Our experiments showed that there was no need for such a procedure in the case of a sealed-off laser. After introduction of the fresh mixture, the catalyst was activated by the rf discharge for 10 – 15 s. The activation process could be visualised by a change in colour of the positive discharge column from bluish white to rose, as well as by an increase in the laser output power. The activity of the catalyst was preserved during operation with the same mixture.

Figure 5 shows the regions of the plasma emission spectrum for a transverse rf discharge in a quartz tube with and without a catalyst. The bluish white colour of the discharge plasma without a catalyst is caused by an increase in the concentration of CO molecules due to the dissociation of CO₂ molecules, which is accompanied by an increase in the B¹Σ – A¹Π emission intensity of CO molecules in the bands located at 451.1, 483.2 and 519.4 nm.

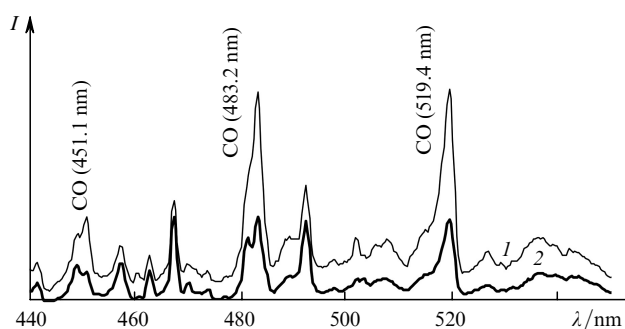


Figure 5. Comparison of the plasma radiation spectral regions of transverse rf discharge in a quartz tube (1) without a catalyst and (2) with a catalyst.

In some cases, the use of a catalyst led to a short-circuiting of the discharge current through the coating. A partial removal of the coating from the tube walls prevented such a short-circuiting but reduced the area of the coating and hence lowered its efficiency.

It was found in the experiments that a moderately dense catalyst layer virtually does not conduct current, displaying, however, a rather high catalytic activity. In this case, the coating is formed which consists of isolated microscopic gold crystals in which atoms are coupled through a strong metallic bond. The average thickness of such a coating is larger than the thickness of a continuous metallic gold layer for the same amount of the deposited material. Our experiments showed that an appreciable conduction appeared when the average coating thickness (determined from the dependence of the coating conductivity on its thickness measured by multiple-beam interferometry [19]) was 30 nm and more, while no conduction was observed for the thickness smaller than 20 nm.

On the other hand, it is known that crystals of size of a few nanometres have the highest catalytic activity towards oxidation of carbon monoxide to carbon dioxide even in the absence of excited oxygen molecules or atoms [20]. There-

fore, the cathode sputtering technique can be employed for depositing nonconducting coatings with a high catalytic activity with respect to the CO₂ regeneration, which can be used in transverse rf discharges.

Figure 4 shows the dependences of the laser output power and efficiency on the rf power supplied to the discharge for a tube with and without a catalyst. These dependences are qualitatively similar to those presented in [16, 21] for a dc discharge. In both cases, the output power is almost doubled when the catalyst is used, and the parameters for the sealed-off regime are almost identical to those obtained for the mixture exchange regime (see Fig. 4). The sealed-off regime differs from the mixture exchange regime in that the maximum power is achieved for a larger energy input, in agreement with the results obtained in [16, 21].

The test of the coating stability to the discharge action for tens of hours did not reveal any change in the appearance or catalytic activity of the coating.

An increase in the activity of a gold catalyst observed with increasing the temperature of one of the discharge channel walls to 50 °C was reported in some papers (see references in [13]). We measured the dependence of the output power of the model laser on the temperature of a cooling oil in the temperature range 15 – 75 °C. The results are presented in Fig. 6. One can see that, as the temperature is increased to 45 °C, the output power changes weakly and rapidly decreases as the oil temperature is further increased.

