Effect of HBr additives into the active media of copper-vapor and copper-halide lasers

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Investigations of the output parameters of CuBr(Cu)+HBr laser systems have shown that HBr additives to the CuBr laser active medium result in an increase of the laser output power and efficiency that compares, in this case, with that of a CuBr laser with H_2 additives. It has been found that the vapor of working substance are additionally produced in gas-discharge tubes (GDTs) less than 2 cm in diameter due to expulsion of copper atoms from the GDT walls by HBr. The results of simulating the kinetics of the copper vapor laser active medium with HBr additives show that the enhancement of the laser performance parameters can be explained by the processes of chemical transformation of copper from the solid phase into the gas one.

Introduction

The main attention of the investigations into the metal vapor lasers (MVL) is now being given to the lasers, whose active media are modified by various halogen compounds of the working metal with the following addition of hydrogen or active additives into the active medium.[1,2] This modification enhances energy and frequency characteristics, as well as the beam quality of such lasers. This fact determines an intense use of such lasers in modern systems for high-speed recording of optical information, microprocessing of materials, sensing of the atmosphere, laser separation of isotopes, etc.[2–4]

Since the copper vapor laser is the most efficient source of visible radiation among all metal vapor lasers, the most widely used types of active media with the modified kinetics are:

1. Copper vapor lasers with hydrogen additives.
2. Copper bromide (chloride) lasers with hydrogen additives.
3. Copper vapor lasers with hydrogen halide (HBr, HCl) additives.
4. Hybrid copper vapor lasers (HyBrID, HyClID).
5. Copper vapor lasers with other additives (Cs, Zn, Sc, Ni, Ag, etc.).

The enhanced energy and increased optimal frequencies for a copper vapor laser achieved due to molecular hydrogen additive were reported for the first time as early as in 1980.[5] Quite recently Kazaryan et al.,[6] have reported on the beginning of industrial production of Kristall LT-30, -40, -50, and -70Cu sealed-off active elements with hydrogen additives at the Istok Industrial Association (Russia). The use of hydrogen additives has increased the laser output power by 1.5 times. Hydrogen additives in the active medium of copper halide laser not only doubled the output power, but also significantly increased the laser efficiency.[7] The maximum practical efficiency for this class of lasers is 3% (Ref. 8).

The idea of using hydrogen additives in the active medium of metal salt lasers has received its further development in hybrid lasers, in which hydrogen halide molecules flowing through the MVL active medium interact with metal copper and form CuBr molecules with their following dissociation.[9] This allows operation, as in the CuBr laser, at the wall temperatures being threefold lower than those in a copper vapor laser without additives. It was just this MVL modification that has demonstrated the maximum output power higher than 200 W from one active element, the efficiency higher than 3% (Ref. 10), and the highest mean specific output power of 2 W/cm^3 (Ref. 11).

Since at that time it was believed that the enhancement is caused by the presence of halide molecules, the characteristics of unmodified MVL were compared with those of MVL with H_2, Br_2, HBr, and HBr + H_2 additives.[12] It has been shown that HBr additives almost doubled the output power. This has led to the development of a new type of metal vapor lasers with improved kinetics. The use of HCl + H_2 simultaneously almost tripled the output power as compared with that of MVL of the same size, but without additives.[13]

Unfortunately, many questions concerning the causes for enhancement of the energy and frequency characteristics of these types of MVL are still to be addressed. But the main hypothesis is essentially based on the presence of hydrogen halide molecules in the active medium. These molecules have a high dissociative attachment cross section, which favors the decrease in the pre-pulse electron concentration.

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The use of other additives as active admixtures (Ag, Cs, Sc, Ni, Zn, and others)[14–16] is still of pure scientific interest, since the H_2 additives have a more pronounced effect on the laser parameters.

Despite the highest output characteristics were obtained with hybrid lasers and lasers with improved kinetics, the application of these lasers is limited because they are flow-through systems. The Ne + H_2 mixture can be efficiently used in sealed-off Cu and CuBr lasers only for some period, since then hydrogen enters in GDT into chemical reactions and its concentration decreases.

