POPULATION INVERSION MECHANISM OF THE UV Cu⁺ LASER IN A PULSED LONGITUDINAL NANOSECOND Ne-CuBr DISCHARGE

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Abstract

Optimal discharge conditions for UV laser operation are found for two different active zone diameters of the laser tube - 9 and 5.2 mm. With diameter reduction a record average laser power of 550 mW is obtained at multiline output. The highest peak pulse and average output powers are measured on the 248.6-nm laser line for the UV Cu⁺ lasers - 2.2 W and 440 mW, respectively. The obtained specific average laser power is 30.5 mW.cm⁻³. A simplified kinetic model is made.

1. Introduction

The main interest in Cu⁺ lasers comes from opportunities for UV lasing. A stable operating Cu⁺ laser on the 248.6-nm line with a narrow linewidth and a high beam quality can be used in injection-locking systems with KrF excimer laser. At an average output power of about 500 mW, UV CuBr vapor laser can be applied in microelectronics, microbiology and genetic engineering as an alternative light source instead of the low-power KrF excimer laser. In [1] a laser oscillation on four Cu⁺ lines 248.6 nm, 252.9 nm, 260.0 nm, and 270.8 nm has been reported for the first time in a nanosecond pulsed longitudinal Ne-CuBr discharge with pulse repetition frequency 12 - 30 kHz. With an improved discharge tube design average output powers of 270 mW at multiline output and of 210 mW on the 248.6 nm line have been obtained in [2]. It was shown in [2] that the lower level population of the 248.6-nm laser line depends strongly on the Ne and Cu⁺ metastables concentration. In both investigations the copper atoms are produced by an electron impact dissociation of the CuBr molecules during the discharge period. In [2] it is also shown that the presence of Br atoms in the discharge leads to a reduction of the Ne metastables density via Penning ionization and, hence, to a Cu⁺ metastables density decrease and an increase of the output laser power. It is assumed that the reduction of the inside tube diameter could lead to a Ne and Cu⁺ metastables decay and hence to a considerable increase of the inversion population and the output parameters of the UV Cu⁺ lasers. That is why a comparatively investigation on two laser tubes with inside diameters of 9 mm and 5.2 mm is carried out.

2. Experimental apparatus

The construction of the basic discharge tube is shown in Fig. 1. The reduction of the inside tube diameter is obtained by using of different quartz inserts. The basic tube of 18-mm inside diameter is made of fused silica. The active length is 86 cm. The CuBr powder is placed in five quartz side- arm reservoirs. Quartz tube inserts of inside diameter of 9 mm (laser tube T1)
and 5.2 mm (laser tube T2) are sleeved in the basic tube. In each insert five hole are made above the reservoirs for CuBr vapor diffusion into the active zone.

![Diagram of discharge tube](image)

**Fig. 1. Construction of the discharge tube**

The ion CuBr vapor laser is excited by an electrical scheme known as the Interacting Circuits (IC) scheme [1,3], which can provide tube voltage and discharge current pulses up to 14 kV and 150 A with duration about 150 ns and pulse repetition frequency up to 30 kHz.

### 3. Experimental results

The utilization of a laser tube of the design, shown in Fig. 1, allows to carry out an investigation on laser tubes with different active zone diameter. The dependence of the average output power on the Ne buffer-gas pressure at active zone diameters of 9 and 5.2 mm is presented in Fig. 2. In spite of the active volume decrease a considerable increase of the average output power is observed. At active zone diameter of 5.2 mm (tube T2) record average output powers of 550 mW at multiline output and 440 mW on the 248.6-nm laser line are obtained. An increase of the optimal Ne pressure from 15 to 20 Torr with the reduction of the active zone diameter is observed.

