

## Gain and continuous-wave laser oscillation on the 1315 nm atomic iodine transition pumped by an air-helium electric discharge

B. S. Woodard,<sup>1</sup> J. W. Zimmerman,<sup>1</sup> G. F. Benavides,<sup>1</sup> D. L. Carroll,<sup>1,a)</sup> J. T. Verdeyen,<sup>1</sup> A. D. Palla,<sup>1</sup> T. H. Field,<sup>1</sup> W. C. Solomon,<sup>2</sup> S. J. Davis,<sup>3</sup> W. T. Rawlins,<sup>3</sup> and S. Lee<sup>3</sup>

<sup>1</sup>CU Aerospace, 2100 S. Oak St., Suite 206, Champaign, Illinois 61820, USA

<sup>2</sup>306 Talbot Laboratory, University of Illinois, 104 S. Wright St., Urbana, Illinois 61801, USA

<sup>3</sup>Physical Sciences Inc., 20 New England Business Center, Andover, Massachusetts 01810, USA

(Received 13 May 2008; accepted 23 June 2008; published online 14 July 2008)

Herein the authors report on the demonstration of gain and a continuous-wave laser on the 1315 nm transition of atomic iodine using the energy transferred to  $I(^2P_{1/2})$  from  $O_2(a^1\Delta)$  produced by a radio-frequency-excited electric discharge sustained in a dry air-He-NO gas mixture. Active oxygen and nitrogen species were observed downstream of the discharge region. Downstream of the discharge, cold gas injection was employed to raise the gas density and lower the temperature of the continuous gas flow. Gain of  $0.0062\% \text{ cm}^{-1}$  was obtained and the laser output power was 32 mW in a supersonic flow cavity. © 2008 American Institute of Physics. [DOI: 10.1063/1.2957678]

The classical chemical oxygen iodine laser (COIL) reported by McDermott *et al.*<sup>1</sup> 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 demonstrated by Carroll *et al.*<sup>2,3</sup> in a supersonic flow cavity. Subsequent efforts have demonstrated gain<sup>4-6</sup> and lasing<sup>5,6</sup> in other ElectricOIL configurations since the earlier demonstrations.<sup>2,3</sup> For an excellent and comprehensive topical review of discharge production of  $O_2(^1\Delta)$  and ElectricOIL studies, see Ionin *et al.*<sup>7</sup> The demonstrations of ElectricOIL systems raise the possibility of other potential gas mixtures that can be electrically excited and transfer their energy to the iodine atom for subsequent lasing. One option is the use of air because the operational logistics are potentially easier than with  $O_2$ .

In this letter, we report on the demonstration of gain and a continuous-wave (cw) laser on the  $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$  electronic transition of the iodine atom at 1315 nm pumped by resonance excitation transfer from  $O_2(^1\Delta)$  produced in an electric discharge sustained in a dry air-He-NO gas mixture. There are considerably more excited states produced in this discharge gas mixture because the  $N_2$  contained in air will be excited into many vibrational and electronic levels. A block diagram of the flow tube setup is shown in Fig. 1. A radio frequency (rf) electric discharge at 13.56 MHz operating between two transverse electrodes was used as the excitation source. 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 Zimmerman *et al.*<sup>8</sup>

The supersonic diagnostic cavity has a Mach 2 nozzle with windows that serve as view ports. The subsonic diagnostic duct has four windows through which simultaneous measurements are made of the optical emission from  $O_2(^1\Delta)$  at 1268 nm,  $I(^2P_{1/2})$  at 1315 nm, and the gain/absorption proportional to  $[I(^2P_{1/2})] - 0.5 \cdot [I(^2P_{3/2})]$ . 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 and 1315 nm.

