

# Spatial and recovery measurements of gain in an electric oxygen-iodine laser

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## ABSTRACT

Recent investigations of an Electric Oxygen-Iodine Laser system have shown that computational modeling over-predicts the laser power output measured in experiments for similar gain conditions. To help resolve this discrepancy, detailed 2-axis mapping of gain and gain recovery measurements downstream of an operating laser cavity were performed. Modeling and analyses of the gain recovery experiments indicate that when the pumping rate of  $I(^2P_{1/2})$  by  $O_2(a^1\Delta)$  is reduced by an effective factor of approximately 4 as a result of an unknown competing reaction, the calculations are well matched to the experimental gain recovery measurements. The agreement between the measured and modeled laser power extraction also significantly improves when the reduced effective pumping rate is used. The results suggest that there may be a competing reaction that effectively reduces the forward pumping rate as compared to the classical chemical oxygen-iodine laser kinetics rates. Understanding of this kinetic process should enable us to accommodate or eliminate its impact on ElectricOIL performance.

## 1. INTRODUCTION

The electrically driven oxygen-iodine laser (ElectricOIL) that was first demonstrated by Carroll *et al.* [Carroll, 2004a; Carroll, 2005] 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  $O_2(a)$ ]. Since the first reporting of a viable electric discharge-driven oxygen-iodine laser system (also often referred to as EOIL or DOIL in the literature), there have been a number of other successful demonstrations of gain [Rawlins, 2005a; Verdeyen, 2006; Hicks, 2007] and laser power [Verdeyen, 2006; Hicks, 2007], as well as a recent demonstration of gain and lasing from an air-helium discharge [Woodard, 2008a]. Computational modeling of the discharge and post-discharge kinetics [Stafford, 2005; Palla, 2006; Palla, 2007] has been a useful tool in ElectricOIL development, allowing analysis of the production of various discharge species [ $O_2(a^1\Delta)$ ,  $O_2(b^1\Sigma)$ , O atoms, and  $O_3$ ] and determination of the influence of  $NO_x$  species on system kinetics. Ionin *et al.* [Ionin, 2007] provide a comprehensive topical review of discharge production of  $O_2(a)$  and various ElectricOIL studies. The highest gain in an ElectricOIL device reported to date is  $0.17\% \text{ cm}^{-1}$ , with an output power of 12.3 W [Zimmerman, 2008].

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  $O_2(a)$ ? For example, for the highest reported power case mentioned above, we have a laser output power of 12.3 W for a flow that has approximately 210 W of power carried downstream by  $O_2(a)$ . While the BLAZE model [Palla, 2006; Palla, 2007] has demonstrated good agreement with the experimental measurements for production of various discharge species and laser gain, Fabry-Perot laser simulations with the BLAZE model indicate that for this case that we should be able to extract over 160 W in laser power, yet we are only getting a little over 12 W. Three possible causes seem the most likely suspects: (i) potential strong non-uniformities in the gain due to thermal boundary layers in the nozzle, (ii) an unknown chemical kinetic process that is somehow inhibiting (slowing) the power extraction process, and (iii) optical losses that dissipate power before it is outcoupled (this latter possibility is discussed by Carroll *et al.* [Carroll, 2008]). In this paper, we will focus on the first two possibilities through measurements of spatial gain in a supersonic ElectricOIL system.

## 2. DIAGNOSTICS SUITE AND DISCHARGE SETUP

A Princeton Instruments/Acton Optical Multi-channel Analyzer (OMA-V, 1024-element InGaAs array) with a 0.3-m spectrometer and a 600 g/mm grating blazed at 1.2  $\mu\text{m}$  was used for measurements at 1268 nm. An Apogee E47

CCD camera coupled to a Roper Scientific/Acton Research 0.15-m spectrometer was used to measure the emission of  $O_2(b^1\Sigma)$  at 762 nm (from which flow temperature was determined using the method described by Carroll *et al.* [Carroll, 2003]). These optical diagnostics were fiber coupled using either Oriol model #77538 glass fiber bundles or ThorLabs 600  $\mu\text{m}$  x 5 m multimode fibers. Micro-Motion CMF and Omega FMA mass flow meters were used to measure the flow rates of the gases. Pressures in the flow tubes were measured with MKS Instruments and Leybold capacitance manometers. The incident and reflected powers to the radio-frequency (RF) matching network were measured by a Bird ThruLine model 43 wattmeter (RF power presented herein is the difference of the incident and reflected powers). Matching the power to the discharge was achieved using a traditional PI-matching network with a tapped air-core transformer.

