acta phys. slov. 39 (1989), No. 2

ON THE DISSOCIATION OF CO_2 IN THE WAVEGUIDE DISCHARGE LASERS[1)]

NOVÁK, M.,[2)] LÁSKA, L.,[2)] Prague

The limitation of lifetime of sealed-off cw waveguide discharge CO_2 lasers expressed by the decrease of the output power is predominantly due to an irreversible dissociation of CO_2 which causes a change in the discharge properties, by which the inversion of the laser levels deteriorates. In this contribution we present the results of the first measurements of the dissociation of CO_2 in connection with measurements of the output power. Furthermore, a construction of a quartz sealed-off CO_2 waveguide laser, whose output power of 2.9 W remained stable within 200 working hours, is reported.

I. INTRODUCTION

Waveguide CO_2 lasers are relatively small devices, characterized by a great energy density (hundreds of W/cm^2) of the laser beam in the continues-wave (cw) regime [1, 2]. This is made possible by a discharge with high current densities at higher pressure in a capillary made of a dielectric material, forming at the same time an optical waveguide. The waveguide together with the optical resonator support in most cases the EH_{11} transversal mode of the electromagnetic field so that up to 98 % of the total output energy can be radiated in this way [3—5]. The laser activity depends upon an inversion population between the vibrational level 00°1 and the levels 10°0 and 02°0 of the CO_2 molecule. The upper laser level is populated by electronic collisions and especially by the resonance transition of energy from the near lying level of the vibrationally excited N_2 molecule ($v = 1$). To the depopulation of the lower levels there mainly contribute V—T transitions between CO_2-molecules and He-atoms. In there collisions the He-atoms receive energy, which is then transported to the walls of the discharge tube. The high density of the active medium in the capillary presents a considerable energy dissipated in the collisions of the excited molecules and the temperature on the axis of the capillary reaches hundreds of °C [1].

[1)] Contribution presented at the 7th Symposium on Elementary Processes and Chemical Reactions in Low Temperature Plasma, STARÁ TURÁ-DUBNÍK, June 13—17, 1988

[2)] Institute of Physics, Czech. Acad. Sci., Na Slovance 2, 180 40 PRAGUE, Czechoslovakia

Therefore in order to reach a higher inversion population it is necessary to decrease the temperature of the mixture by an intensive cooling. This leads to a demand for a sufficiently thin and a highly thermally conductive capillary wall. In case of a compact construction, where the mirrors are mounted directly on the ends of the waveguide, it is advisable if the tube material has a minimum thermal dilatation.

In the particle balance of the electric discharge in the molecular gases, the role of the wall processes becomes more significant with the decreasing diameter of the discharge tube. For example, the recombination coefficient of the oxygen atoms can vary over 5 orders of magnitude depending on the kind, quality and temperature of the wall; in a similar way there changes also the volume density of the O-atoms in the discharge [6]. Immediately after the ignition of the discharge in CO_2 the dissociation proceeds with the time constant of about 1 s, expressed by the equation

$$CO_2 \rightleftharpoons CO + \frac{1}{2} O_2 \qquad (1)$$

and in the equilibrium the fraction of the dissociated CO_2 molecules may reach 60—70% [7]. A further decrease in the partial pressure of CO_2 is caused by the loss of oxygen due to the oxidation of the metal electrodes and due to an absorption on the sputtered metal film in their proximity. This process represents an effective sink for the oxygen slowing down the reaction (1) in the reversed direction and thus increasing the dissociation degree even further.

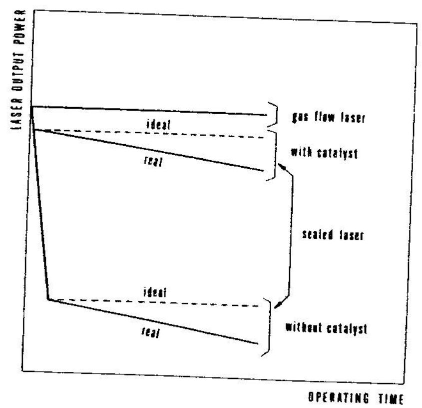

Fig. 1. Schematic plot of the lifetimes of different construction types of CO_2 laser with ideal and real electrodes.

This all leads to a decrease of the radiated laser power and influences negatively the life-time of the sealed-off systems.

Fig. 1 (after [7]) shows schematically the life-time of various construction types of CO_2 lasers. It is seen that the life-time can be increased by using suitable elctrodes and catalysts, which enhance the reverse recombination of CO_2. One of the possibilities is to use perovskite oxide cathodes with $Ln_{1-x}M_xM'O_3$ [7] (Ln — lanthanoid, M — alkalic earth, M′ — transition metal). Their advantage is a high catalytic activity for the oxidation of CO starting at the temperature of 200 °C; in contrast to the metal electrodes their sputtering rate is considerably lower, and instead of consuming the oxygen they release it.

