CHARACTERISTICS OF A PHOTODISSOCIATION LASER BASED ON METHYL IODIDE

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Methyl iodide was one of the first substances yielding generation at the $5^2P_{1/2} \rightarrow 5^2P_{3/2}$ transition of the iodine atom (wavelength $\lambda = 1.315$ μ) on the basis of the pulsed-photolysis method [1]. It was subsequently shown [2, 3] that the pressures of methyl iodide most suitable for generation were much smaller than those of other compounds, while the energy yield in the generation pulse was also lower. This is evidently why subsequent investigations were mainly carried out with perfluoroalkyl compounds, as a result of which hardly any data relating to the energy and time characteristics of generation during the photolysis of CH_3I are available. All that can be said is that an oscillogram of a generation pulse some 5 μsec long with a power of the order of 10 W was presented in [4].

In this investigation we set ourselves the task of obtaining more detailed information regarding the time and energy characteristics of generation during the photolysis of CH_3I; we study the effect (observed in a number of investigations – see [5]) of pumping the working substance through the system on these characteristics. For gasdynamic lasers the development of an inverted population is closely associated with the kinetics of the reduction in temperature of the working medium during the adiabatic expansion of the gas. For electrical-discharge lasers of high power the convective cooling of the gas due to pumping improves their energy characteristics. For lasers which involve excitation during photodissociation no investigations have yet been carried out with a through current of the working substance. In this paper we shall present some experimental results obtained during the operation of a pulsed laser without any through current and also in the presence of a transverse current of the working mixture through the resonator volume. The working substances are methyl iodide CH_3I and a mixture of this with helium.

The experiments were carried out in the apparatus schematically depicted in Fig. 1, where 1 are pulse lamps, 2 is the vacuum chamber, 3 are the reflectors, 4 is the supply line, 5 is the ignition line, 6 is a plexiglas window, 7 are mirrors, and 8 is the collector. The working substance was pumped by two series-connected IFP-5000 pulse lamps arranged at 23 mm from one another within the vacuum chamber. In order to increase the flow of pumping radiation, Duralumin reflectors were employed. The resonator cross section was approximately equal to 4.5 cm^2. The methyl iodide was passed into the interior of the vacuum chamber through four slots cut along a steel tube. The size of each slot was 0.2×60 mm^2, the internal diameter of the steel tube was 15 mm, the length was 400 mm, and the distance from the slot to the resonator axis was 18 mm. During the flow of the working substance the chamber was evacuated by means of a VN-6 pump giving a limiting vacuum of ≤ 0.05 mm Hg. The pressure in the chamber was measured with an oil manometer and a standard vacuum gage; a supplementary vacuum gage measured the pressure inside the slotted tube.

In the laser apparatus one of the resonator mirrors had a compact gold coating and was spherical with a radius of curvature $R = 300$ cm, the other being plane and having a dielectric coating with a transmission of 2% at the generation wavelength of $\lambda = 1.315$ μ. The mirrors lay at a distance of 176 cm from one another at the ends of the chamber. Under typical conditions the pumping energy was 600 J ($C = 33.3$ μF, $U = 6$ kV). The length of the pumping pulse was measured with a photocell of the F-1 type having UFS-1 and UFS-5 light filters and the generation period, with an FÉU-28 photomultiplier and a low-resistance load, so that the time constant was less than 1 μsec. In the measurements we used an S1-51 double-beam oscillograph (with memory), and in order to weaken the signal we added light filters of the NS type. The duration of the pumping pulse at half-intensity level was 80-90 μsec, and the period of generation was 10-20 μsec with a delay of 30-40 μsec relative to the onset of lamp operation. The energy yield of the generation was measured with an IMO-2 instrument and

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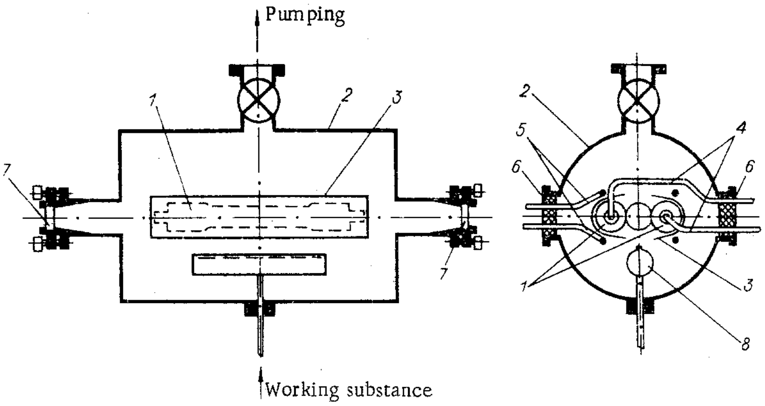