Micromotion CMF and Omega FMA mass flow meters were used to measure the flow rates of the gases. The  $I_2$  concentration was measured by a method developed by Physical Sciences Inc. (PSI) and is based on the continuum absorption of molecular iodine at 488 nm. Details of this diagnostic are described by Rawlins *et al.*<sup>9</sup> 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.<sup>10</sup> 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 line

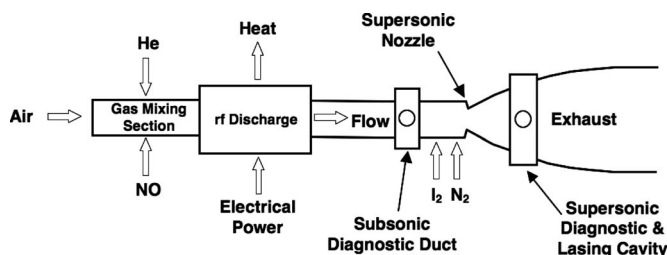


FIG. 1. Schematic of the experimental apparatus.

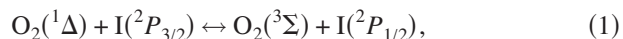
<sup>a)</sup> Author to whom correspondence should be addressed. Electronic mail: carroll@cuaerospace.com.

shapes 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  $Y = O_2(^1\Delta)/[O_2(^3\Sigma) + O_2(^1\Delta) + O_2(^1\Sigma) + 1.5 O_3 + 2O]$ ) were obtained from OMA-V calibrations to more complex 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<sup>11</sup> and Davis *et al.*<sup>10</sup> As noted above there are many more excited species in this gas discharge with air in the mixture due to the presence of  $N_2$ . It is important to note that there is a strong  $N_2(B)$  line around 1220 nm that grows significantly in strength as the discharge power is increased. At very high powers this  $N_2(B)$  spectrum overwhelms the  $O_2(^1\Delta)$  spectrum thereby making it progressively more difficult to analyze the  $O_2(^1\Delta)$  yield data.

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 placed in mounts for the laser power trials. Two mirrors with 2 m radius of curvature, purchased from Advanced Thin Films (ATF), formed a stable optical cavity. Measurements of transmission,  $T$ , indicated a transmission of  $0.003\% \pm 0.001\%$ . Direct measurements of mirror reflectivity,  $R$ , and absorption/scattering,  $AS$ , were unavailable, but previous reflectivity measurements of similar mirrors indicated that a fraction of the remainder of  $1-T$  is in  $AS$  losses for similar high reflectivity mirrors. Thus, we estimate that the mirrors each had a reflectivity of  $99.996\% \pm 0.001\%$ . The mirrors were separated by approximately 34 cm. An infrared 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 in obtaining positive gain. Electric discharges sustained in moderate pressures (5–15 Torr) of oxygen are prone to arcing and constriction. The production of excited oxygen and nitrogen 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  $7.8 \times 10^{-11} \text{ cm}^3/\text{molecule s}$ ,<sup>12</sup> and the backward rate is  $1.04 \times 10^{-10} \exp(-403/T) \text{ cm}^3/\text{molecule}$ .<sup>13</sup> The equilibrium rate constant ratio of the forward to backward reactions is  $K_{eq} = 0.75 \exp(403/T)$ ,<sup>13</sup> where  $T$  is the gas temperature. The yield of  $O_2(^1\Delta)$  for optical transparency as a function of temperature is  $Y_{OT} = 1/[1 + 1.5 \exp(403/T)]$ .<sup>14</sup> Note that the backward rate is slower,  $K_{eq}$  is larger, and  $Y_{OT}$  is lower as  $T$  decreases.

Several flow conditions were found that resulted in positive gain using the configuration shown in Fig. 1. A typical

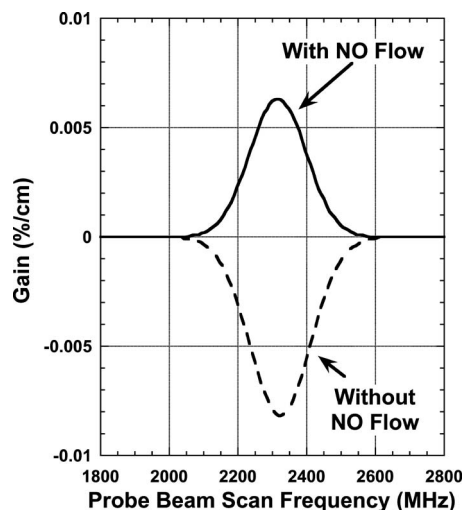


FIG. 2. Gain line shape in the supersonic cavity as a function of probe beam scan frequency with and without NO in the discharge gas mixture. Discharge power was 1500 W.

