

ACTIVE MEDIA

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Amplification and gas-dynamic parameters of the active oxygen–iodine medium produced by an ejector nozzle unit

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***Abstract*. The gain, the temperature, and the absolute velocity of the supersonic active oxygen – iodine medium produced by an ejector nozzle unit were determined by the technique of high-resolution diode laser spectroscopy. The gain in the active medium is formed at less than 44 mm from the nozzle unit for an absolute flow velocity $v \approx 600$ m s^{-1}. Upon dilution of oxygen by primary nitrogen in the ratio of 1 : 6.9, the gain of the active medium amounts to 7×10^{-3} cm^{-1}, the temperature of the active medium to 200 K, the absolute flow velocity to 580 m s^{-1}, and the pressure to 58 Torr. As the dilution is increased to 1 : 13.5, the gain reduces to 4.5×10^{-3} cm^{-1}, the temperature lowers to 180 K, the velocity of the active medium increases to 615 m s^{-1}, and the pressure increases to 88 Torr. The increase in the initial content of water vapour in the oxygen flow results in an increase in the temperature and a decrease in the gain of the active medium.**

Keywords: *oxygen – iodine laser, iodine atom, diode spectroscopy.*

1. Introduction

A high ($\sim 20\,\%$) efficiency of an oxygen – iodine laser with a high total pressure of the active medium in the resonator has been demonstrated in Ref. [1]. The active medium is produced by an ejector nozzle unit, at the outlet of which there occurs a supersonic mixing of three flows that serve different functions. Singlet oxygen as the energy carrier arrives at the ejector array from the jet generator of singlet oxygen (JGSO) and flows into the mixing chamber through a series of slots with a Mach number $M \sim 1$. High-pressure primary nitrogen initially at room temperature flows into the mixing chamber through a series of cylindrical nozzles and is accelerated to $M > 2$ at a short distance. A low-pressure flow of the mixture of nitrogen with molecular iodine at a temperature of ~ 340 K is injected between the two flows. It was shown in Ref. [1] that a nearly full mixing of the three flows occurs over a length of several centimetres. The nozzle array elaborated permits obtaining an oxygen – iodine active medium with a pressure of over 100 Torr recovered in the supersonic diffuser.

The technique of high-resolution diode laser spectroscopy is employed with advantage to determine the gain, the temperature of the active medium of an oxygen – iodine laser, and the collisional broadening coefficients for the laser transition [2 – 4]. Here, this technique was used to determine the gain, the temperature, and the absolute velocity of the active medium produced by the ejector nozzle unit.

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2. Experiment and diagnostics

Fig. 1 shows a schematic of the setup for measuring parameters of the active medium produced by the ejector nozzle array of a supersonic oxygen – iodine laser. (The production of the active oxygen – iodine medium by the ejector nozzle unit and its design are described in detail elsewhere [1].) The initial height of the mixing chamber is 16 mm and the width is 50 mm. At a distance of 11 mm from the nozzle unit, the large walls of the mixing chamber are inclined by 2° relative to the chamber axis, so that the height of the mixing chamber at a distance of 64 mm from the nozzle unit is ~ 20 mm. The side walls of the mixing chamber were optical wedges with an apex angle of 3° through which the beam of a diagnostic diode laser was injected. The evacuation of the active medium was accomplished with a mechanical AVZ-125 pump. A vessel with a volume of 4 m^3 pre-pumped to a pressure of less than 1 Torr was opened to afford a short-term increase in the volume flow rate of the gas medium. During the admissions of gases through the nozzle array, measurements were made of a pressure p_1 in the JGSO, a pressure p_2 in front of the oxygen nozzles, a near-wall pressure p_3 in the mixing chamber, and a p_4 pressure in the Pitot tube. The opening for measuring p_3 is the wall of the mixing chamber, at a distance of 64 mm from the nozzle array. The mouth of the Pitot tube was located at the centre of the lateral section of the mixing chamber, at 2.25 cm downstream from the opening for a p_3-pressure gauge.

