

Gain recovery in an electric oxygen-iodine laser

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(Received 1 October 2008; accepted 13 December 2008; published online 14 January 2009)

Recent investigations of an electric oxygen-iodine laser system have shown that computational modeling overpredicts the experimentally measured power output for similar gain conditions. This discrepancy is potentially due to an unknown reaction that competes with the forward pumping of $I(^2P_{1/2})$ by $O_2(a^1\Delta)$. Measurements of gain recovery downstream of an operating laser cavity were performed. Modeling of this experiment shows that reducing the forward pumping rate by an effective factor of approximately 4 to simulate a competing mechanism results in the computational modeling matching the experimental gain recovery measurements, and in improved agreement between the measured and modeled laser power extraction. © 2009 American Institute of Physics. [DOI: 10.1063/1.3064163]

The electrically driven oxygen-iodine laser (ElectricOIL) that was first demonstrated by Carroll *et al.*^{1,2} 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 $O_2(^1\Delta)$]. Subsequent efforts have demonstrated gain^{3–5} and lasing^{4–6} in other ElectricOIL configurations since the first demonstrations. Ionin *et al.*⁷ provided a comprehensive topical review of discharge production of $O_2(^1\Delta)$ and various ElectricOIL studies.

In this letter the authors report on gain recovery measurements downstream of an operating resonator in an electric oxygen-iodine laser device. The $O_2(^1\Delta)$ is produced by a transverse capacitive 13.56 MHz discharge in O_2 –He–NO mixture. The quartz discharge tube has a 4.9 cm inside diameter, and the electrodes are 25.4 cm long. More information on the performance of the transverse rf discharge sustained in O_2 –He–NO gas mixtures used in the experiments presented herein can be found in Woodard *et al.*⁸ The $O_2(^1\Delta)$ -rich flow (primary) exiting the discharge region passes through a water-cooled heat exchanger and then passes through a duct containing injectors for iodine vapor carried by helium (secondary) and cooled nitrogen (tertiary). The combined flow is then expanded through a Mach 2 nozzle, which has purged mounts on either side of the supersonic flow section that accept 2 in. (50 mm) optics into which wedged windows can be placed for gain measurements, or laser mirrors can be placed for laser oscillation. For this particular set of experiments, a special insert was manufactured in order to hold a 1 in. (25 mm) high-reflectivity laser mirror at each mount centerline inside of the wedged windows, allowing a gain probe beam to pass upstream and downstream of the active resonator. The diagnostic setup for this experiment is illustrated in Fig. 1. The gain length of the laser cavity is 5 cm. Two 99.996% reflective mirrors purchased from AT Films, each with 2 m radius of curvature and

1 in. diameter formed a stable optical cavity. The mirror separation was approximately 33 cm.

Simultaneous measurements of the optical emission from $O_2(^1\Delta)$ at 1268 nm and $O_2(b^1\Sigma)$ at 762 nm were made in the subsonic section downstream of the discharge. A Roper scientific optical multichannel analyzer with a 1024-element InGaAs LN_2 cooled array interfaced to an Acton Research SP-2300i spectrometer was used for measurements at 1268 nm. A thermoelectric-cooled Apogee charge coupled device was used to measure the spectra of the $O_2(b^1\Sigma)$ transition at 762 nm. Gas flow temperatures can be extracted from the rotational spectra of the $O_2(b^1\Sigma)$ measurements. Both instruments were 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 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.*⁹ Pressure in the subsonic and supersonic flow regions was measured by capacitance manometers from MKS and Leybold.

Measurements of gain (or absorption) were made using the Iodine-Scan Diagnostic (ISD) developed by PSI.¹⁰ 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 compo-

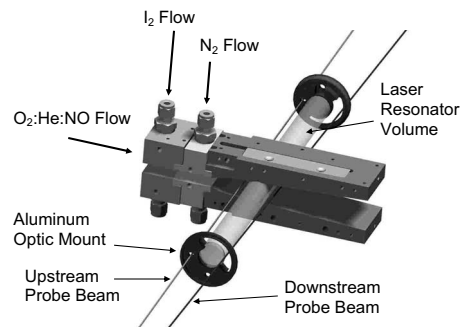


FIG. 1. Illustration of the experimental apparatus.