set of conditions are 3.0 mmol/s of  $O_2$  mixed with 12.0 mmol/s of  $N_2$  to create approximately 15.0 mmol/s of dry air which is diluted with 75.0 mmol/s of He and 0.1 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. The NO also significantly reduces the concentration of atomic oxygen which has been shown to quench the desired  $I(^2P_{1/2})$  state.<sup>15,16</sup> A secondary stream of  $\approx 0.040$  mmol/s of  $I_2$  with 12.0 mmol/s of secondary He diluent was injected  $\approx 27.3$  cm downstream from the exit of the discharge. A tertiary flow of 155 mmol/s of cold  $N_2$  gas ( $\approx 120$  K) was injected further downstream to lower the temperature and to raise the pressure to improve the performance of the nozzle within our vacuum system. The pressures in the subsonic diagnostic duct and in the supersonic diagnostic cavity were 44.0 and 2.0 Torr, respectively.

$O_2(^1\Delta)$  yield measurements were made as a function of rf discharge power and were found to be linear with rf discharge power to as high as 1500 W with the transverse discharge. At a pressure of 45 Torr and 1500 W of rf power, the yield was measured to be  $\approx 9\%$  with a gas temperature of  $\approx 400$  K, whereas at a pressure of 23 Torr and 800 W of rf power, the yield was measured to be  $\approx 16\%$  with a gas temperature of  $\approx 540$  K. It is interesting to note that we typically measure gas temperatures that are significantly higher (as much as 50–100 K) with  $O_2$ -He gas discharges than with these air-He discharge gas mixtures; this is another clear indication that large amounts of power are being absorbed and retained by excited  $N_2$  states, i.e., the energy is not being thermalized as heat into the flow.

BLAZE-IV (Ref. 17) simulations were performed for these flow conditions and yields and indicated that this magnitude of yield should be sufficient for gain and lasing in our experimental configuration. In order to simulate the performance of the  $O_2$ - $N_2$  discharge component of an electric nitrogen-oxygen-iodine laser device 17 species and 119 reactions were added to the BLAZE-IV model. These computations also indicated lower flow temperatures with  $N_2$  in the discharge gas mixture (not shown for brevity).

Gain was then measured for the above flow conditions at 1750 W of rf discharge power and is shown in Fig. 2 and

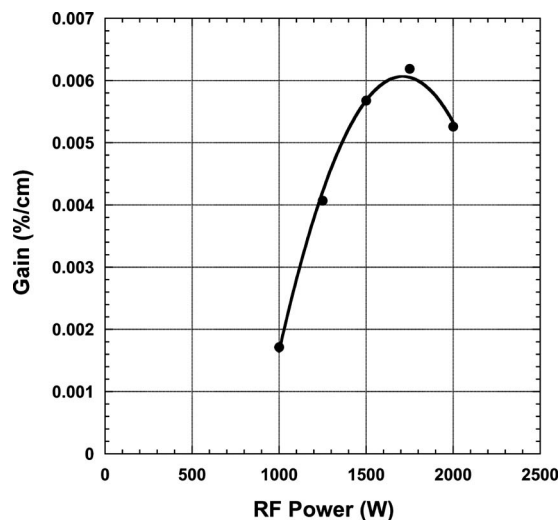


FIG. 3. Gain in the supersonic cavity as a function of rf discharge power.

peaks at  $0.0062\% \text{ cm}^{-1}$  at line center. The line shape indicates a temperature of  $\approx 180 \text{ K}$ . Figure 2 also shows the important effect of NO for this laser system where there is optical gain with NO in the discharge gas mixture, but there is optical absorption when NO is removed from the mixture. Figure 3 shows the gain as a function of rf power. Interestingly as the rf power is reduced below approximately 900 W only optical transparency is observed rather than absorption. We believe this phenomenon is due to a lack of molecular iodine dissociation (and hence little or no atomic iodine) at this point due to O atoms being reduced by both the presence of NO through a cyclic recombination<sup>8</sup> and through three-body recombination of O atoms at the higher pressure of 45 Torr for these cases. This hypothesis is supported by Fig. 2 which shows absorption when the NO was removed from the discharge gas mixture.