II. EXPERIMENTAL ARRANGEMENT AND RESULTS

The construction of the cw waveguide laser, realized in the Gas Discharge Department of the Institute of Physics of the Czech. Acad. Sciences reported previously used either external mirrors [8, 9] or a diffraction grating [10] in a gas-flow regime. The maximum total output power of 2.9 W and the efficiency of 2.8% were obtained. In a closed system the power of 1—1.4 W with an efficiency of 3—4% was obtained during several hours only.

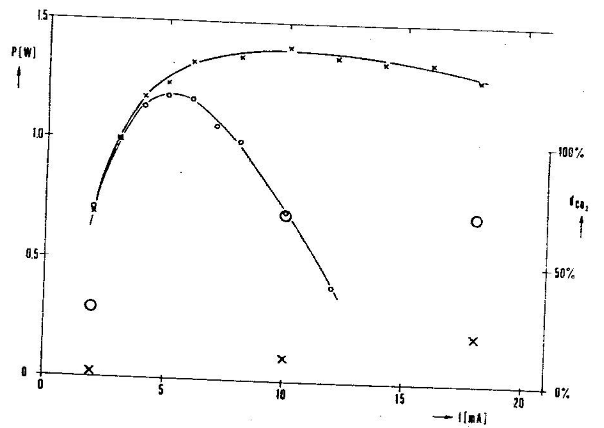

Fig. 2. The dependence of CO_2 waveguide laser output power (small marcs) and dissociation (large marcs) on the discharge current for two different residence times of particles in the system (o = 150 ms, x = 5 ms).

Since the dissociation degree can be modified either by changing the discharge current (with increasing current the dissociation increases) or by changing the supply of the fresh gas (in a gas flow regime), simultaneous measurements of the output lasser power and the dissociation degree were performed by using mas-spectrometer analysis (see also [9]). Fig. 2 shows the dependences of the laser power on the discharge current and simultaneously the dependence of the dissociation degree (great symbols) on the current for two flow rates of the

gas mixture, expressed in the total residence time of the molecule in the discharage system. The residence time of 150 ms is of about the same order as the time to reach the dissociation equilibrium, the residence time of 5 ms is much smaller. In spite of the fact that this dependence is complicated by the above mentioned dependence of the output power on temperature (and therefore on current), the importance of the disociation degree is evident.

Below, the first tests of the output power and the lifetime of a new construction of a sealed-off laser system are reported. This laser consists of a quartz capillary with the inner diameter of 2.7 mm and the length of 25 cm. It is water cooled and has an adjoint volume for the working gas mixture (about 300 cm^3). The resonator consists of two planar mirrors — a hard gold-coated copper mirror and a germanium one with interference coatings for the reflexivity of 90%. This Ge mirror is on the outer surface coated with an antireflex film and serves simultaneously as an output window. The mirrors are fixed directly on the discharge tube in the proximity of the ends of the waveguide capillary and they also seal same the discharge tube. The experiments were made with a DC input power from 25 to 125 W, a perovskite oxide cathode and in a mixture of $CO_2 : N_2 : He$ in the ratio of 1 : 1 : 4 at a total pressure 3—7.4 kPa either in the system closed by a valve, or — the long-life measurements — in the sealed-off system at the total pressure of 5 kPa.

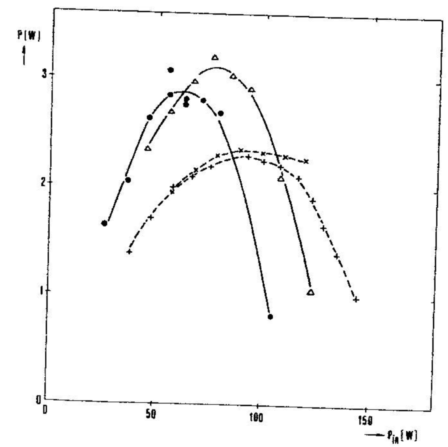

Fig. 3. The dependence of the CO_2 waveguide laser output power on the input power in the sealed-off (full line) and the gas-flow (dashed line) regime.: +, o — 5.3 kPa, ×, Δ — 7.4 kPa.

