

Influence of nitrogen on CO-laser characteristics

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Abstract—The effect of a nitrogen addition on plasma discharge of CO lasers in plasma-chemical processes is discussed here. It is shown that a nitrogen addition improves the laser characteristics and changes the composition of a laser active medium. The reduction of CO highly-excited vibrational-states populations with a current is smaller in the case of mixtures with nitrogen additions. The addition of nitrogen significantly decreases the CO dissociation level and concentration of C atoms created in plasma-chemical reactions of a laser discharge.

The processes of molecular dissociation, following plasma-chemistry, changing the composition of a laser active-medium and laser characteristics, determine the long-term effective operation of molecular sealed-off lasers (working without a gas exchange in the laser discharge tube).

These processes are extremely important for the CO-laser operation. The vibrationally excited $\text{CO}(X^1\Sigma, v)$ molecules, involved in laser-radiation generation, hardly relax during collisions with the initial component of an active mixture, however the ratio of their relaxation could significantly increase when the collision partners are some products of the above mentioned plasma-chemical processes [1]. It is well known (see e.g. [2] and the references therein), that in the sealed-off CO-lasers usually mixtures with significant concentrations of nitrogen are applied, which supports better energy-characteristics and improves the long-life performance of the laser.

The addition of nitrogen molecules to the laser active-mixture results in an increase of population of vibrationally excited CO molecules due to vibrational-vibrational (VV') relaxation processes. However, there are some important indicators pointing to the fact that improvements of CO-laser operation after a nitrogen addition does not necessary result from the additional mechanism of CO states excitation. The addition of nitrogen to an He-CO mixture leads to a significant change in plasma-chemical products. Firstly, some new nitrogen-containing products appear: N atoms, molecules CN, NO, etc. Secondly, nitrogen and nitrogen-containing products influence the plasma composition of an initial He-CO mixture.

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Up to now the influence of nitrogen on plasma-chemistry of a CO laser has rarely been investigated. There is a lack of comprehensive models for plasma-chemistry of He-CO-N₂ mixtures (except some common feature with the plasma of a CO₂ laser [3-5]), which would help to estimate the level of degradation of the initial laser mixture. However, it has been shown in [5] that the CO₂ laser plasma with a high nitrogen content, $30\% < [\text{N}_2] < 50\%$, ensuring proper storage of vibrational energy in CO₂ lasers, is characterized by low electron temperature and thus, small chemical reactivity, i.e. a small level of CO₂ dissociation. From the investigations it follows that mixture decomposition decreases with an increase of N₂ content in CO₂ laser mixtures. In contrast, electron temperature does not decrease after nitrogen additions to He-CO plasma. The temperature is constant or even slightly increases (up to 15%) after nitrogen additions.

However, there is a small amount of experimental data on products of plasma-chemistry in such discharges and on products dependence on experimental conditions and discharge time-duration. The concentrations of CN molecules have been measured in flowing He-CO-N₂ discharges [6-8] and in sealed-off systems, and theoretically determined based on a model of plasma-chemical processes - see Ref. [9].

Thus, the aim of this work is to determine the influence of nitrogen additions to He-CO mixtures on CO-laser characteristics, including CO vibrational excitation. The experiments were performed under conditions typical for discharge tubes of CO-laser working in sealed-off and gas flowing regimes.

The experimental set-up has been already described in a previous work [9]. The water-cooled discharge tube from molybdenum glass, 17 mm in diameter, was used in the experiment. The lengths of a discharge zone was 50 cm. Hollow cylindrical electrodes from tantalum were installed in the side branches of the discharge tube at a distance of 4 cm from its axis. The discharge was excited using DC currents of 10-100 mA. The experiment was performed for initial He-CO and He-CO-N₂ mixtures. Relative CO and N₂ concentrations were varied in the range 3÷15%. The gas pressure was set at 5÷15 Torr. The setup can work in sealed-off and gas-flowing regimes (gas velocities 1÷3 m/s were applied). Initially, the gas

mixtures used in the experiments were carefully purified by using a system of traps with silica-gel and zeolite.

The emission spectra from the discharge positive-column were registered using a spectrometer in the range 300÷6000 nm, in order to determine the respective populations of vibrationally excited states of CO($X^1\Sigma$) and gas temperature along the symmetry axis of the discharge tube as well as its radial profile. The determination was done by analysing the rotational structure of CO vibrational bands.

The vibrational distribution of CO molecules in the ground electronic state ($X^1\Sigma$) was determined from the spectra of IR (infrared) molecular emission at the ($v \rightarrow v-1$) fundamental frequency, and the first ($v \rightarrow v-2$) overtone.

During the experiment the overtones emission was registered in the neighbourhood of the symmetry axis (~3 mm) in a discharge positive column. The spectra were studied using a Czerny-Turner monochromator with a diffraction grating 300 and 1200 lines/mm. Cooled (by liquid nitrogen) photoresistors HgCdTl and uncooled InSb were used as photodetectors.

The lack of cataphoresis significant influence during the measurements in systems without gas flows was proved. It was confirmed by registering He line intensities and CO molecular band at various points along the symmetry axis of the discharge tube using an optical waveguide.