Recently it has become possible to employ selective reversible hydrogen generators of the SRNV type produced by the Impul'snye Tekhnologii Company (Ryazan, Russia) in sealed-off laser tubes.[17]

The aim of this work was to study the effect of hydrogen halide (HBr) additives on the energy characteristics of Cu and CuBr lasers, as well as to analyze the results of theoretical studies of the HBr effect on the copper vapor laser characteristics and the experimental results obtained, for the first time, for the sealed-off CuBr + HBr system with a specialized HBr generator.

Experiment

The energy characteristics of CuBr lasers with HBr additives have been studied using gas-discharge tubes (GDTs) with the adjustable length and diameter of the active zone. Besides, two GDT designs were employed. These designs differed in the method of feeding the working vapor into the active zone: independent heating of CuBr containers[18] and self-heating mode.[19] The parameters of these GDTs are tabulated below.

GDTs had a built-in HBr generator, which not only generated HBr into the laser active medium, but also evacuated it back into the generator. The HBr concentration was regulated in a wide range by the heater temperature, and the optimal additive was determined as that which peaked the output power. At the constant heater temperature, the HBr concentration was established at a certain level.

The GDT was pumped by the traditional scheme of the direct discharge of a KVI-3 working capacitor into the GDT through a switch.[18] The GDTs 1–4 with the switched power up to 3 kW were pumped through a water-cooled TGI1-1000/25 thyratron. In the case of switching the power higher than 3 kW for GDT 5, the pumping was performed by the scheme of alternate operation of two water-cooled TGI1-1000/25 thyratrons. To increase the voltage applied to the GDTs with large interelectrode gaps (GDTs 4, 5), a pulsed cable transformer was used as in Ref. 20. The transformer was made as a PVTFE cable with fluoroplastic insulation winded around ten 100×60×15-mm^3 ferrite rings; the number of windings was three.

The current and voltage pulses were recorded with a Rogowski coil and a low-inductance divider in TVO-type resistors, while the emission was recorded with an FK-22 coaxial photocell. Pulses were displayed on the screen of a Tektronix TDS-3032 oscilloscope. The output power was measured with an IMO-2 power meter, and the GDT wall temperature was measured using a chromel–alumel thermocouple.

The first measurements of the output energy characteristics of GDTs under study were conducted as follows. The GDTs without HBr additives were brought to steady-state operating conditions and the output power was measured (the values are given in the Table). Then the heater of the HBr generator was turned on, and HBr was fed into GDT. The optimal HBr concentration at the initial stage was judged visually by the diameter of the discharge filament.

Results and discussion

The experiments showed that the process of establishing the steady-state operating conditions was significantly different in lasers with GDTs having small (< 2 cm) and large (> 2 cm) diameter. The gradual feed of HBr into the discharge in GDTs 1 and 2 (the output power in pure Ne under the steady-state conditions was 0.15 and 0.5 W, respectively) led first to an increase of the output parameters and then to their decrease because of the excess of the working substance vapor. This could be judged visually from ejection of the working substance into the near-electrode and near-end zones.

Dimensions and power parameters of GDTs under study

GDT #	Diameter, cm	Length, cm	Pump power, W	Output power		
				without H_2 and HBr, W	with H_2, W	with HBr, W
1	1.1	16	400–500	0.15	0.8	0.8
2	1.6	36	800	0.5	2.5	2.5
3	2.6	76	1400–1500	5	10	10.5
4	3.6	120	2500–3000	13	25 (36)	26 (38)
5	5.3	145	4000–5000	22	41 (55)	~40 (55)

Note: The values in parentheses are the output power for GDTs 4 and 5 obtained with the aid of a pulsed cable transformer. The buffer gas (Ne) pressure was 15–30 Torr, the pulse repetition frequency was 17–22 kHz; GDTs 1, 2 are with independent heating of containers, GDTs 3–5 employ a self-heating design.

The further feed of HBr led to the discharge quenching, and therefore a need arose in decreasing the temperature of the heaters of CuBr containers. Only after that the output power achieved maximum: 0.8 and 2.5 W. Thus, for example, to achieve maximum power with GDT 2 (2.5 W), the output power was set at 0.3 W through adjusting the temperature of the heaters of CuBr containers and then HBr was fed.