Measurements of gain (or absorption) were made using the fiber-coupled Iodine-Scan Diagnostic (ISD) developed by Physical Sciences Inc. (PSI) [Davis, 1996]. Since the ISD uses a narrow band diode laser, measurements of the lineshapes are also 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.

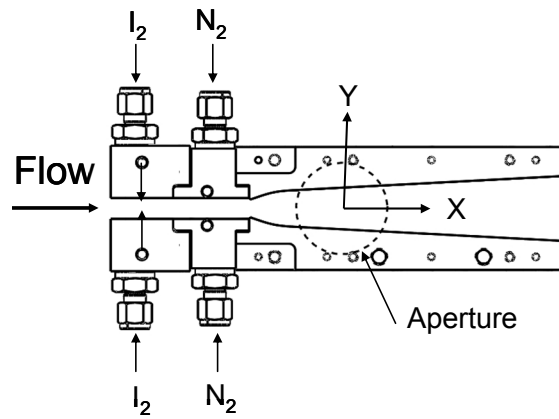
The  $O_2(a)$  for the experiment was produced by a transverse capacitive 13.56 MHz discharge in an  $O_2$ -He-NO mixture. The quartz discharge tube has a 4.9-cm i.d., and the copper foil electrodes are 25.4 cm long. The discharge flow conditions were 7 mmol/s  $O_2$  diluted in 33 mmol/s He and  $\approx 0.1$  mmol/s NO with a discharge pressure of approximately 20 to 24 Torr (depending on tertiary flow conditions) and RF input of 700 W. These flow rates and pressures result in an  $O_2(a)$  yield of  $\approx 18\%$ . The yield varied during the experiments by no more than the statistical error of the measurement, and was therefore approximately constant throughout the experiments. 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.* [Woodard, 2008b].

### 3. SPATIAL GAIN MEASUREMENTS

As discussed above, the BLAZE model provides good agreement with the measured laser gain, but over-predicted the extracted power by as much as an order of magnitude. One potential explanation for this discrepancy between the model-predicted laser power and the experimental measurement was attributed to the limited dimensionality of the model, which assumed flow uniformity across the nozzle height; the gain, which is strongly dependent on temperature, could be strongly influenced by a large thermal boundary layer which would result in a large temperature variation across the nozzle channel. Thus, experimental measurements to establish the vertical gain profile were performed.

Small signal gain measurements were recorded along two axes in the ElectricOIL 4<sup>th</sup> Generation Cavity (CAV4) supersonic nozzle. The aperture size is approximately 44.5 mm parallel to the flow (x-axis) and 20 mm perpendicular to the flow (y-axis). The gain probe was translated repeatedly along each axis and the gain was periodically recorded at the center of the aperture (labeled as the origin in Fig. 1) and compared with previous measurements to insure that minimal temporal drift occurred. In many cases, the system operated near continuously for two to three hours with minimal variation in performance.