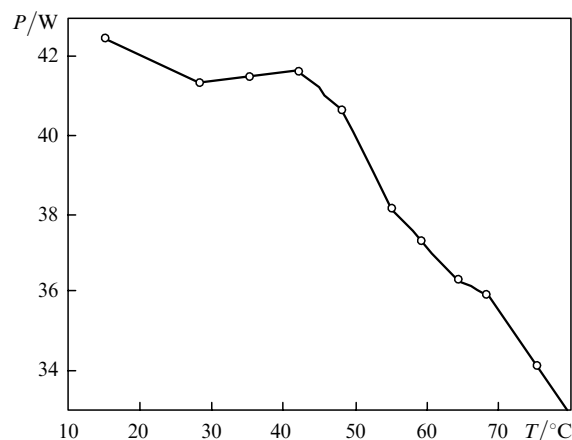


Figure 6. Dependence of the radiation power P in the sealed-off regime with a catalyst on the temperature T of the cooling liquid.

4. Experimental laser

The results obtained in model experiments were used to design and build an experimental model of a 400-W sealed-off laser. The scheme of this laser and its external view are shown in Figs 7 and 8, respectively. Four rigidly connected quartz tubes were enclosed in a square steel casing. 120-cm-long electrodes of the type shown in Fig. 2d were fastened along the tubes. The catalyst was deposited on the inner walls of the tubes over the entire length of the electrodes. The ends of the tubes protruding from the casing were tightened in duralumin mirror blocks. The mirror blocks were fastened to the flanges of the casing through positioning inserts.

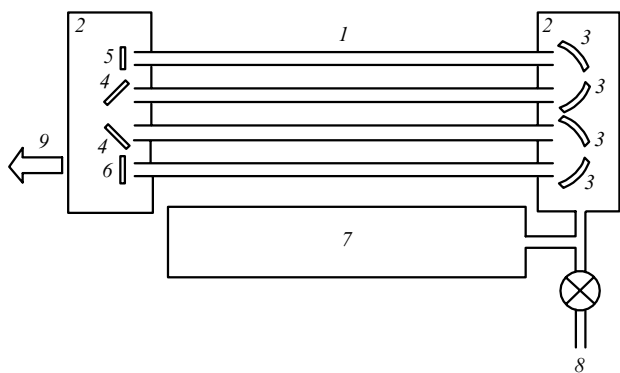


Figure 7. Scheme of the 400-W experimental laser and series-connected discharge tubes: (1) discharge tubes; (2) mirror block; (3) fold mirrors for focusing and correction; (4) plane deflecting mirrors; (5) rear mirror; (6) output mirror; (7) ballast volume; (8) to the evacuating system and gas inlet; (9) radiation output.

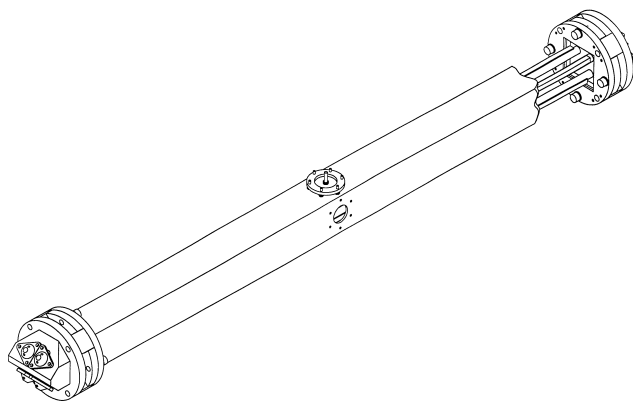


Figure 8. General view of the experimental laser. Part of the casing near the flange on the right side is removed to reveal the arrangement of discharge tubes.