To investigate the amplification spectrum of the active oxygen – iodine medium, we used a measuring complex involving a Physical Science Inc. high-resolution diode laser spectroscope tuned to the resonance frequency of the $^2P_{1/2}$ $(F = 3) \rightarrow {}^2P_{3/2}(F = 4)$ transition. The spectrum of the diode laser emission line was close to a Lorentzian with the linewidth $\Delta\nu = 8$ MHz. The spectrum was scanned within ± 1500 MHz relative to the line centre of the $^2P_{1/2}(F = 3) \rightarrow {}^2P_{3/2}(F = 4)$ transition.

The small-signal gain on the strongest $^2P_{1/2}(F = 3) \rightarrow {}^2P_{3/2}(F = 4)$ transition of atomic iodine

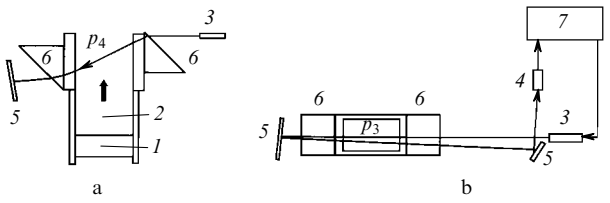

Figure 1. Schematic of the experimental setup: side view (a) and flow-on view (b): (*1*) nozzle unit; (*2*) mixing chamber; (*3*) probe laser; (*4*) photodetector; (*5*) mirrors; (*6*) prisms.

$$g(X) = \frac{7}{12}\Delta N \frac{A\lambda^2}{8\pi}\Phi(X), \tag{1}$$

where ΔN is the total inverse population density on the $^2P_{1/2} \rightarrow {}^2P_{3/2}$ transition; $A = 5.1\ \mathrm{s}^{-1}$ is the $^2P_{1/2}(F=3) \rightarrow {}^2P_{3/2}(F=4)$ transition probability; $\lambda = 1.315\times\ 10^{-4}$ cm is the radiation wavelength; $\Phi(X)$ is the form factor; $X = (\nu - \nu_0)$ is the radiation frequency shift relative to the $^2P_{1/2}\ (F=3) \rightarrow {}^2P_{3/2}(F=4)$ line centre. The inverse population density ΔN is determined by the density of atomic iodine N_1, the relative content of singlet oxygen $Y = [\mathrm{O}_2(^1\Delta)]/[\mathrm{O}_2]$, and the temperature T of the active medium:

$$\begin{aligned}\Delta N &= \left[\mathrm{I}\left(^2P_{1/2}\right)\right] - 0.5\left[\mathrm{I}\left(^2P_{3/2}\right)\right] \\ &= \frac{(Y - Y_{\mathrm{th}})(K_{\mathrm{eq}} + 0.5)}{(K_{\mathrm{eq}} - 1)Y + 1} N_{\mathrm{I}}\end{aligned} \tag{2}$$

where $\mathrm{K_{eq}} = 0.75\exp(401\mathrm{K}/T)$ is the $\mathrm{O}_2(^1\Delta) + \mathrm{I}(^2P_{3/2}) \leftrightarrow \mathrm{O}_2(^3\Sigma) + \mathrm{I}(^2P_{1/2})$, exchange equilibrium constant and $Y_{\mathrm{th}} = (2K_{\mathrm{eq}} + 1)^{-1}$ is the threshold fraction of $\mathrm{O}_2(^1\Delta)$ whereby the inversion vanishes.

Under the action of only the collisional and Doppler mechanisms of spectral line broadening, the total form factor $\Phi(X)$ is the Voigt function:

$$\Phi(X) = \left(\frac{\ln 2}{\pi}\right)^{1/2} \frac{\Delta\nu_{\mathrm{L}}}{\pi\Delta\nu_{\mathrm{D}}} \int_{-\infty}^{\infty} \frac{\exp\left(-Z^2 4\ln 2/\Delta\nu_{\mathrm{D}}^2\right)}{(X-Z)^2 + (\Delta\nu_{\mathrm{L}}/2)^2}\,\mathrm{d}Z. \tag{3}$$

The collisional width $\Delta\nu_{\mathrm{L}}$ is proportional to the static pressure in the flow: $\Delta\nu_{\mathrm{L}} = \alpha(T)p$, where $\alpha(T)$ is a temperature-dependent coefficient. The total Doppler width (in megahertz) is

$$\Delta\nu_{\mathrm{D}} = 14.49\sqrt{T}, \tag{4}$$

where T is the absolute temperature in Kelvins. For a probe beam directed at an angle φ with the normal to the flow velocity, the amplification spectrum shifts by $\delta\nu = (\sin\varphi)\upsilon/\lambda$ owing to the Doppler effect, where υ is the absolute velocity of the active medium.