The tests of GDTs 3–5 have shown that the behavior of the generation has no tendencies inherent in the previous GDTs. The feed of HBr resulted in a gradual power increase from 5, 13, 22 to 12, 26, and 40 W, respectively, without changing the thermal insulation layer on the CuBr containers. The use of the pulsed cable transformer has allowed an extra increase of the power in GDT 4, 5 to 38 and 55 W to be obtained.

To analyze the mechanisms of the influence of HBr additives on the output characteristics of a copper vapor laser, we have developed a detailed nonstationary kinetic model of the Cu–Ne–H_2–HBr (Cu–Ne–H_2–Br_2) active medium. The model allows us to calculate and study the populations of the energy levels of copper, neon, and bromide atoms, the ion densities of these elements, the electron temperatures, and the laser radiation intensity at two wavelengths (λ = 510.6 and 578.2 nm). For detailed description of the model, see Ref. 21. The model accounts for the ground and two first vibrational states of the HBr molecule, as well as the ground state of the bromide molecule. For the atomic bromide, the model accounts for the ground and the first excited state, as well as the ground states of the negative and positive ions. The ground state of the CuBr molecule was taken into account as well. The model included a total of about 240 kinetic reactions.

It is obvious that we cannot directly draw the analogy between the results of simulation of a copper vapor laser with HBr additives and the experimental data obtained for the copper bromide laser with the same additives. At the same time, we know that there is a lot in common between hybrid lasers, lasers with improved kinetics, and copper bromide lasers. Thus, a copper vapor laser with HBr additives differs from the hybrid one only by the absence of working mixture flow, while the working temperatures in the laser with improved kinetics practically correspond to that in ordinary high-temperature copper vapor laser.

The study of the kinetics of the active medium of a copper vapor laser with hydrogen bromide additives has shown[21] that the use of even the overstated constant of recovery of the HBr molecule does not provide for high equilibrium concentration of hydrogen bromide. In spite of this, injection of hydrogen bromide significantly modifies the kinetics of the laser active medium. The following two processes exert a principal impact on the laser operation: variation of the concentration of copper atoms and variation of the electron concentration.

Injection of hydrogen bromide additives leads to a significant decrease in the pre-pulse electron concentration during the time gap between pulses in the processes:

$$\mathrm{HBr}\ (\nu = 0,\ 1,\ 2) + e \to \mathrm{H} + \mathrm{Br}^- . \qquad (1)$$

The increase in the concentration of copper atoms in the active medium upon injection of hydrogen bromide additives occurs due to two parallel processes:

1. Increase in the temperature of the gas discharge tube due to a decrease in the electron concentration upon injection of hydrogen bromide into the active element of the laser;
2. Appearance of additional copper atoms due to conversion of copper from the solid phase into the gas one:

$$\left.\begin{aligned} \mathrm{Cu\ (solid) + Br\ (gas) \to CuBr\ (gas)},\\ \mathrm{Cu\ (solid) + HBr\ (gas) \to CuBr + H\,(gas)}, \end{aligned}\right\} \qquad (2a)$$

$$\left.\begin{aligned} &\mathrm{CuBr\,(gas)} + e\,\mathrm{(Cu, H, CuBr, H_2)} \to\\ &\to + \mathrm{Br\,(gas)} + e\,\mathrm{(Cu, H, CuBr, H_2)},\\ &\mathrm{CuBr\,(gas)} \xrightarrow{\text{thermal}} \mathrm{Cu\,(gas) + Br}. \end{aligned}\right\} \qquad (2b)$$