**Figure 1. Schematic of CAV4 flow channel. The physical aperture illustrated is a result of the window/mirror mounts.**



### 3.1. Experimental setup

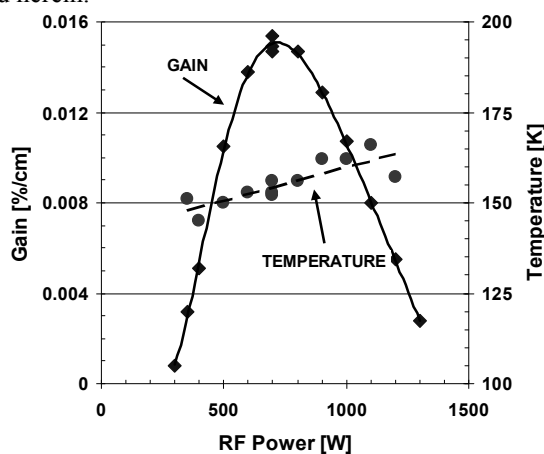
The discharge configuration and flow rates through the discharge are as described in Sect. 2. The  $O_2(a)$  and  $O_2(b)$  measurements are made just downstream of the discharge using a purged diagnostic block. Following the diagnostics, the flow passes into the CAV4 nozzle where a secondary flow of approximately 25  $\mu\text{mol/s}$  of iodine diluted in 20 mmol/s of helium is mixed into the primary flow. A tertiary flow of temperature regulated nitrogen is subsequently mixed into the flow. The tertiary flow provides two principle functions: (i) the main function is to cool the gases from the RF discharge, and (ii) the tertiary flow promotes mixing upstream of the nozzle throat, improving the distribution of iodine in the primary flow.

Two nitrogen tertiary flow conditions were studied. First, 40 mmol/s of nitrogen at room temperature was injected into the flow channel. When mixed with the primary flow and accelerated to approximately Mach 2.0 (geometric Mach 2.4 nozzle), the resulting flow temperature is about 160 K. Second, the flow rate of nitrogen was increased to 72 mmol/s and the nitrogen temperature chilled to approximately 95 K. The resulting flow temperature in the nozzle becomes approximately 110 K. Henceforth, the room temperature nitrogen and chilled nitrogen tertiary cases shall be referred to as the warm flow and cold flow cases, respectively.

### 3.2 Spatial Gain Data

The aperture available to the gain diagnostic and the axes used while plotting the measurements is illustrated in Fig. 1. Gain was measured 50 mm downstream from the nozzle throat as a function of RF system power for the room temperature case described above, Fig. 2. A power of 700 W was clearly optimal for the described conditions, and thus 700 W was utilized for all of the experiments described herein.

Figure 2. Gain and supersonic flow temperature vs RF power in the laser cavity. Flow conditions (in mmol/s): 7  $O_2$ , 34 He, 0.06 NO, 0.025  $I_2$  carried by 20 secondary He, 40 tertiary  $N_2$  at 300 K,  $P_{\text{total}} \sim 20$  Torr.



The results from mapping gain along the x-axis, as defined in Fig. 1, are provided as Figs. 3a and 3b, for the warm and cold flow cases, respectively. The position  $x=0.0$  in Fig. 3 references a point 50 mm downstream from the nozzle throat. Figures 4a and 4b illustrate the measured gain and temperature distribution perpendicular to the flow across the nozzle (at the  $x=0.0$  position) for the warm and cold flow cases, respectively. A number of points can be made about the data:

1. The gain improves significantly in the warm flow case (Fig. 3a) as the flow proceeds through the nozzle and the temperature drops. This illustrates the strong sensitivity of gain to temperature changes when not pre-cooled. Temperature and gain essentially mirror one another.
2. Figure 3b shows the expected rise in gain when NO is added to the discharge gas mixture; this has been observed previously [Carroll, 2004a; Rawlins, 2005a]. It is interesting to note that the addition of NO only raises the  $O_2(a)$  yield from 15.4% to 17.4%, yet there is a significant improvement of  $\sim 60\%$  in the gain accompanying the addition of NO. This is because of the changing concentrations of additional species in the flow, namely oxygen atoms. The O atoms both deactivate  $O_2(a)$  and quench  $I^*$  and since NO reduces the presence of O atoms, the gain improves, regardless of the fact that removing the oxygen atoms resulted in some flow heating (discussed below). Figure 3b illustrates the importance of managing undesirable species in the flow and furthermore shows the relatively flat gain in the flow direction of the resonator.