We used two different optical schemes for scanning the amplification spectrum. In the first case, the diode laser beam was deflected with a prism through an angle $\varphi = 27.5^\circ$ from the normal to the direction of the flow velocity and entered the mixing chamber at a distance of 81 mm from the nozzle array to exit at 55 mm from the array. After reflection from the mirror, the probe laser beam retuned to the active medium at the same angle with the normal to the direction of the flow velocity, and its intensity was recorded by a photodetector.

After a double passage through the active medium, the net amplification spectrum for a probe laser beam is proportional to

$$\exp\left\{\frac{L_{\mathrm{a}}[g_1(X-\delta\nu) + g_2(X+\delta\nu)]}{\cos\varphi}\right\} - 1, \tag{5}$$

where $L_{\mathrm{a}} = 5$ cm is the length of the active medium; $g_1(X-\delta\nu)$ and $g_2(X+\delta\nu)$ are the gains for the forward and backward passages of the probe laser beam through the active medium.

In the second case, the optical prisms deflecting the probe laser beam were absent, and the diode laser beam was normally incident ($\varphi = 0$) with respect to the direction of the flow velocity. In this scheme, the net amplification for the probe laser beam is approximately equal to the net small-signal amplification upon double passage of the active medium $\exp\{L_{\mathrm{a}}[g_1(X) + g_2(X)]\} - 1$. The uncertainty in tuning the angle of incidence of the probe beam is $\sim 1^\circ$ for both optical systems.

The amplification spectrum was scanned for 5 s; during this period, all pressures and the amplification spectrum were stationary. The complex hardware and software allowed us to record the amplification spectrum in real time and calculate the functions $g_1(X-\delta\nu) + g_2(X+\delta\nu)$ or $g_1(X) + g_2(X)$, depending on the measuring optical configuration. Assuming the g_1 and g_2 functions to be the Voigt functions, it is possible to extract the Gaussian and Lorentzian components from them. In the case of oblique incidence of the probe laser beam, $g_1(X-\delta\nu)$ and $g_2(X-\Delta\nu)$ are determined separately. In the case of normal incidence, the average gain $[g_1(X) + g_2(X)]/2$ is found. The amplification spectrum of the active medium obtained when the probe laser beam was incident at an angle $\varphi = 27.5^\circ$ to the normal to the flow is exemplified in Fig. 2. Subsequent mathematical processing of the spectra permitted determining the gain coefficient $g(0) = g_1(0) + g_2(0)$ of the active medium at the centre of the $^2P_{1/2}(F=3) \rightarrow {}^2P_{3/2}(F=4)$ line, as well as determining $\delta\nu, \Delta\nu_{\mathrm{L}}$ and $\Delta\nu_{\mathrm{D}}$. Note that the total width of the Lorentzian form factor is $\Delta\nu_{\mathrm{L}} + \Delta\nu$.

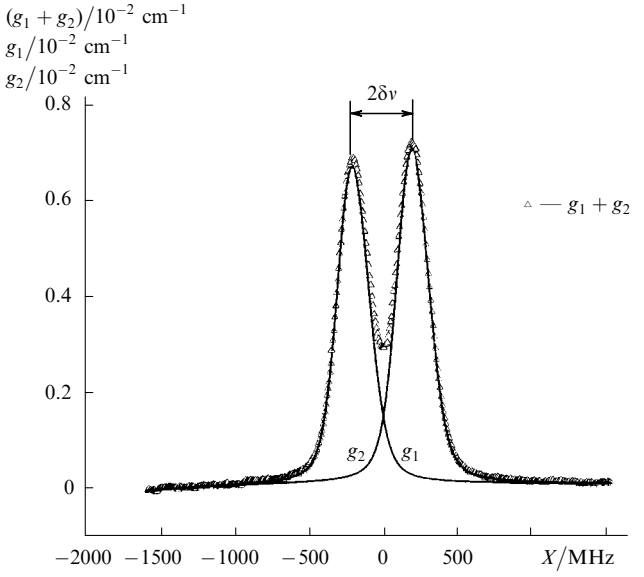

Figure 2. Experimental (points) and calculated (curves) amplification spectra of the active medium for an oblique incidence of the probe laser beam.