The laser resonator was subsequently installed around the supersonic cavity. For the above flow conditions and 1500 W rf power, a laser output power of 32 mW was obtained. The beam shape was a rounded rectangle with a length of  $\approx 1.9 \text{ cm}$  (the same as the clear aperture of the mirror mounts) and a height of  $\approx 1.1 \text{ cm}$ . For reference, the initial measurement of laser action using an ElectricOIL with a supersonic laser cavity at  $\approx 1.3 \text{ Torr}$  produced 220 mW.<sup>3</sup>

In conclusion, gain and cw laser action were measured on the  $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$  electronic transition of the iodine atom at 1315 nm pumped by a near resonant energy transfer from  $\text{O}_2(^1\Delta)$  produced in an electric discharge sustained in a dry air-He-NO gas mixture. There are a large number of excited  $\text{N}_2$  states produced in this discharge gas mixture in addition to  $\text{O}_2(^1\Delta)$ ; it is likely that some of these states can

influence the production/destruction of  $\text{I}^*$ , but further investigation is required to determine if these states play a net positive or negative role. Cold  $\text{N}_2$  was injected to lower the temperature of the flow and shift the equilibrium of atomic iodine in favor of the  $I(^2P_{1/2})$  state. This in combination with a supersonic flow cavity produced sufficient population inversion to observe gain of  $0.0062\% \text{ cm}^{-1}$  followed by laser oscillations when two  $\approx 99.996\%$  reflectivity mirrors were used to form an optical resonator surrounding the gain medium. The laser output power for these experimental conditions was 32 mW. Potential operational logistics make the use of air rather than  $\text{O}_2$  an interesting option, but the trade offs in performance need to be further assessed.

This work was supported by DARPA Contract No. HR0011-07-C-0054 through a subcontract from PSI. The authors gratefully thank D. M. King, J. K. Laystrom, and A. Roberts for their technical assistance.

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

<sup>2</sup>D. 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).

<sup>3</sup>D. 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).

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

<sup>5</sup>J. 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).

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

<sup>7</sup>A. A. Ionin, I. V. Kochetov, A. P. Napartovich, and N. N. Yuryshv, *J. Phys. D: Appl. Phys.* **40**, R25 (2007).

<sup>8</sup>J. W. Zimmerman, D. King, A. Palla, J. Verdeyen, D. Carroll, J. Laystrom, G. Benavides, B. Woodard, W. Solomon, W. Rawlins, S. Davis, and M. Heaven, *Proc. SPIE* **6261**, 62611R1 (2006).

<sup>9</sup>W. T. Rawlins, S. J. Davis, S. Lee, M. L. Silva, W. J. Kessler, and L. G. Piper, "Optical diagnostics and kinetics of discharge-initiated oxygen-iodine energy transfer," AIAA Paper No. 2003-4032, 2003.

<sup>10</sup>S. J. Davis, M. G. Allen, W. J. Kessler, K. R. McManus, M. F. Miller, and P. A. Mulhall, *Proc. SPIE* **2702**, 195 (1996).

<sup>11</sup>G. D. Hager, personal communication (25 October 2002).

<sup>12</sup>R. G. Derwent and B. A. Thrush, *Faraday Discuss. Chem. Soc.* **53**, 162 (1972).

<sup>13</sup>G. P. Perram and G. D. Hager, Final Report No. AFWL-TR-88-50, 1988.

<sup>14</sup>J. Hon, G. Hager, C. Helms, and K. Truesdell, *AIAA J.* **34**, 1595 (1996).

<sup>15</sup>V. N. Azayazov, I. O. Antonov, S. Ruffner, and M. C. Heaven, *Proc. SPIE* **6101**, 61011Y (2006).

<sup>16</sup>D. 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).

<sup>17</sup>A. D. Palla, J. W. Zimmerman, B. S. Woodard, D. L. Carroll, J. T. Verdeyen, T. C. Lim, and W. C. Solomon, *J. Phys. Chem. A* **111**, 6713 (2007).