- The addition of NO raises the temperature of the flow, Fig. 3b, due to the additional chemical reactions that effectively convert O atoms into O<sub>2</sub>. Despite the additional heating of approximately 10 K, the gain improves significantly when NO is added. Since the flow is so much colder in the cold flow case, the gain is less sensitive to the additional heat in this case. In other words, the additional heating due to chemical reactions with NO is not a significant issue with an appropriate amount of pre-cooling.
- For the cold flow cases where the temperature of the flow (< 120 K) results in a very low yield required for optical transparency,  $Y_{OT}$  (discussed below in Sect. 4), the gain moving downstream in the nozzle stays relatively uniform. For these cases, even when the temperature continues to drop through the nozzle, the gain remains fairly steady because  $Y_{OT}$  is already less than ~2%.
- Figures 4a and 4b show, in both the warm and cold flow cases, that the gain distributions perpendicular to the flow are mostly flat across much of the resonator volume, but go to zero near the walls due to the thermal boundary layer. Figure 4b better illustrates the rise in temperature near the walls because of a stronger gain signal. The gain signal in the warm flow case is weaker and turns sharply towards zero near the walls such that getting reliable temperature measurements from the gain profile became difficult. The uniformity helps show that good mixing is achieved with the tertiary flow injection just prior to the nozzle.

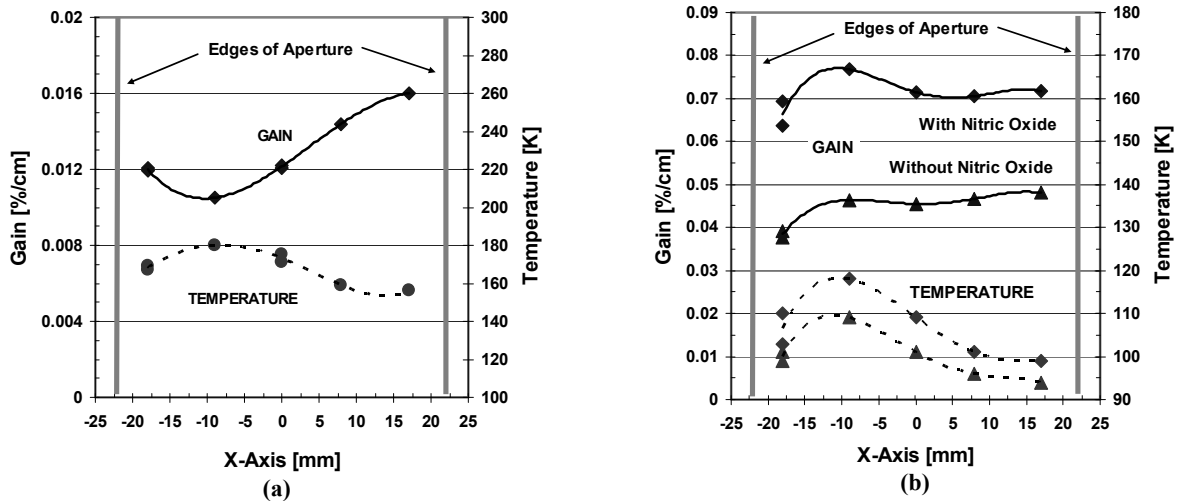


Figure 3. Gain and temperature map in the direction of the supersonic flow. X=0 is center of aperture. Flow conditions were: 7 O<sub>2</sub>, 34 He, 0.06 NO, 700 W, with (a) 40 N<sub>2</sub> at 300 K,  $P_{total} \sim 20$  Torr, and (b) 73 N<sub>2</sub> at 90 K,  $P_{total} \sim 24$  Torr.