Unfortunately, no data on the mechanisms of the processes of interaction of metal copper with hydrogen halide molecules, their cross sections, and rates for typical experimental conditions are available in the literature. Nevertheless, we can use our experimental data on the increase in the concentration of copper atoms upon injection of hydrogen bromide additive into the active medium of the copper bromide laser. In our opinion, the excess concentration of atoms of the working substance in the discharge zone of GDTs 1 and 2 is connected just with the processes of (2a) type: apart from the fact that CuBr pairs come to the discharge from containers, Br atoms and HBr molecules additionally expulse copper atoms attached to GDT walls. However, it should be noted that in large-diameter GDTs 3–5 this effect was not observed. We believe that the processes (2a) are less efficient in large-diameter GDTs. Thus, the energy injected into a unit volume in the large-diameter tubes is much lower, and, as a consequence, dissociation of HBr molecules and the concentration of active bromide are lower. So the main process of additional production of copper bromide [Cu (solid) + Br (gas) → CuBr (gas)] becomes negligible. Besides, in the large-diameter GDTs the length of diffusion of HBr molecules to the GDT wall, where the reactions of type (2) occur, increases and significant radial inhomogeneity of the gas temperature is observed.[22] Under these conditions, the main role in improvement of the lasing characteristics is likely played by the process of dissociative electron attachment to the HBr molecule.

In addition to these processes, the injection of hydrogen bromide into the active medium[21] lowers the degree of population depletion of the ground state of the copper atom and changes the electric characteristics of the discharge.

The injection of an HBr additive decreases both the pre-pulse electron concentration and the maximum electron concentration during the pump pulse (see Fig. 1). The HBr additive causes some increase in the maximum electron temperature and slows down its drop during the first stage of the afterglow (first 20 μs), see Fig. 2. This is connected, as in the case of hydrogen chloride, with the weaker (as compared to H_2 additive) effect of elastic Coulomb collisions in the afterglow because of the decrease in the concentration of positive ions in plasma due to the ion–ion recombination reactions involving the negative bromide ion. Nevertheless, the injection of hydrogen bromide into the active medium causes no significant decrease in the pre-pulse electron temperature.

As in the case of hydrogen chloride, the injection of hydrogen bromide is accompanied by the increase in the degree of dissociation of molecular hydrogen, and this leads to a decrease in the concentration of vibrationally excited hydrogen molecules and increases the conductivity of plasma of the active discharge. The changes in the time dependence (mostly in the period of late afterglow) of the electron temperature and the degree of population relaxation of the metastable levels of the copper atom are likely connected with just the above factors of the impact of the hydrogen bromide additives on the molecular hydrogen.

At the first stage of the afterglow period (from 1 to 20 μs) the concentration of copper atoms in the metastable states relaxes more and more slowly. This is connected with the decreasing number of the quenching electron collisions with copper atoms in the metastable states because of the decreasing electron concentration. At the same time, the concentration of copper atoms in metastable states decreases insignificantly.