Both ends of the grounded electrodes were connected to each other and to the lead on the casing via copper buses. The electrodes to which the rf voltage was applied were insulated from each other, each having its own lead in the middle of the casing (Fig. 8). The service life of the sealed-off laser was increased by connecting a ballast volume (6 L) to the discharge volume. The discharge tubes were cooled by a flow of transformer oil circulating through the inner cavity of the casing at a rate of 17 L min^{-1} . They were connected to the rf generator through matching resonance LC circuits. By choosing the inductances appropriately, these circuits were tuned in such a way that the discharge current in all the tubes was the same and the reflected power was minimum.

We employed the same rf generator that was used in the model experiment. The generator power was 2 kW at a frequency of 27.1 MHz. The laser power achieved the maximum for an optimal matching of the generator and the load, when the maximum power was supplied by the generator to the load. The power supplied to a unit volume of the active medium for the maximum input power of 2 kW was about 3 W cm^{-3} , which is about half the optimal value obtained in the model experiment for tubes with the catalyst. The optimal effective current density 25 mA cm^{-2} of the rf discharge, which is typical for such a specific power input, corresponds to the anomalous burning regime of a

weak-current rf discharge. At the same time, the transverse structure of the discharge emission observed from the end-faces of the tubes was similar to that observed in the model experiment (see Fig. 2d).

The optical cavity calculation was performed for the actual construction of a 400-W laser. The negative thermal lens was taken into account by assuming that for the rated output power of the laser, about 3 kW of input rf power in the form of heat is released in the rf discharge and this heat is carried to the walls by heat conduction. Four discharge sections of length 120 cm each were combined in a single cavity with the help of fold mirrors mounted at an angle of 45° to the axis of the tubes and reflecting the radiation successively from one tube to the next (see Fig. 7). For a total length L of the cavity equal to 5.8 m and an inner diameter 13 mm of the tube, the Fresnel number $N_F = 0.7$. For tubes of such a diameter, the open cavity regime ($N_F > 1$) can be obtained for $L < 2.75 \text{ m}$, and the wave-guide regime ($N_F < 0.3$) is realised for $L > 11 \text{ m}$.

The scheme shown in Fig. 9 was used to obtain the stable open-cavity regime taking into account compensation for the concave thermal lens formed in the discharge sections. The radiation in each tube propagated, as in a stable nearly semi-confocal resonator (taking the negative thermal lens into account), while the resonator as a whole consisted of four identical stable resonators connected in series. Output mirror (1), rear mirror (5), and fold mirrors (4) were plane.

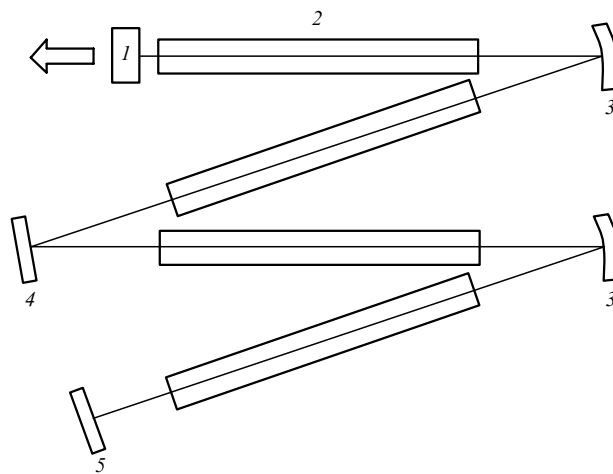


Figure 9. Full-length resonator diagram used for calculations: (1) partially reflecting output mirror; (2) discharge section; (3) two-mirror focusing element shown as a spherical mirror; (4) two-mirror deflecting element shown as of a plane mirror; (5) rear mirror, the half-length cavity is obtained by connecting the elements (1-2-3-2-5) [deflecting element (4) is replaced by plane rear mirror (5)].