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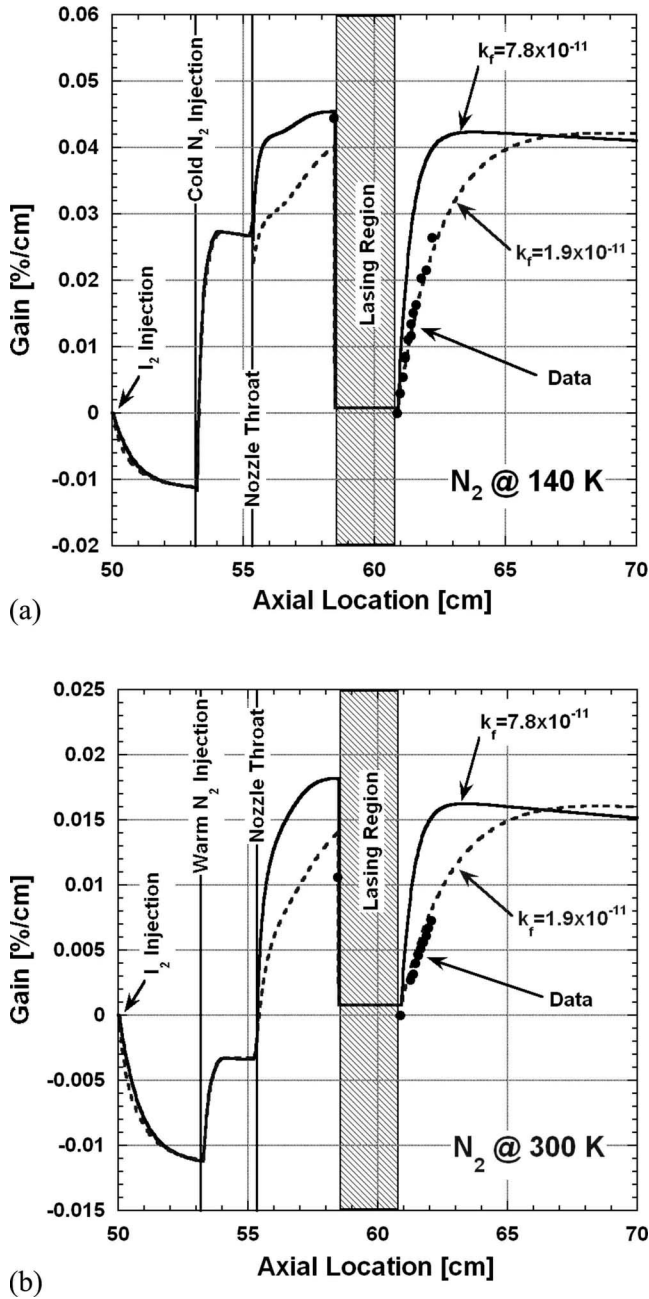


FIG. 2. Comparison of gain data with BLAZE-V predictions as a function of axial location and $I+O_2(^1\Delta)\leftrightarrow I^*+O_2(^3X)$ forward pumping rate for a lower temperature flow with (a) 74 mmol/s of tertiary N_2 injected at 140 K and (b) 50 mmol/s of tertiary N_2 injected at 300 K.

nents 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 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. Laser power measurements were made with a Scientech Astral™ model AC5000 calorimeter interfaced to a Scientech Vector™ model S310 readout. An infrared detection card from New Focus, Model 5842, with response between 800–1600 nm, was also used to observe the intensity profile of the beam.

The primary flow conditions for the gain recovery measurements were 7 mmol/s O_2 diluted in 33 mmol/s He and

0.05 mmol/s NO. The power input to the discharge was 700 W. 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.^{11,12} The $O_2(^1\Delta)$ yield measured downstream of the discharge for these conditions was $\approx 18\%$, which corresponds to 170 W stored in the $O_2(^1\Delta)$ flow. A secondary stream of ≈ 0.02 mmol/s of I_2 with 20.0 mmol/s of secondary He diluent was injected 32.4 cm downstream from the exit of the primary discharge. Gain recovery measurements were taken for two tertiary N_2 flow inputs: (a) 74 mmol/s N_2 at 140 K and (b) 50 mmol/s N_2 at 300 K. These flow conditions result in laser cavity temperatures of approximately 115 and 164 K for cases (a) and (b), respectively. In case (a) the discharge operated at 24 Torr. In case (b) the discharge operated at 20 Torr.