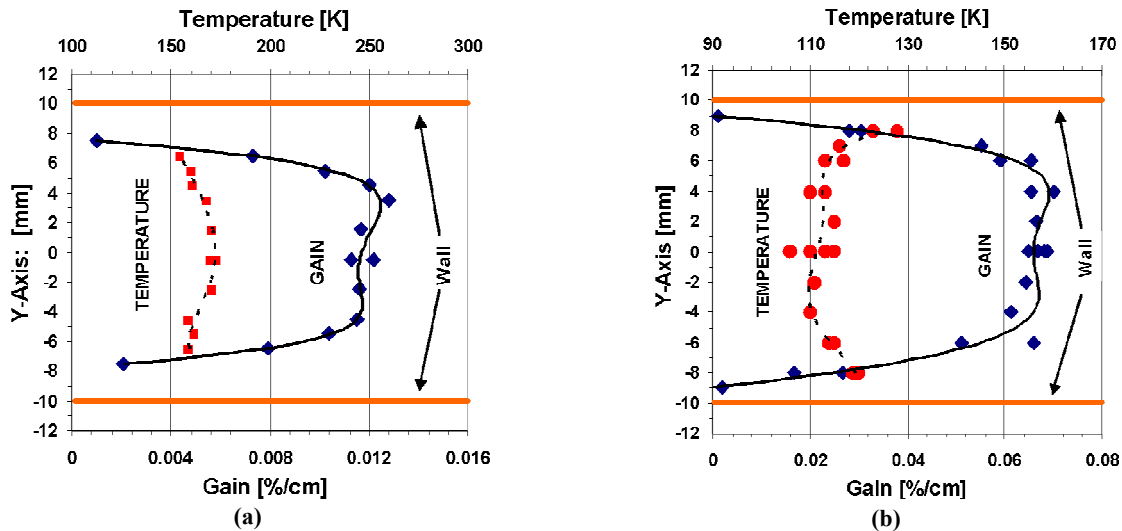


Figure 4. Gain and temperature map perpendicular to the supersonic flow. Y=0 is centerline of flow channel. Flow conditions were: 7 O<sub>2</sub>, 34 He, 0.06 NO, 700 W, with (a) 40 N<sub>2</sub> at 300 K,  $P_{total} \sim 20$  Torr, and (b) 73 N<sub>2</sub> at 90 K,  $P_{total} \sim 24$  Torr.

### 3.3. Discussion

Spatial measurements of the gain within the laser cavity show a gain region that is fairly uniform along the flow axis, and a vertical profile that falls to zero at the flow channel walls due to the influence of the thermal and viscous boundary layer. However, the thermal boundary layer fills only a limited portion of the nozzle and is only a factor in approximately 25% of the nozzle. These 2-axis measurements are qualitatively consistent with those taken for the VertiCOIL nozzle [Keating, 1997] (from which the CAV4 nozzle was derived) and also with those taken for an Israeli COIL configuration [Rybalkin, 2002]. Given the relative uniformity of the gain profiles and qualitative similarity to COIL data that had no extraordinary power extraction issues, we do not believe that flow non-uniformities are a major contributor to the discrepancy between measured and predicted ElectricOIL laser powers.

### 4. GAIN RECOVERY DOWNSTREAM OF LASER RESONATOR

In an effort to understand if there is an unknown chemical kinetic process that is limiting the power extraction we performed measurements of the gain immediately downstream of the lasing region. Proper interpretation of the gain recovery measurements appears to be tied closely to the pumping reaction that produces the lasing species I\*,



In practice, the backward rate  $k_b$  for this reaction is measured, and the equilibrium constant, which is determined from statistical thermodynamics, is used to determine the forward rate  $k_f$ . The equilibrium constant  $K_{eq}$  is given by

$$K_{eq} = \frac{k_f}{k_b} = 0.75 \exp\left(\frac{401.4}{T}\right). \quad (2)$$

The rate of the backward reaction  $k_b$  is well established at room temperature [Derwent, 1971] ( $k_b = 2.7 \times 10^{-11}$  molecules<sup>-1</sup> cm<sup>3</sup> s<sup>-1</sup>), and its temperature dependence was confirmed by Van Marter and Heaven [Van Marter, 1998] to be in good agreement with a simple Arrhenius model which matched the room temperature result and measurements at 150 K. The established forward and backward pumping rates are

$$k_f = 7.8 \times 10^{-11} \text{ molecules}^{-1} \text{ cm}^3 \text{ s}^{-1} \quad (3)$$

and

$$k_b = 1.04 \times 10^{-10} \exp(-401.4/T) \text{ molecules}^{-1} \text{ cm}^3 \text{ s}^{-1} \quad (4)$$

Knowledge of these rates is vital to the modeling and experimental development of oxygen-iodine laser technology. Equation (2) can be used to determine the temperature and O<sub>2</sub>(a) yield levels required to produce conditions for an operational oxygen-iodine laser cavity. Positive gain on the I\*→I transition requires [I\*]/[I] > 0.5, and therefore  $K_{eq}[\text{O}_2(\text{a})]/[\text{O}_2(\text{X})] > 0.5$  [Hon, 1996]. The fractional yield required for optical transparency is then expressed as

$$Y_{OT}(T) = \frac{1}{1 + 2K_{eq}(T)} = \frac{1}{1 + 1.5 \exp\left(\frac{401.4}{T}\right)}, \quad (5)$$

where fractional yield is defined by  $Y = [\text{O}_2(\text{a})] / \{[\text{O}_2(\text{a})] + [\text{O}_2(\text{X})]\}$ .

