

Gain and continuous-wave laser power enhancement with a secondary discharge to predissociate molecular iodine in an electric oxygen-iodine laser

G. F. Benavides,¹ J. W. Zimmerman,² B. S. Woodard,² D. L. Carroll,^{1,a)} J. T. Verdeyen,¹ T. H. Field,¹ A. D. Palla,¹ and W. C. Solomon²

¹CU Aerospace, 2100 S. Oak St., Suite 206, Champaign, Illinois 61820, USA

²University of Illinois at Urbana-Champaign, 306 Talbot Laboratory, 104 S. Wright St., Urbana, Illinois 61801, USA

(Received 4 December 2007; accepted 9 January 2008; published online 31 January 2008)

Herein the authors report on the demonstration of a 50% enhancement in gain and 38% enhancement in continuous-wave laser power on the 1315 nm transition of atomic iodine through the addition of a secondary discharge to predissociate the molecular iodine in an electric oxygen-iodine laser. In the primary discharge the $O_2(a^1\Delta)$ is produced by a radio-frequency-excited electric discharge sustained in an O_2 -He-NO gas mixture, and $I(^2P_{1/2})$ is then pumped using energy transferred from $O_2(a^1\Delta)$. A gain of $0.10\% \text{ cm}^{-1}$ was obtained and the total laser output power was 6.2 W. © 2008 American Institute of Physics. [DOI: 10.1063/1.2839323]

The classical chemical oxygen-iodine laser (COIL) first reported by McDermott *et al.*¹ operates on the electronic transition of the iodine atom at 1315 nm, $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$ (denoted hereafter as I^* and I , respectively). The lasing state I^* is produced by near resonant energy transfer with the singlet oxygen metastable $O_2(a^1\Delta)$ [denoted hereafter as $O_2(^1\Delta)$]. The conventional COIL requires a chemical two-phase process to produce the $O_2(^1\Delta)$ from aqueous basic hydrogen peroxide and Cl_2 gas. Logistic issues with this chemical singlet oxygen generator motivated many investigations into an electrically driven oxygen-iodine laser (ElectricOIL) that was first demonstrated by Carroll *et al.*^{2,3} Subsequent efforts have demonstrated gain⁴⁻⁶ and lasing^{5,6} in other ElectricOIL configurations since the first demonstrations.^{2,3} For an excellent and comprehensive topical review of discharge production of $O_2(^1\Delta)$ and ElectricOIL studies, see Ionin *et al.*⁷ In these prior ElectricOIL experiments investigators relied upon O atoms to provide the predominant mechanism for iodine dissociation. However, with the desire to push the system to higher flow pressures there are fewer O atoms available due to recombination. Thus, to achieve more complete dissociation in ElectricOIL, the use of an iodine predissociator is of particular importance at higher flow pressures. Carroll and Solomon⁸ first hypothesized and calculated that this technology improvement would be very important for the ElectricOIL system to maximize the energy transfer of $O_2(^1\Delta)$ into atomic iodine, and consequently significantly enhance gain and laser power output.

In this letter, the authors report on the demonstration of a 50% enhancement in gain and 38% enhancement in continuous-wave laser power on the 1315 nm transition of atomic iodine through the addition of a secondary discharge to predissociate the molecular iodine in an electric oxygen-iodine laser. In the primary discharge the $O_2(a^1\Delta)$ is produced by a radio-frequency-excited electric discharge sustained in an O_2 -He-NO gas mixture, and $I(^2P_{1/2})$ is then

pumped using energy transferred from $O_2(a^1\Delta)$. A block diagram of the flow tube setup is shown in Fig. 1. The primary radio frequency (rf) electric discharge at 13.56 MHz operating between two transverse electrodes was used as the excitation source for the production of $O_2(a^1\Delta)$. The plasma zone was approximately 4.9 cm in diameter and 25 cm long. More information on the performance of this transverse electric discharge sustained in an O_2 -He-NO gas mixture can be found in Benavides *et al.*⁹ A secondary rf discharge was placed at the exit of the iodine injection holes using electrodes imbedded in injector blocks fabricated out of the machinable ceramic Macor®.