The electric field in the discharge was determined by measuring the voltage drops across the discharge gap and the electrode sheaths. The electrode sheath voltages were found from the voltage measurements in experiments with different discharge lengths. A typical electrode sheath voltage was 350÷400V. The estimated value of the reduced electric field E/N was $(2.1 \div 3.2) \cdot 10^{-16}$ V cm².

Table 1. The CO vibrational temperature [K] under different conditions

| Pressure Current | 5 Torr | | 10 Torr | |
|----------------------------------|----------------------|-------|----------------------|-------|
| | 50 mA | 70 mA | 50 mA | 70 mA |
| He-CO= 95:5 | 1750/ 1700 | 1950 | 1700/ 1600 | 1800 |
| He-CO= 91:9 | 1700 | 1800 | 1700 | 1800 |
| He-CO-N ₂ = 95:3:2 | 2850/ 2700 | 2800 | 2700/ 2600 | 2750 |
| He-CO-N ₂ = 95:4:1 | 2450/ 2400 | 2500 | 2500/ 2400 | 2650 |

The measurements were performed for a CO-laser-generation regime and without laser-radiation generation. In order to enforce laser generation a resonator in a flat/spherical configuration was applied. A fully reflecting metallic mirror (steel covered with gold) had a 5 m curvature radius. The exit mirror (with transmission 85÷95%) was a flat plate of CaF₂ or ZnSe covered by a

dielectric multilayer. The laser energy characteristics under the gas-flow regime were: power 0.4 W and electro-optic efficiency $\eta = 10\%$; under sealed-off regime 0.2 W and $\eta = 4.8\%$, respectively. The resonator was not optimised for generation conditions.

The measurements showed that a nitrogen addition to the He-CO laser medium results in a substantial increase of CO vibrational temperatures. This should be related to the transfer of vibrational energy from nitrogen to carbon oxide due to the VV' exchange. As a result, a stationary state is realised in the laser medium, with a high level of vibrational energy in CO molecules.

Table 1 presents the results of vibrational temperature (T_1) determination:

$$T_1 = E_1 / \ln(N_0/N_1), \quad (1)$$

where E_1 – the energy of the first excited vibrational state; N_0 and N_1 – the populations of the ground and first excited vibrational state of CO molecule. The numbers in Table 1 given in a normal font correspond to the measurements in the gas-flow regime and the numbers in bold – to the sealed-off one. As can be seen, the values for both regimes are relatively close. The reproducibility of results was estimated as 10%.

The presence of nitrogen in the mixture results in increased populations of CO excited vibrational-states, and this effect grows with the considered nitrogen concentrations. Laser generation was observed in He-CO-N₂ mixtures (at the level 0.4 W) but it wasn't observed when nitrogen was absent in the mixture under any considered experimental conditions.

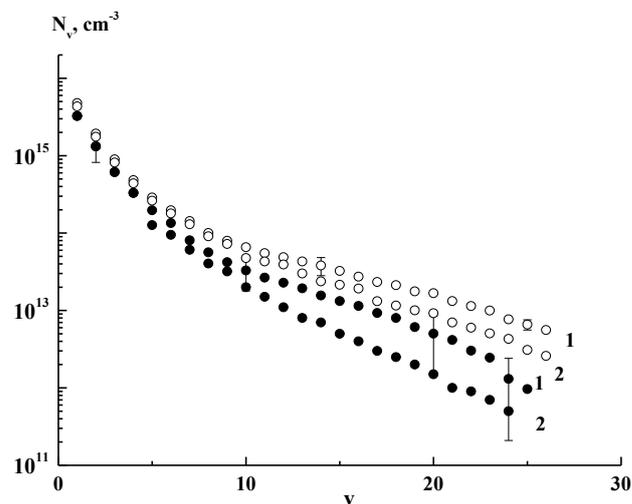


Fig. 1. Populations of CO vibrational states in He-CO and He-CO-N₂ mixtures without laser-radiation generation at various discharge currents; black full circles - He:CO=91:9 mixture, open circles - He:CO:N₂=91:4:5 mixture; curves 1: I = 25 mA, 2: I = 40 mA, pressure P = 12 Torr.

It was found that a decrease of CO vibrational states-populations in the mixtures containing nitrogen proceeds under much higher energy inputs (currents) than in the mixtures without nitrogen. Figure 1 presents CO vibrational energy distributions for mixtures He-CO and He-CO-N₂ for different current values. It can be seen that populations in the He-CO-N₂ mixture fall with significantly slower current values. It should be pointed out that the gas temperatures measured (near symmetry axis) in both mixtures He-CO and He-CO-N₂ were similar and varied in the range 360-480 K. These temperatures do not depend on gas flow.

One of the main causes of a smaller decrease of CO vibrational states-populations under increase of a current value in the mixtures with nitrogen is probably related to a lower CO-dissociation level and lower concentrations of plasma-chemical decomposition products. This should be proved in future investigations.

In conclusion, the performed measurements under conditions characteristic for a plasma discharge. In a CO laser showed that a nitrogen addition to the He-CO mixture leads to significant increases in CO vibrational temperature as well as populations of CO excited states. The reduction of CO highly-excited vibrational-states populations with a current is smaller in the case of mixtures with nitrogen additions. It was also found from the IR spectra that the CO dissociation decreases in the plasma with nitrogen.

More detailed investigations of plasma-chemistry dependence (especially CO concentration) on a nitrogen content should be performed in the future.

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