The laser design also allowed lasing in two independent parallel resonators of half the length. In this case, rear mirror (5) was replaced by a second output mirror, and plane fold mirrors (4) (Fig. 7) were replaced by two plane rear cavity mirrors. As a result, two parallel resonators of half the length were obtained, each in its turn consisting of two series-connected stable resonators of the same type as in the case of full-length resonators. Because the parameters of the focusing mirrors were the same in both cases, the volume occupied by the radiation in each tube was also the same. A

comparison of the total output power of the two parallel resonators with the output power of the full-length resonator under identical conditions allowed us to estimate the cavity losses caused by optical distortions in the active medium.

Focusing elements (3) were made in the form of mirrors reflecting radiation at an angle of 45° as shown in Fig. 7. Cylindrical mirrors were used to correct the astigmatism produced by reflection from the focusing mirror at an angle of 45° . Certain difficulties were encountered during the adjustment of the resonator with cylindrical mirrors due to more stringent requirements on the accuracy of cylindrical axis alignment. The following method was used for final adjustment. A cylinder with crossed graduation marks inscribed at its end was inserted into the aperture of the rear mirror. The image of the cross was observed through the output mirror aperture. The distortions of this image could be removed completely by slight rotations of cylindrical mirrors.

Zinc selenide output mirrors with reflectivities of 30% and 50% were used. Both mirrors were tested in operation with the long resonator. The output laser power was nearly the same in both cases, which indicates that these two values of the reflectivity are close to the optimal value. Output mirrors with a reflectivity of 50% were used in short resonators.

The output power of each of the two parallel half-length resonators was 185 W, which corresponds to an energy extraction of 0.77 W cm^{-1} from a unit length of the active medium with an efficiency of 18.5%. The output power of the full-length resonator was 280 W (0.58 W cm^{-1}) for the 14% efficiency.

In the first approximation, the laser radiation structure in the output beam cross section is a ring about 10 mm in diameter, corresponding to the TEM_{01} mode of a stable open resonator. A more complex diffraction pattern, indicating that the output beam contains radiation reflected at the tube walls, is superimposed on the ring. The effect of tube walls on the lasing process points to the presence of scattered radiation in the resonator and the losses associated with it, which results in a decrease in the laser output power.

The role of optical distortions in the active medium can be judged by comparing the output power (280 W) of the full-length resonator with the total power (370 W) of two half-length parallel resonators. The volume and configuration of radiation in discharge tubes, as well as other conditions affecting lasing, were nearly identical in both cases, the only difference being that optical inhomogeneities of the active medium in the full-length resonator lead to accumulation of uncompensated wave front distortions of the beam, which causes an increase in the losses. We explain the effect of accumulation of uncompensated wave-front distortions by the fact that the concave thermal lens, which was assumed to be axially symmetric in calculations, had a cylindrical component associated with the structure of the transverse rf discharge.

5. Conclusions

The results obtained in this study confirm the possibility of producing an efficient CO_2 laser with a transverse rf discharge in a dielectric tube with an open optical resonator. The successful use of a distributed gold catalyst shows that a high plasmachemical activity of the rf

discharge in the investigated range of parameters may be the main reason of ambiguity in the parameters of such lasers. An increase in the specific energy input leads to the formation of transition type near-electrode layers characterised by a higher energy dissipation and plasmachemical activity than in the layers of a weak-current rf discharge. A catalyst, operating efficiently and stably under these conditions, allows a neutralisation of the harmful effect of the plasmachemical processes. In our opinion, this is the main result of our present study.

We have built the 400-W experimental laser with four discharge tubes of length 120 cm each. The laser was tested with a 2-kW rf generator whose power is not sufficient to obtain the maximum output power. Nevertheless, we managed to achieve quite high values of the parameters: an output power of 0.77 W per unit length (cm) of the active medium with an efficiency of 18.5%.

We have shown that the optical inhomogeneities of the active medium restrict the possibility of increasing the laser power simply by increasing the number of tubes. It seems that the nonuniform heat release in the transverse rf discharge is the main source of optical distortions. This problem can be technically solved by arranging the electrodes along a spiral or by reducing the length of individual discharge sections.

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