Fig. 1

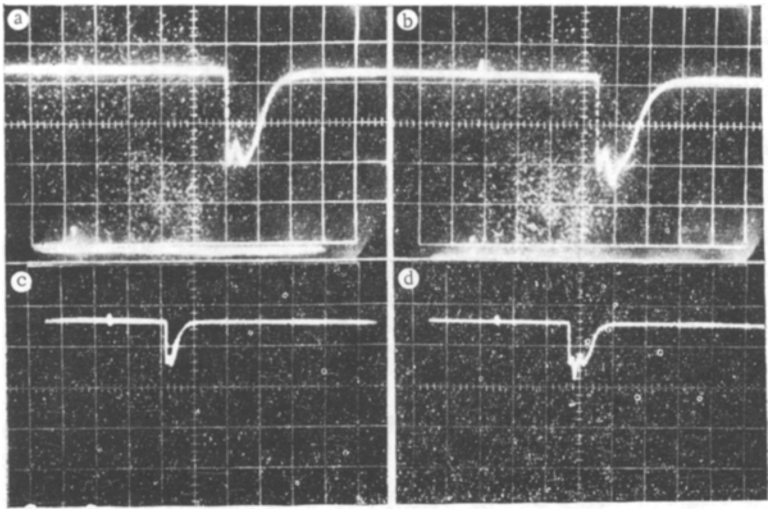

Fig. 2

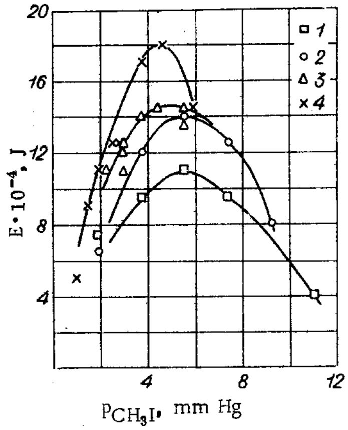

Fig. 3

in order of magnitude amounted to 1-2 mJ. Typical generation pulse oscillograms appear in Fig. 2: a) CH_3I without flow, sweep 10 μsec/division, energy 0.9 mJ; b) CH_3I with flow, sweep 10 μsec/division, energy 1.1 mJ; c) CH_3I : He mixture without flow, sweep 25 μsec/division, energy 1.2 mJ; d) CH_3I : He mixture with flow, sweep 25 μ sec/division, energy 2 mJ. From the time and energy data we calculated the mean power in the generation pulse. Depending on the experimental conditions the average power varied over the range 50-100 W.

Let us give some more detailed consideration to the gasdynamic conditions under which the experiments took place. Table 1 shows the pressures p_1 of the working medium in the tube before outflow from the slots and

TABLE 1

p_1, mm Hg	p_2, mm Hg	G, g/sec	v, m/sec	T, °K	Composition
9	7	0,20	86	280	CH_3I
26	5	0,65	137	261	CH_3I
88	26	1,07	325	232	CH_3I : He 1:6
70	29	0,85	372	231	CH_3I : He 1:7
99	44	1,20	372	231	CH_3I : He 1:7

the pressure in the working chamber p_2 during the flow, indicating the typical experimental values during the measurements. Using p_1 and p_2 and knowing the cross section of the slots carrying the gas flow and the corresponding equations of gasdynamics [6], we determined the gas flow in unit time G, the rate of outflow of the gas from the slots v, and the gas temperature T for conditions of adiabatic expansion. The values of G, v, and T are also given in Table 1. For control purposes we made an experimental determination of the gas flow based on the reduction in its pressure in the original calibrated volume. The experimental data lay very close to the calculated values.

Calculation of the Reynolds number for the outflow of CH_3I and mixtures of the latter with He through the slot under the experimental conditions in question gives values of $Re \approx 100$-400. According to [7] the outflow of a plane jet may only be regarded as laminar for $Re < 30$. Hence, in order to calculate the outflow geometry of the gas stream and the velocity distribution we used the theory of turbulent jets [8]. In particular, for the dependence of the axial gas velocity on the coordinate in the main part of the jet we took the approximate Van der Hegge-Zijnen equation $u_{max}(x)/u_0 = 2.48/(x/d + 0.6)^{1/2}$, where $u_{max}(x)$ is the stream velocity in the direction of outflow (x is reckoned from the slots and $x/d > 10$); u_0 is the initial stream velocity; and d is the slot width. The divergence angle of the jet boundaries on reducing the gas velocity by a factor of two [8] was taken as 11°. For a distance of 18 mm from the slot to the axis of the resonator and a slot width of 0.2 mm we found that the width of the jet close to the resonator axis was 4 mm, while the maximum rate of flow was 1/4 of the rate of gas outflow from the slot. These figures were used for estimating the through flow of the working substance during the generation pulse.