Processing the amplification spectrum yields the following parameters of the active medium: the total inverse population density on the $^2P_{1/2} \rightarrow {}^2P_{3/2}$ transition of atomic iodine

$$\Delta N = \frac{12}{7}\frac{8\pi}{A\lambda^2}\int_{-\infty}^{\infty}\frac{g(X)}{2}\mathrm{d}X, \quad (6)$$

the temperature

$$T = \left(\frac{\Delta\nu_{\mathrm{D}}}{14.49}\right)^2, \quad (7)$$

the flow velocity

$$v = \frac{\delta\nu\lambda}{\sin\varphi}. \quad (8)$$

With a knowledge of the temperature and the absolute velocity of the active medium, it is possible to determine the flow Mach number:

$$M = v\left(\frac{\mu}{kRT}\right)^{1/2} = 14.49\frac{\delta\nu\lambda}{\Delta\nu_{\mathrm{D}}\sin\varphi}\left(\frac{\mu}{kR}\right)^{1/2}, \quad (9)$$

where μ is the average molar weight of the active medium; R is the universal gas constant; $k = 1.4$ is the adiabatic index.

3. Results

The dependence of the parameters of the active medium on the flow rate of primary nitrogen appears to be most significant, for it is precisely this dependence that shows the maximum recovered pressure attainable with simultaneous retention of the conditions necessary for efficient lasing. The limiting pressure recoverable with a supersonic diffuser of constant section area at a pseudoshock is approximately equal to p_4 [5]. In this series of experiments, we recorded the chlorine flow rate through the JGSO $G_0 = 39.2$ mmol s^{-1}, the flow rate of secondary nitrogen $G_2 = 11$ mmol s^{-1}, the flow rate of molecular iodine $G_{\mathrm{I2}} = 0.8$ mmol s^{-1}, and the temperature of the solution in the JGSO $t = -16\,^{\circ}$C, while the variable quantity was the flow rate of primary nitrogen G_1. As G_1 was varied from 150 to 530 mmol s^{-1}, the pressure in the prechamber in front of the nitrogen nozzles varied from 480 to 1450 Torr and the near-wall pressure p_3 in the mixing chamber varied from 7 to 11 Torr.

Figs 3 and 4 show the most important parameters of the active medium in the mixing chamber as functions of the flow rate of primary nitrogen. It turned out that the inverse population density decreases with the flow rate of primary nitrogen, this reduction being approximately proportional to that of the gain. Consequently, the narrowing of the Doppler width of the amplification spectrum is compensated for by the growth of the collisional width with increase in the flow rate of primary nitrogen.

Increasing the flow rate of primary nitrogen is attended with a growth of its pressure at the lateral section of cylindrical nozzles, and the outflow of primary nitrogen becomes progressively different from the calculated one, i.e., the ratio between the pressure at the lateral section of cylindrical nozzles and the pressure in the mixing chamber increases. (The theory of development of the initial portion

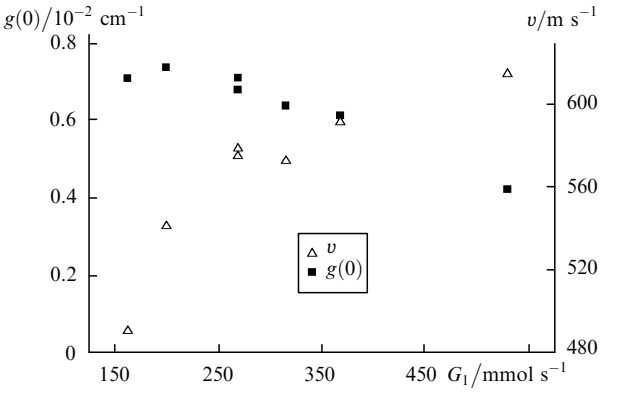

Figure 3. Gain $g(0)$ and velocity of the active medium v as functions of the flow rate G_1 of primary nitrogen.