The supersonic diagnostic cavity has a Mach 2 nozzle with purged mounts into which windows can be placed for measurement of the gain or laser mirrors can be placed for laser oscillation. The subsonic diagnostic duct has multiple purged windows through which simultaneous measurements are made of the optical emission from $O_2(^1\Delta)$ at 1268 nm and $O_2(b^1\Sigma)$ at 762 nm. A Roper Scientific Optical Multi-channel Analyzer (OMA-V) with a 1024-element InGaAs LN_2 cooled array interfaced to an Acton Research SP-2300i monochromator was used for measurements at 1268 nm. A thermoelectric-cooled Apogee charge-coupled device is used to measure spectra of the $O_2(b^1\Sigma)$ transition at about 762 nm. Both instruments are fiber coupled to enable instrument positioning flexibility and excellent measurement repeatability.

Micro-Motion CMF and Omega FMA mass flow meters were used to measure the flow rates of the gases. The I_2

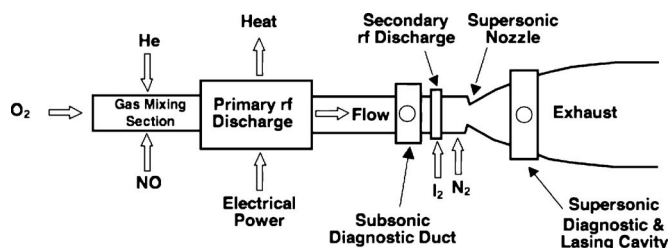


FIG. 1. Schematic of the experimental apparatus.

^{a)} Author to whom correspondence should be addressed. Electronic mail: carroll@cuaerospace.com.

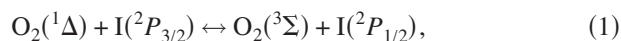
concentration was measured by a method developed by Physical Sciences Inc. and is based on the continuum absorption of molecular iodine at 488 nm. Details of this diagnostic are described by Rawlins *et al.*¹⁰ Pressure in the subsonic and supersonic flow regions were measured by capacitance manometers from MKS and Leybold.

Measurements of gain (or absorption) were made prior to running the apparatus as a laser using the iodine-scan diagnostic (ISD) developed by PSI.¹¹ The ISD is a diode laser based monitor for the small signal gain in iodine lasers. The system uses a single mode, tunable diode laser that is capable of accessing all six hyperfine components of the atomic iodine. It was calibrated in frequency to enable automated operation for the (3,4) hyperfine transition for our experiments. A fiber optic cable was used to deliver the diode laser probe beam to the iodine diagnostic regions in the subsonic portion of the flow tube and in the supersonic cavity. Since the ISD uses a narrow band diode laser, measurements of the lineshapes can also be used to determine the local temperature from the Voigt profile.

The windows on the sides of the cavity, when using the gain diagnostic, were wedged and antireflection coated to minimize etalon effects. A single pass configuration (5 cm path length) was used in the supersonic diagnostic section. Measurements of the $O_2(^1\Delta)$ yield {defined as $Y = O_2(^1\Delta) / [O_2(^3\Sigma) + O_2(^1\Delta)]$ } were obtained from OMA-V calibrations involving gain measurements and the relative values of the spectral intensities measured for $I(^2P_{1/2})$ to $O_2(^1\Delta)$ using techniques originally developed by Hager,¹² Davis *et al.*,¹¹ and Rawlins *et al.*¹⁰

Laser power measurements were made with a Scientech Astral™ model AC5000 calorimeter interfaced to a Scientech Vector™ model S310 readout and were made at the same location in the supersonic laser cavity as were the gain measurements. The gain measurements were made first. The mirrors were then put in place for the laser power trials. One 99.990% reflective mirror purchased from Los Gatos Research and one 99.996% reflective mirror purchased from Advanced Thin Films, each with 2 m radius of curvature, formed a stable optical cavity. The mirrors were separated by approximately 34 cm. An infrared (IR) detection card from New Focus, model 5842, with response between 800 and 1600 nm, was also used to observe the intensity profile of the beam.