The waveforms of the laser pulse at active zone diameters of 9 and 5.2 mm are shown in Fig. 3. A decrease of the laser pulse delay from 4.1 to 3.2 µs with respect to the current pulse end is observed with the diameter reduction. A record peak pulse power of 1.9 W is obtained at inside diameter of 5.2 mm (tube T2). The laser pulse duration at the base increases from 14.8 to 20.8 µs with the diameter decrease from 9 to 5.2 mm.
At optimal discharge conditions for inside diameter 5.2 mm the waveforms of the tube voltage and discharge current pulses are given in Fig. 4. The amplitudes of the tube voltage and discharge current are about 10.2 kV and 80 A, respectively. The duration of both pulses is about 120 ns at the base. The time-delay of the discharge current to the tube voltage start is about 25 ns.

The experimental results are summarized in Table I. It can be concluded that with the reduction of the active zone diameter from 9 mm to 5.2 mm the following features of the laser operation are observed: 1) The optimal Ne pressure increases, as the value of $p_{Ne,d}$ is not constant but slightly decreases; 2) The average, peak pulse and specific output powers increase; 3) The laser pulse delay to the current pulse end decreases from 4.1 to 3.2 $\mu$s; 4) The laser pulse duration increases.

For the laser tube T1 the waveforms of the laser pulses on the 248.6 nm line are observed at different reservoir temperatures (1-6) (the CuBr vapor pressure increases from curve 1 to 6). The results are shown in Fig. 5. The laser pulse delay after the end of the excitation electrical pulse decreases from 8 to 4.1 $\mu$s, with the vapor pressure increasing.

4. Kinetic study

Table I: d - active zone diameter; $p_{Ne}$ - optimal Ne buffer-gas pressure; $P_{out}$ - average output power at multiline output; $\tau_d$ - time-delay of laser oscillation with respect to the current pulse end; $\tau$ - laser pulse duration at the base; $P^{SP}_{out}$ - specific average output power; $P^{p}_{out}$ - peak pulse power on the 248.6-nm Cu$^+$ laser line; $f$ - pulse repetition frequency.

<table>
<thead>
<tr>
<th>d, mm</th>
<th>$p_{Ne}$, Torr</th>
<th>$p_{Ne,d}$, Torr.mm</th>
<th>$P_{out}$, mW multiline output</th>
<th>$\tau_d$, $\mu$s</th>
<th>$\tau$, $\mu$s</th>
<th>$P^{SP}_{out}$ mW/cm$^3$</th>
<th>$P^{p}_{out}$, W 248.6 nm</th>
<th>f, kHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.2</td>
<td>20</td>
<td>104</td>
<td>485 (550)</td>
<td>3.2</td>
<td>20.8</td>
<td>27 (30.5)</td>
<td>1.9</td>
<td>19.5-25</td>
</tr>
<tr>
<td>9</td>
<td>15</td>
<td>135</td>
<td>75</td>
<td>4.1</td>
<td>14.8</td>
<td>1.4</td>
<td>0.3</td>
<td>19.5-25</td>
</tr>
</tbody>
</table>

Fig. 4. Waveforms of tube voltage and discharge current pulses.

Fig. 5. Waveforms of laser pulses (1-6) on the 248.6-nm line at different reservoir temperatures.

In Fig. 6 energy levels of the copper, neon and bromine ions, and the neon metastables are shown. The upper laser levels 5s of the UV Cu$^+$ lines are populated via charge exchange collisions.
between the ground state neon ions and the ground state copper atoms.

In the simplified model described below only the 248.6-nm line is considered. The upper level 5s \(^3D_1\) of this line is depopulated via the laser operation transition and radiative transitions to the lower laser level and to five other levels (\(^{13}P^O_1, ^3D^O_{1,2}, ^1D^O_2\)). The 248.6-nm lower level 4p \(^3P^O_2\) is mainly depopulated by UV spontaneous emission to the \(Cu^+\) metastable levels 4s \(^3D_{1,2,3}\), and is populated by Penning impacts between neon metastables and ground state copper atoms. The high population of the \(Cu^+\) metastable levels leads to an increase in the lower laser level population by radiative trapping [4,2]. The lower laser level 4p \(^3P^O_2\) is additionally populated via radiative decay of 5s \(^3D_{2,3}\) (Fig. 6).