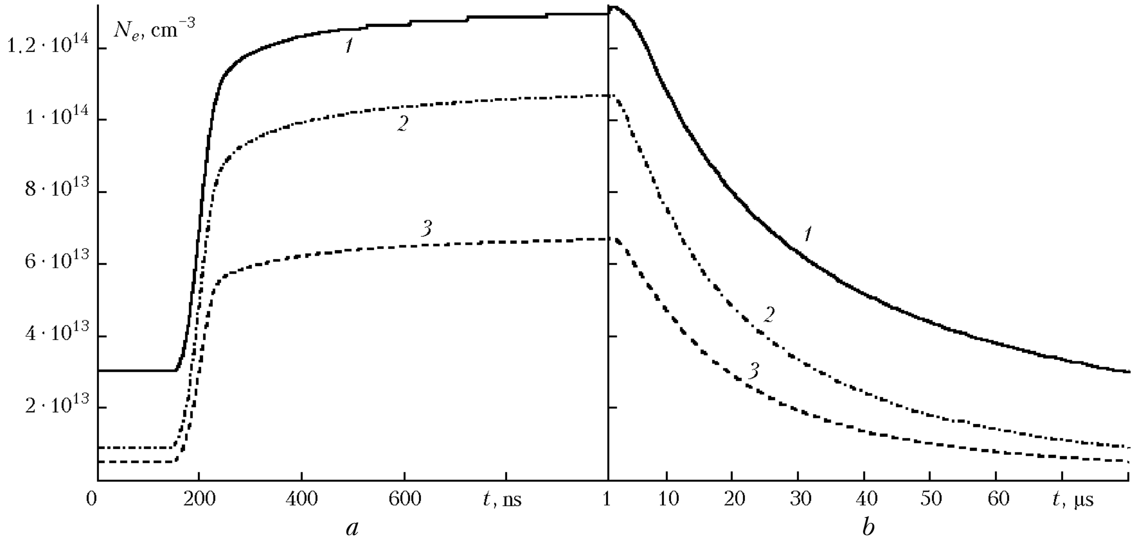


Fig. 1. Time dependence of the electron concentration during the pump pulse (*a*) and the gap between pulses (*b*): $N_{\rm HBr}$ = 0%, $N_{\rm Cu} = 0.8 \cdot 10^{15}$ cm^{-3}, $I_{\max}$ = 840 A (curve *1*); $N_{\rm HBr} = 1.5 \cdot 10^{14}$ cm^{-3}, $N_{\rm Cu} = 0.8 \cdot 10^{15}$ cm^{-3}, $I_{\max}$ = 840 A (curve *2*); $N_{\rm HBr} = 1.5 \cdot 10^{14}$ cm^{-3}, $N_{\rm Cu} = 1.1 \cdot 10^{15}$ cm^{-3}, $I_{\max}$ = 610 A (curve *3*).

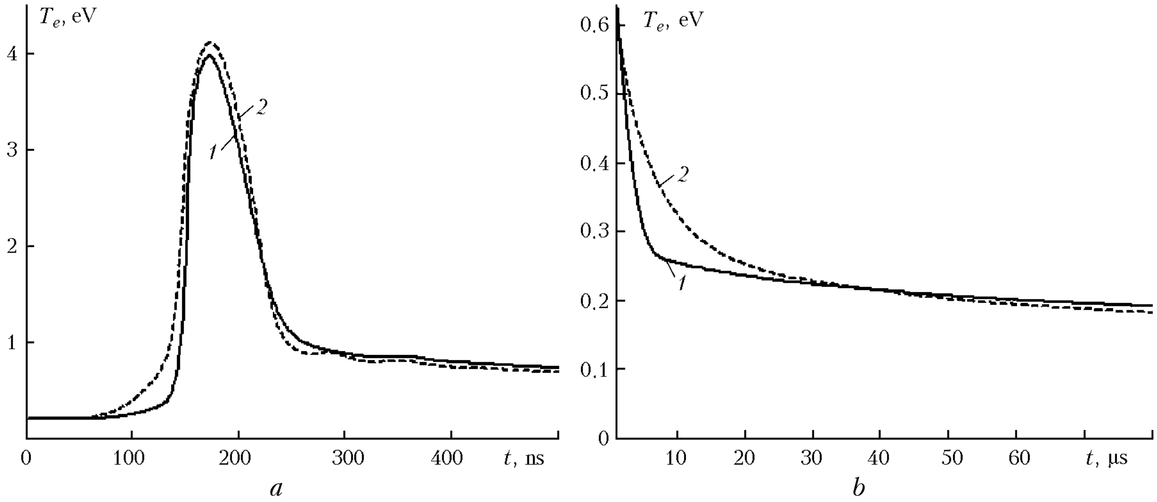
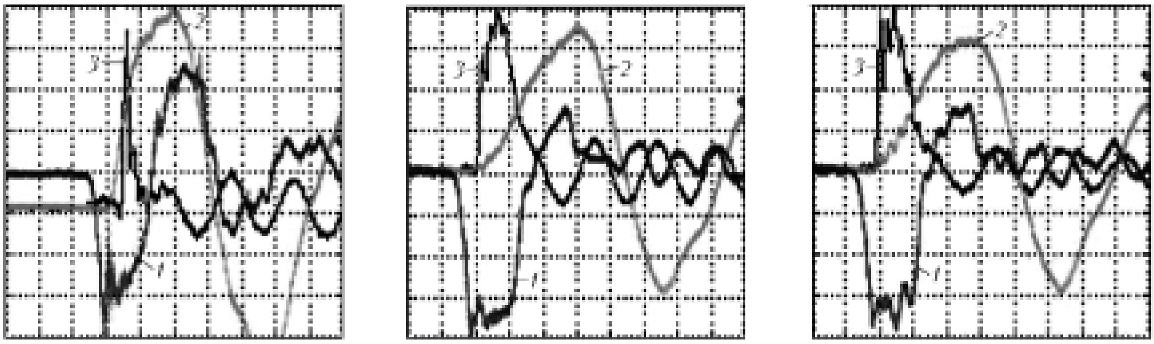