Electric discharge stability and temperature control were found to be critical parameters to obtain positive gain.^{2,3} Electric discharges sustained in moderate pressures (10 s of Torr) of oxygen are prone to arcing and constriction. The production of excited oxygen species by the discharge adds higher levels of complexity to the downstream kinetics when the iodine donor species are added to the flow. (These species are not usually encountered in the purely chemical system.) The critical aspect of temperature control results from the equilibrium of the singlet delta pumping reaction,



where the forward rate is $7.8 \times 10^{-11} \text{ cm}^3/\text{molecule s}$,¹³ and the backward rate is $1.04 \times 10^{-10} \exp(-403/T) \text{ cm}^3/\text{molecule s}$.¹⁴ The equilibrium rate constant ratio of the forward to backward reactions is $K_{eq} = 0.75 \exp(403/T)$,¹⁴ where T is the gas temperature. The yield of $O_2(^1\Delta)$ for optical transparency, Y_{OT} , as a function

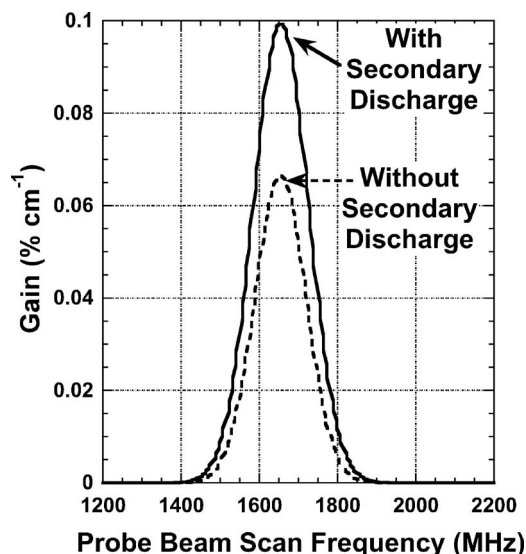


FIG. 2. Gain lineshape in the supersonic cavity as a function of probe beam scan frequency with and without a 100 W rf secondary I_2 dissociation discharge.

of temperature is $Y_{OT} = 1 / [1 + 1.5 \exp(403/T)]$.¹⁵ Note that the backward rate is slower, K_{eq} is larger, and Y_{OT} is lower as T is decreased.

The flow conditions for these gain and laser power experiments with the I_2 predissociator are 10.0 mmol/s of O_2 which is diluted with 50.0 mmol/s of He and 0.05 mmol/s of NO. The discharge production of $O_2(^1\Delta)$ was enhanced by the addition of a small proportion of NO to lower the ionization threshold of the gas mixture and improve discharge stability. The NO also significantly reduces the concentration of atomic oxygen which has been shown to quench the desired $I(^2P_{1/2})$ state.^{16,17} A secondary stream of ≈ 0.045 mmol/s of I_2 with 12.0 mmol/s of secondary He diluent was injected 27.3 cm downstream from the exit of the primary discharge and run through a 100 W rf secondary discharge. A tertiary flow of 100 mmol/s of cold N_2 gas (≈ 100 K) was injected further downstream to lower the temperature and to raise the pressure to improve the performance of the nozzle in our vacuum system. The pressures in the subsonic diagnostic duct and in the supersonic diagnostic cavity were 30.0 and 3.2 Torr, respectively. Measurements in the subsonic diagnostic duct from the $O_2(^1\Delta)$ and $O_2(b^1\Sigma)$ spectra indicated an $O_2(^1\Delta)$ yield of $\approx 13\%$ and a gas temperature of ≈ 450 K for these flow conditions at 700 W of rf power in the primary discharge.

Gain was measured for the above flow conditions at 700 W of primary rf discharge power and 100 W of secondary rf discharge power for the I_2 predissociator. Figure 2 shows the gain at line center which peaks at $0.10\% \text{ cm}^{-1}$ with the I_2 predissociator secondary discharge. For comparison, the best gain previously observed in our system⁹ of $0.067\% \text{ cm}^{-1}$ without this secondary discharge is also shown in Fig. 2; the secondary discharge provides a 50% $[(0.10 - 0.067)/0.067]$ enhancement in gain as compared to the previous best results. The lineshape indicates a temperature of ≈ 120 K. Note that the flow conditions are different between the two cases shown in Fig. 2 and represent, to date, the best gain flow conditions for cases with and without the secondary discharge. The flow conditions without the secondary discharge were $O_2:He:NO = 10:33:0.15$ mmol/s with P_{total}