The spatial gain measurements and corresponding BLAZE-V modeling results are shown in Fig. 2. The modeling results are shown for two values of the forward pumping rate of the reaction $O_2(^1\Delta)+I\leftrightarrow O_2+I^*$. The accepted forward rate for classic chemical oxygen-iodine laser pumping is $k_f=7.8\times 10^{-11}$ molecules $^{-1}$ cm 3 s $^{-1}$ (see van Marter and Heaven¹³), while the modified effective rate which results in improved agreement with the gain measurements in both cases is $k_{f,eff}=1.9\times 10^{-11}$ molecules $^{-1}$ cm 3 s $^{-1}$. In both modeling cases, the backward rate is determined from the well-established equilibrium rate,¹³ $K_{eq}=k_b/k_f=1.3346e^{(-403/T)}$. This four times reduction in forward rate supports a prior hypothesis by Rawlins *et al.*¹⁴ that there is some important unknown kinetic process that is specific to the ElectricOIL system and tied to the pumping process. The result that the same effective rate constant provided good agreement with both sets of data at different temperatures suggests that this ElectricOIL-specific kinetic process is relatively temperature independent.

To reinforce the conclusion from the above comparison of experiment and the BLAZE-V model, the data were compared to a simple analytic model. The spatial derivative of the gain can be expressed as

$$\frac{dg}{dx} = \frac{d}{dx} \left\{ \sigma(T) \left([I^*] - \frac{1}{2}[I] \right) \right\} = \sigma(T) \left\{ \frac{d[I^*]}{dx} - \frac{1}{2} \frac{d[I]}{dx} \right\}, \quad (1)$$

where $\sigma(T)=(7/12)(1.293\times 10^{-17})\sqrt{300/T}$ is the stimulated emission cross section (in cm 2), and the spatial derivatives of $[I^*]$ and $[I]$ can be expressed as

$$\begin{aligned} \frac{d[I^*]}{dx} &= \frac{1}{u} \frac{d[I^*]}{dt} = \frac{1}{u} (k_f [O_2(^1\Delta)][I] - k_b [O_2][I^*] - k_q [O] \\ &\quad \times [I^*]), \quad \frac{d[I]}{dx} = -\frac{d[I^*]}{dx}, \end{aligned} \quad (2)$$

where the backward rate $k_b=k_f/K_{eq}$, k_q is the rate of I^* quenching by oxygen atoms and u is the flow velocity. The influence of oxygen atom quenching is negligible in these cases, and the slope at a given point in the data can be approximated as linear, such that $dg/dx=C$ is constant. Using these approximations, and the above relations, the effective forward rate can be expressed as

$$k_{f,\text{eff}} = \frac{2uC}{3\sigma(T) \left\{ [\text{O}_2(^1\Delta)][\text{I}] - \frac{1}{K_{\text{cq}}} [\text{O}_2][\text{I}^*] \right\}}. \quad (3)$$

Using this simple model, and the measured values of slope from the gain data in Fig. 2, and other inputs to Eq. (3), the values of $k_{f,\text{eff}}$ determined in the (a) chilled and (b) room-temperature N_2 tertiary cases were 1.64×10^{-11} and 1.45×10^{-11} molecules $^{-1}$ cm 3 s $^{-1}$, respectively. Thus, this simple analytical model is in agreement with the assessment from BLAZE-V calculations that there is an unknown mechanism that competes with the forward pumping of I^* by $\text{O}_2(^1\Delta)$.

The measured laser powers were 1.8 and 0.85 W for data in Figs. 2(a) and 2(b), respectively. The predicted laser powers using a forward rate of $k_f = 7.8 \times 10^{-11}$ molecules $^{-1}$ cm 3 s $^{-1}$ were 9.2 and 5.2 W for Figs. 2(a) and 2(b), respectively. When the forward rate was reduced to $k_{f,\text{eff}} = 1.9 \times 10^{-11}$ molecules $^{-1}$ cm 3 s $^{-1}$, the predicted laser powers were reduced to 3.6 and 1.7 W for Figs. 2(a) and 2(b), respectively. Compared to the experiment, the BLAZE-V model predicts a similar ratio of power outputs between the cold and room-temperature tertiary flow conditions as that observed in the experiment (the power output in the room-temperature N_2 case is approximately 50% of the chilled N_2 case), but the predicted powers are still higher than measured, even when the reduced pumping rate that agrees with the gain recovery measurement is used. Optical losses could also be playing a role in reducing the extracted power.