The laser oscillation is observed in the discharge afterglow and the laser pulse delays 3-5 \(\mu s\) after the excitation pulse end (Fig. 3 and 5). The laser pulse duration varies from 15 to 21 \(\mu s\) depending on the inside tube diameter and CuBr vapor pressure (Fig. 3 and 5). The electron temperature of the gas discharge plasma 3 \(\mu s\) - 20 \(\mu s\) after the excitation pulse end is about 0.35-0.5 eV [5,3]. At such electron temperature the excitation and deexcitation of the \(Cu^+\) and Ne\(^+\) levels via direct or stepwise electron collisions can be neglected, because of the small cross section for these interactions [5].

Under these assumptions the kinetic equations for the upper and the lower levels of the 248.6-nm laser line before the laser oscillation start (3-5 \(\mu s\) after the current pulse end) can be written in the following form:

\[
\frac{dN_3}{dt} = K_{CT} \cdot N_{Ne^+} \cdot N_0 - \frac{N_3}{\tau_{32}} - \frac{N_3}{\tau_{3}}
\]

\[
\frac{dN_2}{dt} = K_{p} \cdot N_{Ne^+} \cdot N_0 + \frac{N_3}{\tau_{32}} + \frac{N_3}{\tau_{32}'} + \frac{N_3}{\tau_{32}''} - \frac{N_2}{\tau_{21}}
\]

where \(N_3\) and \(N_2\) are the population densities of the upper 5s \(^3D_1\) and lower 4p \(^3P^O_2\) levels of the 248.6-nm laser line; \(N_{Ne^+}\) and \(N_{Ne^+}\) are the concentrations of the neon ions and metastables; \(N_0\) is the concentration of the ground state copper atoms; \(N_3^+\) and \(N_3^++\) are the population densities of the 5s \(^3D_{2,3}\) levels; \(K_{CT}\), \(K_{p}\) are the rate constants for population of the upper and lower laser levels via charge exchange and Penning collisions, respectively; \(1/\tau_{32} = A_{32}\), \(1/\tau_{32}' = A_{32}'\), \(1/\tau_{32}'' = A_{32}''\) are the probabilities for spontaneous emission from the upper laser and 5s \(^3D_{2,3}\) levels to the lower laser level; \(\tau_{21}\) is the effective lifetime for the lower laser level radiative decay to the three \(Cu^+\) metastable states 4s \(^3D_{1,2,3}\) considering the radiative trapping (taken from
\[ \frac{1}{\tau_3} = \sum_i A_{3i} \] is the probability for the upper laser level radiative decay where the subscript \(i\) refer to all radiative transitions depopulating this level with the exception of the 248.6-nm transition. The term \(N_3 / \tau_{32}''\) can be neglect in (2) because of the large lifetime \(\tau_{32}''\) is 769 ns [6]. For \(N_3\) the following balance equation can be written:

\[
\frac{dN_3}{dt} = K_{CT} N_3 N_0 - \frac{N_3}{\tau_3} - \frac{N_3'}{\tau_3'}
\]

where \(K_{CT}'\) is the rate constant for 5s \(3D_2\) level population via charge exchange impacts;

\[
\frac{1}{\tau_3} = \sum_i A_{3i}'
\]

is the radiative decay probability of this level, and the subscript \(i\) refer to all radiative transitions depopulating this level excepting the transition to the 248.6-nm lower level.

In a hollow cathode with continuous wave (CW) excitation a CW UV laser operation is observed [7]. That is why it can be supposed that at pulsed excitation the laser oscillation will start during the discharge current pulse or immediately after the current pulse end. Moreover in nanosecond pulsed longitudinal Ne-Cu and Ne-CuBr discharges the neon ions density decreases slightly for about 30 \(\mu\)s and is estimated about \(2 \times 10^{12}\) cm\(^{-3}\), while the concentration of the ground state copper atoms increases from \(1 \times 10^{14}\) cm\(^{-3}\) to \(4 \times 10^{15}\) cm\(^{-3}\) [5]. It shows that the laser pulse duration have to be considerably higher than the experimentally observed. In order to explain the observed delay and duration of the laser pulse, kinetic equations are considered below under quasistationary condition \((dN_i / dt = 0)\) and an determination of the reasons leading to an inversion population disturbance is carried out.