Fig. 2. Time dependence of the electron temperature during the pump pulse (*a*) and in between the pulses (*b*): $N_{\rm HBr}$ = 0%, $N_{\rm Cu} = 0.8 \cdot 10^{15}$ cm^{-3}, $I_{\max}$ = 840 A (curve *1*); $N_{\rm HBr} = 1.5 \cdot 10^{14}$ cm^{-3}, $N_{\rm Cu} = 1.1 \cdot 10^{15}$ cm^{-3}, $I_{\max}$ = 610 A (curve *2*).



a *b* *c*

Fig. 3. Oscillograms of the voltage (*1*), current (*2*), and lasing (*3*) pulses for GDT 3: pure Ne (*a*); with H_2 additive (*b*); with HBr additive (*c*) with the working capacitor of 825 pF, pump pulse repetition frequency of 17.2 kHz, pressure of the buffer gas Ne or Ne+HBr (Ne+H_2) of 20 Torr; power supply voltage and current: 6.2 kV and 0.22 A (*a*); 6.2 kV and 0.21 A (*b*, *c*); laser output power: 5 (*a*), 10 (*b*), and 10.5 W (*c*).

The obtained experimental and theoretical results suggest that the HBr additives to the active medium of Cu and CuBr lasers efficiently enhance the output characteristics to the level comparable with the parameters of Cu and CuBr lasers with H_2 additives. Earlier we have measured the output characteristics of these GDTs with H_2 additives in the sealed-off and unsealed versions. In the case of unsealed GDT, H_2 was pumped into a gas line from a gas cylinder. In the sealed-off version, GDT 4 with a SRNV1 hydrogen generator welded to it was operated. The comparison of the output power and efficiency of these lasers demonstrates their virtual identity. Figure 3 depicts the current, voltage, and lasing pulses for GDT 3 in pure Ne and with H_2 and HBr additives under the same excitation conditions and the same temperature of the GDT walls.

The comparison of operation of the studied GDTs with H_2 and HBr generators suggests that the developed HBr generator has no disadvantage inherent in the hydrogen generator when used in the CuBr laser. This disadvantage is connected with the fact that H_2 does not go back into the generator in the case of its excessive concentration in the discharge. Figure 4 shows the evolution of current (*1′–3′*) and voltage (*1–3*) pulses measured within a certain interval as HBr escaped from the discharge.

It can be seen from Fig. 4 that the pulses restore their initial shape *1′*, *1*, corresponding to GDT operation in pure Ne without additives. The speed of the HBr escape from the discharge back into the generator depends on the HBr concentration in the generator.

It should be noted that the developed HBr generator can be then used to degas GDT from atmospheric gases and products that negatively affect the output characteristics of a laser. For example, free bromine is accumulated in CuBr lasers with GDT operation. The experimental results[12,23] along with numerically calculated ones[21] evidence of the negative effect of bromine additives on the characteristics of a copper vapor laser. Thus, according to the calculations, as atomic bromine in the concentration of $1.5 \cdot 10^{14}$ cm^{-3} is injected, the mean output power decreases approximately by 10%.