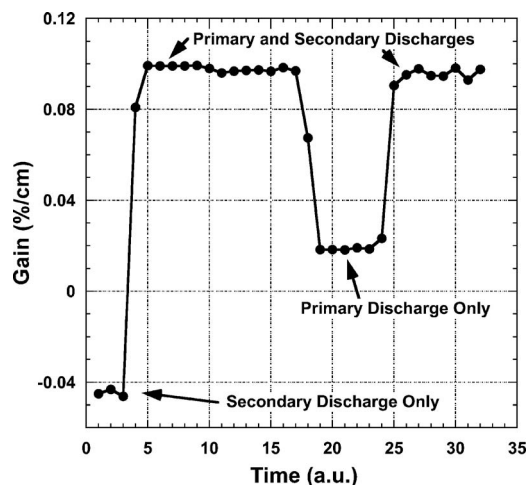


FIG. 3. Gain in the supersonic cavity as a function of time with different combinations of the primary and secondary discharges.

=20 Torr and a primary rf discharge power of 1000 W, and those with the secondary discharge were $O_2:He:NO = 10:50:0.05$ mmol/s with $P_{total} = 30$ Torr and a primary rf discharge power of 700 W (as stated above).

It is important to note that the primary discharge was intentionally reduced to 700 W and thus the total (primary plus secondary) rf discharge power was only 800 W when using the secondary discharge. This reduction in primary discharge power enabled the 50% enhancement to gain and a 20% reduction in total applied power as compared to the earlier 1000 W case from Ref. 9. Therefore, applying power to dissociate the iodine with the secondary discharge is more energy efficient than applying the power required to produce sufficient O atoms in the primary discharge to dissociate the iodine downstream.

Figure 3 shows the gain during a sequence of turning different discharges on and off, starting with the secondary discharge only, followed by both discharges on, followed by data with only the primary discharge, and then again with both discharges on. As anticipated, there is absorption when only the low power secondary discharge is running, showing that the molecular I_2 is at least partially dissociated. The authors hypothesize that some of the secondary discharge power may dissociate not only I_2 but also O_2 , followed by the newly created O atoms helping in the I_2 dissociation process. A test was run where the O_2 flow was set to zero, and the magnitude of the absorption decreased (but did not disappear). Therefore, we conclude that this phenomenon is occurring, but that the secondary discharge is at least partially dissociating the I_2 as desired; further studies of the secondary discharge dynamics and interactions with the primary flow are required. When the primary discharge is turned on, the gain rises quickly to $\approx 0.10\% \text{ cm}^{-1}$. When the secondary discharge is turned off, the gain drops to approximately $\approx 0.02\% \text{ cm}^{-1}$ (note that the gain for these flow conditions without the secondary discharge is significantly lower than the $0.067\% \text{ cm}^{-1}$ gain for the different 20 Torr, 1000 W flow conditions shown in Fig. 2), and when the secondary discharge is again turned on, the gain returns to $\approx 0.10\% \text{ cm}^{-1}$.

The laser resonator was subsequently installed around the supersonic cavity. For the above 30 Torr flow conditions and total rf power of 800 W, a total laser output power of

6.2 W was obtained, which was a 38% improvement to laser power relative to the results of Ref. 9. The beam was elliptically shaped with a length of ≈ 3.5 cm (the same as the clear aperture of the mirror mounts) in the flow direction and a height of ≈ 1.8 cm.