The behavior seen in Fig. 2 appears to be a significant finding for ElectricOIL system development. Although the data sets do not show full recovery of the gain downstream of the resonator due to the limiting aperture of the experimental setup, the computations that match the measured slopes do demonstrate full gain recovery further downstream. Thus, the desired result of full gain recovery is achieved, but the comparison of experiment and modeling demonstrates that this occurs at a slower rate than anticipated. This suggests that whatever the responsible kinetic mechanism is, it acts in a “competing” fashion to the standard pumping reaction, as opposed to a “quenching” reaction that irreversibly removes available energy from the $\text{O}_2(^1\Delta)$ and/or I^* . This is a very important distinction between competing and quenching because a competing kinetic process implies that the energy is still available, but that it will take longer to extract. This result suggests that it may be possible to extract significantly more laser power in an ElectricOIL system by (i) minimizing the effects of the competing process or (ii) the use of a longer resonator in the flow direction (or possibly a folded resonator concept).

One of the significant remaining questions regarding the ElectricOIL system is: Why is the amount of laser power extracted so much lower than the available power in the $\text{O}_2(^1\Delta)$? Measurements of gain recovery rate reported here indicate that there is an ElectricOIL-specific kinetic process that is competing with the classic oxygen-iodine laser pump-

ing reaction. We are not suggesting that the established forward pumping rate is in error, only that there is an additional competing process that is occurring in the ElectricOIL system kinetics due to additional species not present in classic chemical oxygen-iodine lasers. BLAZE-V power calculations with a slower pumping rate constant (that simulates the competing process) can also explain a significant difference between the measured power and what is available to be extracted from the $\text{O}_2(^1\Delta)$. At this time there are still too many uncertainties to draw definitive conclusions; however it appears likely that the difference in ElectricOIL measured laser power and the extractable power available from the $\text{O}_2(^1\Delta)$ is largely due to a competing kinetic reaction with the pumping reaction, and not some error in the established pumping rate. Some future work should be guided toward defining the mechanisms responsible for the reduced gain recovery rate determined here.

This work was supported by the Missile Defense Agency (MDA) through the U.S. Army Space and Missile Defense Command/U.S. Army Forces Strategic Command (USA SMDC). The authors gratefully thank B. Otey (USA SMDC), W. T. Rawlins and S. J. Davis (PSI), M. C. Heaven (Emory Univ.), I. Adamovich, W. Rich, and W. Lempert (Ohio St. Univ.), J. Kotora and D. Podolski (MDA), and G. W. Sutton (Sparta Inc.). They would also like to thank D. M. King, J. K. Laystrom, T. H. Field, and B. Shea for their technical assistance.

¹D. 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).

²D. 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).

³W. T. Rawlins, S. Lee, W. J. Kessler, and S. J. Davis, *Appl. Phys. Lett.* **86**, 051105 (2005).

⁴J. 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).

⁵A. Hicks, S. Tirupathi, N. Jiang, Yu. Utkin, W. R. Lempert, J. W. Rich, and I. V. Adamovich, *J. Phys. D* **40**, 1408 (2007).

⁶S. J. Davis, S. Lee, D. B. Oakes, J. Haney, J. C. Magill, D. A. Paulsen, P. Cataldi, K. L. Galbally-Kinney, D. Vu, J. Polex, W. J. Kessler, and W. T. Rawlins, *Proc. SPIE* **6874**, 68740D (2008).

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

⁸B. S. Woodard, J. W. Zimmerman, J. T. Verdeyen, D. L. Carroll, T. H. Field, G. F. Benavides, A. D. Palla, and W. C. Solomon, High Power Laser Ablation Conference, Taos, NM, 21–24 April 2008 (unpublished).

⁹W. T. Rawlins, S. J. Davis, S. Lee, M. L. Silva, W. J. Kessler, and L. G. Piper, AIAA-Paper-2003-4032 (2003).

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

¹¹V. N. Azyazov, I. O. Antonov, S. Ruffner, and M. C. Heaven, *Proc. SPIE* **6101**, 61011Y (2006).

¹²D. 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).

¹³T. van Marter and M. C. Heaven, *J. Chem. Phys.* **109**, 9266 (1998).

¹⁴W. T. Rawlins, S. Lee, W. J. Kessler, L. G. Piper, and S. J. Davis, AIAA-Paper-2005-5299 (2005).