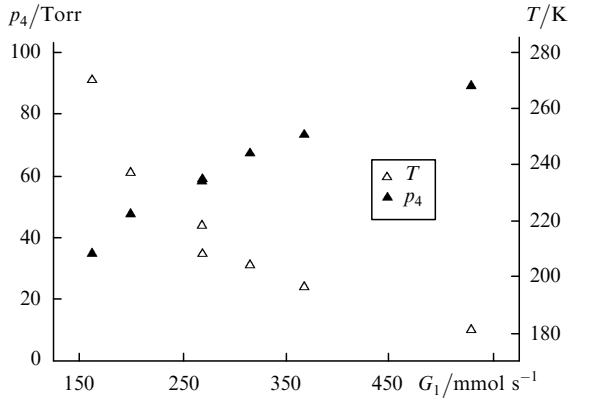

Figure 4. Temperature T and pressure p_4 as functions of the flow rate G_1 of primary nitrogen.

of underexpanded supersonic jets is outlined in detail in Ref. [6]). This results in a stronger compression of low-pressure jets of oxygen and nitrogen–iodine mixture located between the jets of primary nitrogen. In this case, the pressure in the jets of primary nitrogen decreases and the compression of the subsonic flow of the $O_2-I_2-N_2$ mixture causes its velocity to increase.

Some distance away from the nozzle array, the pressure in the jets of primary nitrogen becomes lower than the pressure in the low-pressure flow of the $O_2-I_2-N_2$ mixture, its expansion terminates, and the $O_2-I_2-N_2$ flow acquires a sonic velocity. There commence the reverse compression of the primary nitrogen flow attended with origination of shock waves and the expansion of the $O_2-I_2-N_2$ flow, which is accelerated to the sonic velocity. Subsequently the process of compression and rarefaction of the flows can be repeated over and over again. The higher the flow rate of primary nitrogen, the stronger the initial compression and higher the $O_2-I_2-N_2$ flow pressure and the lower the flow velocity prior to its first compression.

This is confirmed in the experiment by the growth of the pressure p_2 from 25 to 35 Torr as the flow rate of primary nitrogen is increased from 163 to 530 mmol s^{-1}. The higher the $O_2-I_2-N_2$ mixture density and the longer the time period during which it does not experience compression, the higher the losses of $O_2(^1\Delta)$. This can explain the reduction in gain and inversion density with increase in the flow rate of primary nitrogen. The increase in absolute velocity and the reduction in the temperature of the active medium lead to

the growth of the Mach number, calculated by formula (9), from 1.53 to 2.27 as the flow rate of primary nitrogen is increased from 163 to 530 mmol s^{-1}.

Fig. 5 shows the results of measurements of the gain, the temperature, and the collisional width along the flow employing the second optical configuration ($\varphi = 0$). In this case, the flow rates of secondary nitrogen $G_2 =$ 11 mmol s^{-1} and molecular iodine $G_{I2} = 0.8$ mmol s^{-1} as well as the solution temperature $t = -16\,^{\circ}C$ were fixed. One can see that the maximum gain is reached at distances z of less than 44 mm from the nozzle unit. The temperature of the active medium is hardly changed with distance to the nozzle unit ($dT/dz \approx 0$) for both values of the flow rate of primary nitrogen. From the functional pressure dependence of the collisional width $\Delta\nu_L = \alpha(T)p$ and the approximate equality $dT/dz \approx 0$ it follows that

$$\frac{d\Delta\nu_L}{\Delta\nu_L dz} \approx \frac{dp}{p dz},$$

and from the equation of state of a gas $p = NRT$, where N is the molar gas density, it follows that

$$\frac{dN}{N dz} = \frac{dp}{p dz} - \frac{dT}{T dz} \approx \frac{d\Delta\nu_L}{\Delta\nu_L dz}.$$