The condition for inversion population \(N_3 > \frac{g_1}{g_2} N_2\) can be written using (1), (2), (3) in the following form:

\[
\tau_3^C > \frac{g_3}{g_2} \left( \frac{K_{CT} N_0}{K_{CT} N_{Ne}} + \frac{\tau_3^C}{\tau_{32}} + \frac{K_{CT'} \tau_3'^C}{K_{CT} \tau_{32}} \right) \tau_{31}^{eff}
\]

where \(\tau_3^C = (1/\tau_{32} + 1/\tau_3)^{-1}\) and \(\tau_3'^C = (1/\tau_{32} + 1/\tau_3')^{-1}\).

A cross section of \(2.2 \times 10^{-15}\) cm\(^2\) for 248.6-nm upper level population with energy defect \(\Delta E\) of 0.183 eV via charge exchange reaction is calculated in the way given in [8]. Using the method described in [9], based on the dependence of the 248.6-nm spontaneous emission duration on the vapor pressure, the cross section of \(4 \times 10^{-16}\) cm\(^2\) for charge exchange collisions is experimentally determined. The corresponding rate constants for charge exchange reaction are \(3.3 \times 10^{10}\) cm\(^3\)s\(^{-1}\) and \(0.6 \times 10^{10}\) cm\(^3\)s\(^{-1}\). The rate constant for Penning collision population of the 248.6-nm lower level as well as the probabilities (the respective lifetimes) for the radiative transitions involved in the model, are taken from [2,6].

Using the estimated rate constants, as well as the data for neon ions and metastables densities [5] it is obtained that the condition (4) is disturbed at a lifetime \(\tau_{31}^{eff} > 8.5\) ns, which obtained as a result of the radiative trapping [2] at a Cu\(^+\) metastable density higher than \(5.8 \times 10^{12}\) cm\(^{-3}\) for laser tube T1 (9 mm) and \(11.1 \times 10^{12}\) cm\(^{-3}\) for tube T2 (5.2 mm).
5. Discussion and conclusions

The carried out experiments show that with the decrease of the active zone diameter of the laser tube the following dependences are observed:
1) The average, peak pulse and specific output powers increase considerably;
2) The laser pulse delay to the current pulse end decreases;
3) The laser pulse duration increases.

Since the current pulse amplitude remains approximately constant, the discharge current density increases with the reduction of the inside laser tube diameter. That leads to an increase of the Ne* concentration, while the Ne*m density is saturated and, hence, the first term in (4) decreases considerably, i.e. condition (4) is disturbed at higher values of $\tau_{21}^{\text{eff}}$, corresponding to a higher Cu* metastables density.

The effective probability for radiative decay of 248.6-nm lower level $A^{\text{eff}}$ increases with the tube diameter reduction. As in [2,5], it is calculated that the critical value $1.18 \times 10^8$ s$^{-1}$ of $A^{\text{eff}}$ (respectively $\tau_{21}^{\text{eff}} = 8.5$ ns) is reached at about two times higher concentration of Cu* metastables with the diameter decrease from 9 mm to 5.2 mm. Moreover the influence of the different particles diffusion increases, because of the higher gas temperature and closer tube walls.

On the basis of the simplified model results it can be concluded that the observed influence of the laser tube diameter decrease on the output power and laser pulse is due to the higher population of the upper laser level because of the higher Ne* density and mainly to the more effective depopulation of the lower laser level.

Acknowledgment

This work was supported by project NATO SfP 971989 and by Bulgarian Science Fund, Grant F-703.

References