Efficient pulsed chemical oxygen-iodine laser with instantaneous generation of atomic iodine by volumetric discharge

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A pulsed chemical oxygen-iodine laser (COIL) using atomic iodine generated by volumetric discharge of CH₃I is developed and tested. COIL with a gain length of 60 cm is energized by a square pipe-array jet singlet oxygen generator with basic hydrogen peroxide pumping circulations and operated at subsonic gas flow. Maximum output energy of 4.3 J, pulse duration of 50 μ s, specific energy extraction from the active medium of 2.0 J/L, and the maximum chemical efficiency of 12.5% are achieved at a chlorine flow rate of 55 mmole/s.

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A chemical oxygen-iodine laser $(COIL)^{[1]}$ can operate in the continuous wave (CW) and pulsed regimes. The latter can be achieved either by Q-modulation of the gain in a CW COIL or by instantaneous volumetric iodine atom generation. Each of these techniques has some advantages and disadvantages. In the US, Q-modulation has been studied experimentally and operated at several, even tens, of kilohertzs and maximum pulse energy of 0.3 $J^{[2-4]}$. According to Refs. [5-8] instantaneous volumetric iodine generation can be achieved either by means of photolysis or electric discharge dissociation of iodine. Photolysis to produce iodine atoms^[9,10], although able to realize a specific output energy from 2.8 to 3.1 J/L and pulse duration of 2 ms with a small-scale COIL chlorine flow rate of 17 mmole/s, has low electric efficiency (ratio of laser output energy to electric energy), which makes it difficult to scale up. However, the electric efficiency of pulsed discharge is up to $91\%^{[5]}$, which is approximately 300 times higher than that of photolysis. Pulsed COILs based on electric discharge dissociation of iodine have been investigated using small-scale COIL with chlorine flow rate of 14 mmole/s^[5]. Recently, Refs. [11,12] reported a pulsed COIL initiated by discharge at chlorine flow rates of 39, 65, 92, and 105 mmole/s, in which the pulse energy attained 1.1 J, specific energy extraction from the active medium was 1.7 J/L, and chemical efficiency was 6%-3%. In this letter, we report on the demonstration of an efficient subsonic pulsed COIL utilizing iodine atoms generated by volumetric discharge.

Figure 1 shows the experimental setup. Our pulsed COIL system consists of a square pipe-array jet-type singlet oxygen generator (SOG) that includes a basic hydrogen peroxide (BHP) cooling and circulation system, a CH₃I supplier and injector, a laser cavity combined with discharge electrodes, and a vaccum pump system. Experimental results of the square-pipe-array jet SOG (JSOG) have been reported for our previous small-scale JSOG^[13] with nitrogen as buffer gas. In our experiments, the square pipe-array jet-type SOG operated at

chlorine flow rate of 50–60 mmole/s, primary nitrogen of 100–200 mmole/s, and SOG pressure of 20–25 Torr. At the outlet of the SOG, CH₃I was delivered from a tank heated up to 50-70 °C to the main gas flow through the gas injector with the aid of the secondary nitrogen buffer gas. A stable resonant cavity of 160 cm, including gain length of 60 cm, was used, with a transmission of 2.8%for the plane output mirror and a curvature radius of 2.5 m for the reflective mirror. The gain medium, with the pressure of 18–20 Torr, was located at the pulsed gas discharge chamber measuring $60 \times 5 \times 7$ (cm), in which iodine atoms by two electrode of area of $60 \times 5 \text{ cm}^2$ and a gap of 7 cm between the electrodes were produced. In order to realize the homogenous glow discharge, 52 sliding discharge pins inlaid in the cathode were used for pre-ionization, which was important for the discharge of negative gases^[14,15], as described details in Ref. [16].</sup> The storage capacitance was 67.2 nF and loaded 17-kV voltage. The frequency of pulse operation was in the range of 1–5 Hz. Deposited energy was approximately 4– 5 J, and deposition electric efficiency was approximately 40%-60%. The pulsed output energy was measured by an energy detector (J-50MB-YAG-1535, Coherent. Co. Lt.), whereas the pulse duration measured by a Ge detector (New Focus Co. Lt.)