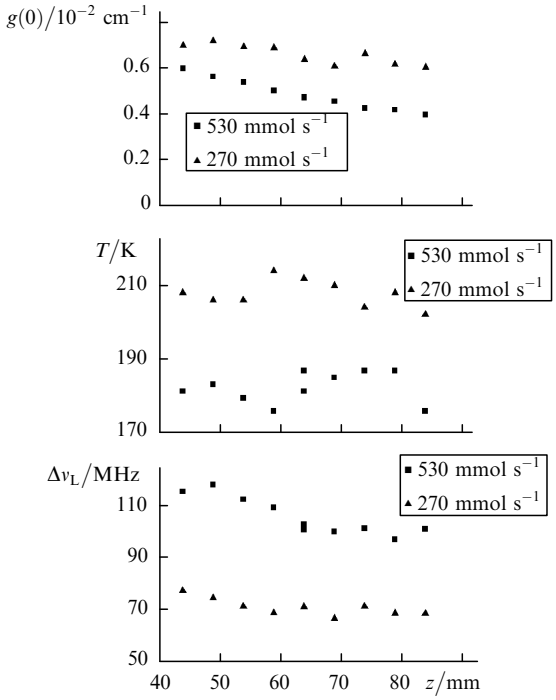

Figure 5. Parameters of the active medium versus distance to the nozzle unit for different G_1.

Since the relative reduction of the collisional width over distances of 64–84 mm along the flow is less than 10 %, the relative reduction of the static pressure and density will be the same. Consequently, a faster decline of the gain along the flow cannot be attributed to the lowering of the density of the active medium alone. Increasing the flow rate of primary oxygen from 270 to 530 mmol s^{-1} results in a faster decrease of the inversion and the gain along the flow. This is attributable in part to a greater rarefaction of gas in the expanding mixing chamber owing to a reduction of the thickness of the boundary layer with increase in the flow rate of primary nitrogen.

Measurements of the parameters of the active medium along the flow were also made for a higher temperature $t = -5.6\,^{\circ}C$ of the alkaline solution of hydrogen peroxide in the JGSO and for a lower flow rate of molecular iodine. The results of measurements of the parameters of the active medium at a distance $z = 64$ mm from the nozzle unit are given in Table 1. In all cases, the maximum gains were achieved at $z < 44$ mm; the temperature was nearly constant along the flow. The water vapour content in the oxygen flow affected only the gain and the temperature: in these cases, too, the rate of gain decay along the flow was, for the same flow rate of primary nitrogen, virtually the same as for $t = -16\,^{\circ}C$ and an iodine flow rate of 0.8 mmol s^{-1}.

4. Discussion of results

It follows from the results obtained that the main processes responsible for the dissociation of molecular iodine, the heat release in the flow of the active medium, and the production of the gain in the active medium take place at distances of less than 44 mm from the nozzle unit in the region of subsonic flow of the $O_2 - I_2 - N_2$ mixture. Then there occurs a fast mixing of $O_2 - I_2 - N_2$ with the high-pressure supersonic flow of primary nitrogen, with the result that the temperature of the active medium lowers drastically.

As revealed by previous experiments, an efficient mixing of the flows takes place at distances of less than 35 mm from the nozzle unit [1]. Subsequently the active medium moves with a velocity $v \approx 600$ m s^{-1} without a noticeable lowering of its temperature, which is testimony to a drastic decrease of the rates of kinetic processes which result in the heat release in the active medium. Water vapour and molecular iodine convert the electron energy stored in $O_2(^1\Delta)$ to heat via the quenching of electronically excited iodine atoms $I(^2P_{1/2})$:

$$O_2(^1\Delta) + I(^2P_{3/2}) \rightarrow O_2(^3\Sigma) + I(^2P_{1/2}),$$

$$I(^2P_{1/2}) + I_2 \rightarrow I(^2P_{3/2}) + I_2,$$

$$I(^2P_{1/2}) + H_2O \rightarrow I(^2P_{3/2}) + H_2O.$$

Table 1.

G_1/mmol s^{-1}	t/°C	G_{12}/mmol s^{-1}	$g(0)/10^{-2}$ cm^{-1}	T/K	$\Delta\nu_L$/MHz	$\Delta N_0/10^{14}$ cm^{-3}	p_3/Torr	p_4/Torr
270	–16	0.8	0.64	212	71	9.6	8.5	58
270	–5.6	0.8	0.51	232	86	8.2	9.3	65
530	–16	0.8	0.47	181	100	7.5	11	88
560	–5.6	0.8	0.3	204	112	5.2	12	85
560	–16	0.4	0.25	164	116	4	10.8	81

The results obtained show that the increase in water vapour content in the oxygen flow is responsible for a temperature rise and a lowering of the gain in the active medium, but does not affect the character of their behaviour. Reducing the flow rate of molecular iodine results in a lowering of the temperature of the active medium, but does not exert effect on the rate of variation of the gain along the flow.