The stabilization of the laser power after the change of parameters (especially of the current) lasts due to the thermal equilibrium from about 5 to 10 minutes. Instability of the laser power within this time (see, for ex., the scattering of the experimental points in fig. 3) can be ascribed to an instability of the discharge, but more likely to a drift of the laser regime between diverse spectral lines (in

[10] a lasing on 40 lines was demonstrated) due to a change of the length of the resonator, caused by the thermal dilation of the discharge tube. The output laser power in dependence on the input discharge power for two total pressures (5.3 and 7.4 kPa, without gas flow) is presented in fig. 3. In comparison with the equivalent measurements of the gas-flow regime (in a slightly different configuration — [8]) both curves show more expressive maxima, which are shifted to a lower input power (a similar character of dependences is shown also in [7]). The corresponding efficiencies for the maximum power vary between 4.3 and 5.2%.

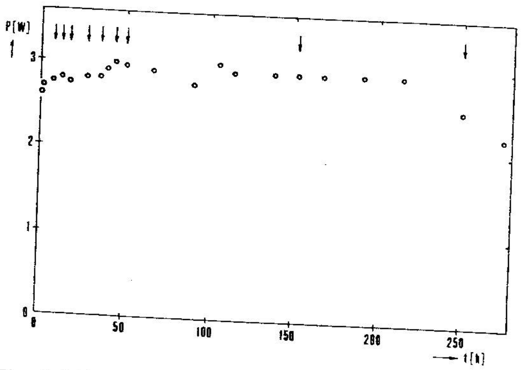

Fig. 4. Lifetime of the sealed-off CO_2 waveguide laser.

In fig. 4 the time dependence of the output power of the sealed-off system is shown. After an initial small increase the output power remained within the first 200 working hours stable on the level of 2.9 W (with occasional interruptions — see arrows in fig. 4). After further 150 hours the power decreased to 1.9 W. It follows from this that the used catalyst remarkably influences the dissociation equilibrium in the system. The limitation of the lifetime of the laser can be ascribed not only to a gradual decrease of the efficiency of the catalytic reaction on the cathode causing a disappearence of CO_2 from the mixture, but also to a degradation of the dielectric coatings on the output mirror, caused both by the dielectric losses in Ge and by the influence of the ambient atmosphere on the electrostatically charged window.

III. CONCLUSION

In the cw regime of the CO_2 waveguide laser the influence of the perovskite oxide cathode on the dissociation degree and consequently on the lifetime of the sealed-off system was tested with a good result. The output power 2.9 W of the laser with the beam diameter (Gaussian) of about 1.5 mm did not decrease noticably during 200 hours of operation. Even if these results are only preliminary, it is evident that this laser could be a suitable tool for a number of laboratory and technical applications. The results of a numerical solution to balancing the laser level populations, based on the Cohen model [11], should serve as an optimization of this CO_2 waveguide laser performance.

REFERENCES

[1] Dumitras Dan, C.: *Inter. Conf. and School Lasers and Applications.* Bucharest 1982, 483.
[2] Degnan, J. J.: Appl. Phys. *11* (1976), 1.
[3] Marcatili, E. A. J., Schmeltzer, R. A.: The Bell System Technical Journal (1964), 1783.
[4] Abrams, R. L., Bridges, W. B.: IEEE J. Quant. El. QE-9 (1973), 940.
[5] Conder, P. C., Redding, J. R., Jenkins, R. M.: Infrared Phys. *25* (1985), 61.
[6] Mašek, K., Láska, L.: Czech. J. Phys. B*30* (1980), 805.
[7] Iehisa, N., Fukaya, K., Matsuo, K., Karubè, N.: J. Appl. Phys. *59* (1986), 317.
[8] Novák, M., Urbánková, H.: *5th Conf. Lasers in Research and Industry.* Palkovice 1985, 53.
[9] Urbánková, H., Novák, M.: *Conf. Appl. Optics '87.* Praha 1987, 124.
[10] Urbánková, H.: Czech. J. Phys. B*37* (1987), 911.
[11] Mašek, K., Rohlena, K.: *VI. Symposium on Elem. Proc. and Chemical Reactions in Low-temperature plasmas.* Jelšava-Hrádok 1986.

Received July 13th, 1988
Accepted for publication October 18th, 1988

О РАСТВОРЕНИИ CO_2 В ВОЛНОВОДОВЫХ РАЗРЯДНЫХ ЛАЗЕРАХ

Ограничение на время жизни волноводовых разрядных лазеров, которое выражено уменьшением мощности на выходе, в основном определяется необратимым растворением CO_2, которое обуславливает изменения свойств разряда, разрушающих инверсионную заселенность уровней лазера. В предлагаемой работе приведены результаты первых измерений растворения CO_2 в связи с измерением мощности на выходе. Более того, сообщается о конструкции кварцевого волноводового лазера с мощностью 2,9 W, остающейся постоянной на протяжении 200 часов.