The experimental results are shown in Fig. 3: 1) CH_3I without flow; 2) $CH_3I : He = 1 : 7$ mixture without flow; 3) CH_3I with flow; 4) $CH_3I : He = 1 : 7$ mixture with flow. The horizontal axis gives the partial pressure of CH_3I in the chamber, and the vertical axis gives the energy of the generation pulse obtained under various experimental conditions. The results are given for pure CH_3I and for a CH_3I : He mixture in molar proportion 1 : 7. Similar qualitative results are obtained for 1 : 6 and 1 : 8 mixtures. In the pressure range $p_{CH_3I} = 2 - 10$ mm Hg studied the curves have a maximum in the region 4-6 mm Hg. The existence of the maximum may be understood from a consideration of limiting cases. For low CH_3I pressures the optical density of the medium falls and the absorption of pumping energy in the reflectors increases. A reduction in the absorption of pumping energy by the medium leads to a reduction in the energy of the generation pulse. For high CH_3I pressures the frequency of the inelastic collisions of the excited I* atoms with the CH_3I molecules increases, which is the main reason [9] for the deactivation of the I* atoms. This leads to a reduction in generation energy at high pressures.

Comparison of the Fig. 3 curves shows that the lowest energy yield occurs for pure CH_3I without flow-through. The mixture of methyl iodide with helium under static conditions and pure CH_3I under flow conditions gives approximately the same pressure dependence, the energy maximum being 1.4 times greater than that of curve 1. The greatest energy yield is obtained for a flow of CH_3I : He through the resonator (relative increase 1.8-2 times relative to curve 1).

Analysis of all the oscillograph data led to the following results. For a flow of CH_3I through the resonator there was a slight lengthening of the generation pulse by 1-3 μsec. The energy increment compared with the data obtained without the CH_3I flow was due to the rise in average generation power. For a flow of CH_3I : He mixture the lengthening of the generation pulse was more considerable and varied in different experiments from 2 to 10 μsec. However, there was an increment in the mean power by comparison with the generation of the mixture in the absence of flow in this case also. The characteristic delay times and generation pulse lengths (40 and 20 μsec) are too short for the macroscopic motion of the medium through the resonator to lead to any appreciable displacement of the medium. An estimate of this linear displacement from the foregoing relationships leads to a value of the order of tenths of a millimeter. Thus, the total irradiated mass of CH_3I molecules increases by no more than 5% on account of the flow. This estimate (together with the increment in the average power of generation) leads to the conclusion that when the substance flows through the resonator there is a change in the threshold characteristics of the active medium, promoting an increased energy in the generation pulse.

It is natural to associate such changes with temperature changes of the whole working medium. Let us therefore give some further attention to energy evolution during the photodissociation of the CH_3I molecules. In an individual act of photodissociation $h\nu = E_1 + E_2 + E_3$, where $h\nu$ is the energy of the pumping quantum; E_1 is the dissociation energy of the CH_3I molecules with respect to the C–I bond; E_2 is the excitation energy of the I atom; and E_3 is the excess energy of the dissociation products, ultimately leading to an increase in the temperature of the medium. Starting from the known values of $h\nu \approx 5$ eV, $E_1 = 2.5$ eV and $E_2 = 0.94$ eV we find that $E_3 \approx 1.5$ eV. An estimation of the temperature rise of the medium without helium during the generation pulse for a degree of dissociation of the medium equal to 2-3%† leads to a change of $\Delta T = 40\text{-}60°$. Thus the energy evolution within the volume of the resonator due to the primary act of photodissociation during the generation pulse leads to slight heating of the substance. The velocities of the secondary recombination processes at the working pressures are low and make no contribution to the heating of the medium over these periods. In the experiments without flow-through the role of helium in raising the energy yield of generation evidently amounts to the fact that as a result of elastic collisions it accelerates the "cooling" of the excited iodine atoms, which at the instant of the dissociation of the CH_3I acquire an excess kinetic energy. The reduction in the Doppler linewidth of the radiation from the I* atoms due to this "cooling" increases the amplification factor of the active medium and raises the energy yield in the generation pulse.

In the experiments involving the flow of the working substance through the resonator the gas temperature falls slightly during its adiabatic expansion after passing the slots. Table 1 gives the gas temperatures, from which we see that for CH_3I the fall in temperature (from room level) is 20-30°, while for the CH_3I : He mixture it reaches 60°. For a qualitative explanation of the observed changes in generation energy with and without a flow of working substance we may reasonably assume that on reducing the translational temperature in the gas flow the frequency of the inelastic collisions between the excited iodine atoms and the CH_3I molecules diminishes. For quantitative estimates we should have to know how rapidly equilibrium is established between the temperature of the excited iodine atoms and the temperature of the medium with and without the presence of helium. However, this problem demands a separate investigation.

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†The degree of dissociation was determined from data relating to the energy yield in the generation pulse, allowing for the loss of particles along the channel of inelastic collisions and the threshold inverted population.