The rapid decline of gain and inversion along the flow can be explained by the lowering of $O_2(^1\Delta)$ content or atomic iodine density. As shown by an analysis of the kinetics of the oxygen–iodine medium, the lowering of $O_2(^1\Delta)$ content along the flow cannot be the cause of so rapid a decline of the gain. In addition, this is indirectly confirmed by the facts that the temperature of the active medium does not rise along the flow and the decay of gain along the flow is independent of the initial content of water vapour and the flow rate of molecular iodine. Since the pressure and the total density decrease along the flow considerably slower than the gain, the atomic iodine density can lower only due to their adherence to the aerosol surface or recombination. Aerosol can be present in the oxygen flow due to carrying away of the working solution from the JGSO or can be produced due to condensation of water vapour and the residual molecular iodine upon mixing with the flow of cold primary nitrogen.

The flow temperature was calculated by formula (7) under the assumption that no additional broadening mechanism makes a contribution to the Gaussian component $\Delta\nu_D$ of the line spectrum. When the motion of the active medium is turbulent, chaotic pulsations of the transverse velocity should bring about an additional broadening of the amplification spectrum, which can make contributions both to the Lorentzian and Doppler components if the distribution of the turbulent velocity pulsations is non-Gaussian. The average value of flow velocity pulsations for $v \sim 600$ m s^{-1} can be comparable with the average thermal velocity of heavy iodine atoms at a temperature of ~ 200 K [6, 7].

The transverse components of the flow velocity, which originate as periodic compression or rarefaction waves in the interaction of supersonic gas jets, can also be responsible for additional broadening of the amplification spectrum. If it is assumed that there are no additional mechanisms of spectral broadening, the resultant data would suffice to calculate the flow deceleration parameters – the temperature $T^* = T(1 + 0.2M^2) = T + (k-1)\mu v^2/(2kR)$ and the pressure $p^* = p_3(1+ 0.2M^2)^{3.5}$. With the knowledge of the initial values of the temperature and flow rates of all the gases and the deceleration temperature T^*, it is possible to calculate the thermal power released in the active medium and determine the losses of $O_2(^1\Delta)$ in the course of its production. Under these assumptions, our estimates made proceeding from the results given in Figs 3 and 4 show that the losses of $O_2(^1\Delta)$ increase with increase in flow rate of primary nitrogen. This is evidence in favour of the assumption that raising the flow rate of primary nitrogen increases the duration of stay and the pressure of the $O_2 - I_2 - N_2$ mixture in the region of its subsonic flow, where the iodine dissociation and the $O_2(^1\Delta)$ quenching occur for the greater part.

5. Conclusions

The diode laser spectroscopy of the $^2P_{1/2}(F = 3) \rightarrow {}^2P_{3/2}$ $(F = 4)$ working laser transition of atomic iodine allows a determination of not only the gain and the temperature of the active medium of an oxygen–iodine laser, but its gas-dynamic parameters as well, the absolute flow velocity in particular. In the active medium produced by an ejector nozzle unit with dilution of oxygen by primary nitrogen in the ratio of 1:6.9, the gain coefficient amounts to 7×10^{-3} cm^{-1}, the temperature of the active medium is 200 K, the absolute flow velocity ~ 560 m s^{-1}, and the pressure about 58 Torr.

As the dilution is increased to 1:13.5, the gain reduces to 4.5×10^{-3} cm^{-1}, the temperature drops to 180 K, the velocity of the active medium rises to 615 m s^{-1}, and the pressure rises to 88 Torr. The amplification in the active medium is produced by an ejector nozzle unit at distances of less than 44 mm and then decays monotonically, and the higher the flow rate of primary nitrogen, the stronger the decay. Increasing the initial content of water vapour lowers the gain but does not change its decay rate along the flow, and increases the temperature of the active medium. In all nozzle-unit operating modes investigated, the temperature of the active medium is hardly changed along the flow direction.

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