In conclusion, the authors observed a 50% enhancement in gain and 38% enhancement in continuous-wave laser power on the 1315 nm transition of atomic iodine through the addition of a secondary rf discharge to predissociate the molecular iodine at the injection location near the supersonic laser cavity in an electric oxygen-iodine laser. A gain of $0.10\% \text{ cm}^{-1}$ was obtained and the laser output power was 6.2 W in a stable cavity with one 99.990% reflective mirror and one 99.996% reflective mirror. Applying power to dissociate the iodine with the secondary discharge is more energy efficient than applying excess power in the primary discharge to produce sufficient O atoms for iodine dissociation downstream. Further improvements to the iodine dissociator are expected to provide improvements to the gain, laser power, and additional power savings over the current design. Note that the results presented herein represent approximately a 50-fold improvement in gain and laser power that has been achieved since the initial demonstrations in 2004–2005.^{2,3}

This work was supported by the Missile Defense Agency (MDA) through the U.S. Army Space and Missile Defense Command (USA SMDC). The authors gratefully thank B. Otey (USA SMDC); W. T. Rawlins and S. J. Davis (Physical Sciences Inc.); M. C. Heaven (Emory Univ.); J. Kotora and D. Podolski (MDA); and G. W. Sutton (Sparta Inc.). They would also like to thank D. M. King and J. K. Laystrom for their technical assistance.

¹W. McDermott, N. Pchelkin, D. Benard, and R. Bousek, *Appl. Phys. Lett.* **32**, 469 (1978).

²D. L. Carroll, J. T. Verdeyen, D. M. King, J. W. Zimmerman, J. K. Laystrom, B. S. Woodard, N. Richardson, K. Kittell, M. J. Kushner, and W. C. Solomon, *Appl. Phys. Lett.* **85**, 1320 (2004).

³D. L. Carroll, J. T. Verdeyen, D. M. King, J. W. Zimmerman, J. K. Laystrom, B. S. Woodard, G. F. Benavides, K. Kittell, D. S. Stafford, M. J. Kushner, and W. C. Solomon, *Appl. Phys. Lett.* **86**, 111104 (2005).

⁴W. T. Rawlins, S. Lee, W. J. Kessler, and S. J. Davis, *Appl. Phys. Lett.* **86**, 051105 (2005).

⁵J. T. Verdeyen, D. L. Carroll, D. M. King, J. K. Laystrom, G. F. Benavides, J. W. Zimmerman, B. S. Woodard, and W. C. Solomon, *Appl. Phys. Lett.* **89**, 101115 (2006).

⁶A. Hicks, S. Tirupathi, N. Jiang, Yu. Utkin, W. R. Lempert, J. W. Rich, and I. V. Adamovich, *J. Phys. D* **40**, 1408 (2007).

⁷A. A. Ionin, I. V. Kochetov, A. P. Napartovich, and N. N. Yuryshev, *J. Phys. D* **40**, R25 (2007).

⁸D. L. Carroll and W. C. Solomon, *Proc. SPIE* **4184**, 40 (2000).

⁹G. F. Benavides, A. D. Palla, D. M. King, D. L. Carroll, J. T. Verdeyen, J. K. Laystrom, T. H. Field, J. W. Zimmerman, B. S. Woodard, and W. C. Solomon, *AIAA Paper No. 2007-4623* (2007).

¹⁰W. T. Rawlins, S. J. Davis, S. Lee, M. L. Silva, W. J. Kessler, and L. G. Piper, *AIAA Paper No. 2003-4032* (2003).

¹¹S. J. Davis, M. G. Allen, W. J. Kessler, K. R. McManus, M. F. Miller, and P. A. Mulhall, *Proc. SPIE* **2702**, 195 (1996).

¹²G. D. Hager personal communication (25 October 2002).

¹³R. G. Derwent and B. A. Thrush, *Discuss. Faraday Soc.* **53**, 162 (1972).

¹⁴G. P. Perram and G. D. Hager (Air Force Weapons Laboratory, Kirtland Air Force Base, Final Report No. AFWL-TR-88-50 1988).

¹⁵J. Hon, G. Hager, C. Helms, and K. Truesdell, *AIAA J.* **34**, 1595 (1996).

¹⁶V. N. Azayazov, I. O. Antonov, S. Ruffner, and M. C. Heaven, *Proc. SPIE* **6101**, 61011Y (2006).

¹⁷D. L. Carroll, J. T. Verdeyen, D. M. King, J. W. Zimmerman, J. K. Laystrom, B. S. Woodard, G. F. Benavides, K. Kittell, and W. C. Solomon, *IEEE J. Quantum Electron.* **41**, 213 (2005).