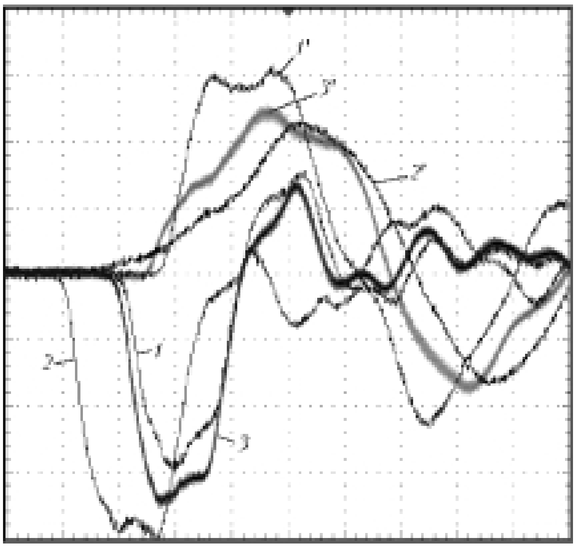


Fig. 4. Evolution of current (*1′–3′*) and voltage (*1–3*) pulses in the process of HBr escape from the discharge in the GDT 3 into the HBr generator; pulses correspond to laser operation in pure Ne without additives (*1′*, *1*), with the added HBr concentration higher than the optimal one (*2′*, *2*), 20 min after the HBr generator was turned off (*3′*, *3*).

Note that, according to the results of Ref. 21, virtually all injected bromine is in the atomic state (according to the calculations, as molecular bromine in the concentration of $1.5 \cdot 10^{14}$ cm^{-3} is injected, the pre-pulse concentration of molecular bromine is only $4.57 \cdot 10^{11}$ cm^{-3}). The proper choice of the operating conditions of the HBr generator will likely help in decreasing the bromine concentration in the discharge.

Service life of the active element

The first results on the use of CuBr–HBr lasers show that these lasers have the features characteristic of HyBrID lasers, in particular, the problem connected with the appearance of dendrites, which constrain the aperture of the active zone, in the channel of small-diameter GDTs after several tens of hours of operation.[9] To remove this phenomenon, Jones and Little[9] proposed to control the hydrogen concentration and the tube temperature by selecting accurately the thermal insulation in order to avoid local overheating. As to our experiment, we conducted the maintenance work. The discharge was initiated in GDT in pure Ne without vapor of the working substance, and the consumed power was a bit higher than the power needed for steady-state operation. After heating the GDT under these conditions for some time, dendrites were removed into cold zones (traps).

Conclusion

We have studied the output parameters of CuBr(Cu) + HBr laser systems. Experiments with five GDTs of different size have shown that injection of the HBr additive into the active medium of the CuBr laser enhanced the output power and efficiency (up to 2 times) similarly to the case of H_2 additives. The analysis of electrical characteristics (amplitude and duration of pump pulses) of the CuBr + HBr and CuBr + H_2 lasers under the same pumping conditions has also demonstrated their identity.

In addition, it was found that the vapor of the working substance is generated in the active zone of the CuBr + HBr laser with GDT of 2-cm diameter not only due to heating of the CuBr powder, but also due to expulsion of copper atoms from GDT walls by hydrogen bromide (bromine) in the reactions of type (2), which is similar to the operating principle of the HyBrID laser. The similarity with the hybrid laser is in deposition of dendrites in the GDT channel as well. In GDTs with the diameter larger than 2 cm, no increase in the concentration of the working substance vapor due to this effect was observed most probably because the process (2) proves to be inefficient owing to the long length of diffusion of HBr molecules to the wall. So the enhancement is provided for by the process of dissociative attachment of electrons to HBr molecules.

The paper also presents the results of simulation of the kinetics of the active medium of a copper vapor laser with HBr and Br_2 additives. The calculated results confirm the experimentally observed decrease in the mean output power at accumulation of the bromine additive in the active medium.

To explain the enhancement of the output characteristics of copper vapor and copper bromide lasers at injection of hydrogen bromide, it is necessary to consider other mechanisms, for example, processes (2) of chemical transformation of copper from the solid phase into the gas one. This confirms the conclusions drawn in Ref. 21.

The tests of the HBr generator have shown that the concentration of the HBr additive in the laser can be controlled within a wide range. When used in CuBr lasers, this generator differs from the known H_2 generators by the property of reversibility. The excessive HBr concentration leading to the decrease in the output power can be readily removed by turning the generator heating off.

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