The equilibrium relation, Eqn. (2), can also be used along with measurements of iodine flow rate, iodine dissociation fraction, and gain (or absorption) measurements to determine the O<sub>2</sub>(a) yield. This O<sub>2</sub>(a) yield measurement can then be used to calibrate the 1268 nm emission to the density of O<sub>2</sub>(a). This method of calibrating the O<sub>2</sub>(a) emission was first introduced by Rawlins *et al.* [Rawlins, 2003] and is used in our ElectricOIL research [Carroll, 2004b] to analyze the yield performance of various discharge configurations. This method is also useful in determining the O<sub>2</sub>(a) yield at locations within the nozzle laser cavity (downstream of the iodine injection) where using the 1268 nm emission is difficult due to low density, low partial pressure of O<sub>2</sub>(a), and varying geometry.

#### 4.1. Experimental Setup

For this particular set of experiments, two identical dual optic mounts were designed and fabricated that each hold a 1" laser mirror on the internal vacuum side in addition to a 2" window on the external side. Gain measurements can be made simultaneously through openings in the holder both upstream and downstream of the lasing region. The downstream aperture was large enough to permit translation of the gain probe over a 1.2 cm distance. The diagnostic setup for this experiment is illustrated in Fig. 5. 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" diameter formed a stable optical cavity. The mirror separation was 33 cm.

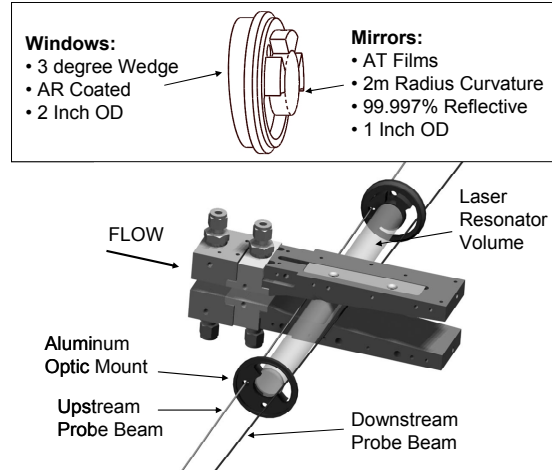


Figure 5. Experimental setup for gain recovery measurements.

#### 4.2. Data

Figures 6a and 6b show data for the two sets of flow conditions having different injection temperatures of the tertiary  $N_2$  gas to determine if there are any substantial changes in the kinetics as a function of gas temperature. Corresponding BLAZE-V simulations were performed for these cases. For both temperatures, the BLAZE-V calculations with a standard  $I + O_2(a) \leftrightarrow I^* + O_2(X)$  pumping rate (the backward rate is fully determined by the equilibrium constant and the forward rate) of  $k_f = 7.8 \times 10^{-11}$  molecules $^{-1}$  cm $^3$  s $^{-1}$  were found to show a much steeper rise in gain as a function of distance than exhibited by the data.