Because the chemical efficiency of CW COIL is defined as the percentage of laser output power over the most power, we suppose that chlorine first completely changes



Fig. 1. Schematic of the experimental setup.

to singlet oxygen and then completely changes to excited iodine atoms:

$$\eta_{\rm chem} = \frac{P_{\rm cw}}{91.4 (\rm kJ/mol) \times f_{\rm Cl_2}},\tag{1}$$

where 91.4 kJ/mol is the energy stored in 1 mol of excited iodine atom, $f_{\rm Cl_2}$ is the flow rate of chlorine (mol/s), and $P_{\rm cw}$ is the laser output power.

The chemical efficiency for a pulsed COIL based on instantaneous volumetric iodine atom generation can be defined as the percentage of laser pulse energy extracted from the maximum energy in the discharge volume, supposing that chlorine completely first changes to singlet oxygen and then completely changes to excited iodine atoms:

$$\eta_{\rm chem} = \frac{E_{\rm pulse}}{5.37({\rm J/Torr}\cdot {\rm L}) \times P_{\rm O_2} \times V_{\rm dis}},\qquad(2)$$

where 5.37 J/(Torr·L) is the energy in 1 Torr and 1 L of excited iodine atom at a standard state, which equals 91400/ (760×22.4) because the temperature is at standard state temperature; $E_{\rm pulse}$ is the laser pulse energy; $P_{\rm O_2}$ is the partial pressure of oxygen in the cavity, supposing that chlorine completely changes to (singlet) oxygen; $V_{\rm dis}$ is volume of discharge.

Bun	n Cl ₂ Flow . Rate (mmole/s)	Primary	Secondary	$CH_{3}I$	Pressure	Pressure	Output Energy (J)	Chemical Efficiency (%)
No		N_2 Flow	N_2 Flow	Flow	in the	in the		
NO.		Rate	Rate	Rate	Cavity	SOG		
		(mmole/s)	(mmole/s)	(mmole/s)	(Torr)	(Torr)		
1	52.2	110.0	172.7	3.80	19.6	21.4	3.17	9.4
2	52.1	111.0	173.9	3.85	19.7	22.4	3.02	9.0
3	51.7	115.7	153.0	3.93	18.1	20.9	3.04	9.5
4	51.3	133.0	132.8	3.94	18.3	22.7	3.01	9.2
5	54.0	133.0	133.0	3.94	18.3	23.7	3.07	9.0
6	56.3	133.0	157.0	3.96	20.7	25.0	3.20	8.6
7	58.4	133.0	157.0	3.97	20.7	23.5	3.20	8.4
8	57.5	157.0	133.0	3.98	20.3	25.0	3.03	8.2
9	54.4	174.7	111.6	3.80	19.5	25.2	4.29	12.5
10	54.0	111.8	174.2	3.80	19.7	24.1	3.40	9.9
11	53.4	111.2	174.3	3.90	19.6	24.1	3.20	9.4

Table 1. Summary of the Main Experimental Results

The main experimental results are listed in Table 1. We can see that the pulse output energy is in the range of 3.0–4.3 J at chlorine flow rate of 52–58 mmole/s, and that the chemical efficiency is 8.2%–12.5%. To our knowledge, the results of COIL, which adopts transverse electric discharge and tends to scale up, show the most efficient pulsed COIL initiated by volumetric discharge. This result may be due to the improvement of the SOG efficiency and the performance of pre-ionized technique^[16].



Fig. 2. Pulse energy measured by the coherent energy detector.



Fig. 3. Pulse profile measured by the new focus detector.

The measured pulse energy is shown in Fig. 2. We can see that the laser pulses are 29 and nearly stable at 8-s operation time. The pulse profile measured by Ge detector is presented in Fig. 3. The pulse duration was approximately 50 μ s.

In conclusion, we develope and test a pulsed multijoule COIL with iodine atoms generated by volumetric discharge of CH_3I . This subsonic pulsed COIL system is demonstrated with high chemical efficiency. The maximum output energy is 4.3 J per pulse, and specific energy extraction from the active medium of 2.0 J/L is obtained. The potential maximum chemical efficiency is as high as 12.5% at a chlorine flow rate of 55 mmole/s.

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