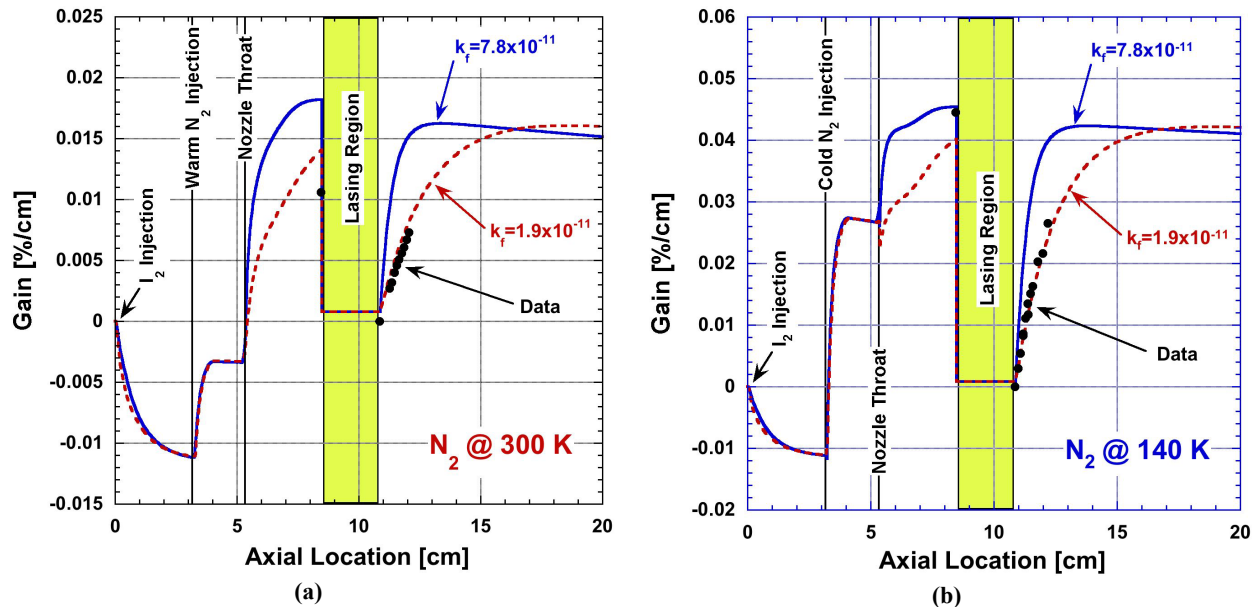


Figure 6. Comparison of gain data with BLAZE-V predictions as a function of axial location and  $I + O_2(^1\Delta) \leftrightarrow I^* + O_2(^3X)$  forward pumping rate for a low temperature flow with (a) 50 mmol/s of tertiary  $N_2$  injected at 300 K and (b) 74 mmol/s of tertiary  $N_2$  injected at 140 K.

A number of subsequent BLAZE-V computations were performed to simulate the equivalent of a competing process to the pumping reaction, and a forward rate of  $k_f = 1.9 \times 10^{-11}$  molecules $^{-1}$  cm $^3$  s $^{-1}$  was found to give the best agreement with both sets of data, Figs. 6a and 6b; approximately a factor of 4x slower than anticipated. Since the standard COIL pumping rate is well established [Van Marter, 1998], this finding supports a prior hypothesis by Rawlins *et al.* [Rawlins, 2005] that there is some important unknown kinetic process that is specific to the ElectricOIL system and tied to the pumping process. Note that the simulations predict that the gain with the slower

forward rate of  $k_f=1.9 \times 10^{-11}$  is higher downstream of the lasing region than upstream of the lasing region. This is a consequence of the fact that for this much slower pumping rate that the  $I^*/I$  ratio does not reach equilibrium before the laser cavity, but when given more time downstream of the resonator it is able to do so; therefore the gain ends up higher downstream of the resonator with the slower rate. The result that the same rate constant provided good agreement with both sets of data at different temperatures suggests that whatever this ElectricOIL-specific kinetic process is, it is relatively temperature independent.

Table 1 shows that the measured laser powers were 0.85 W and 1.8 W for data in Figs. 6a and 6b, respectively. The predicted laser powers using a forward rate of  $k_f=7.8 \times 10^{-11}$  molecules<sup>-1</sup> cm<sup>3</sup> sec<sup>-1</sup> were 5.2 W and 9.2 W for Figs. 6a and 6b, respectively. When the forward rate was reduced to  $k_f=1.9 \times 10^{-11}$  molecules<sup>-1</sup> cm<sup>3</sup> sec<sup>-1</sup> the predicted laser powers were reduced to 1.7 W and 3.6 W for Figs. 6a and 6b, respectively. Compared to the experiment, the BLAZE-V model predicts a similar ratio of power outputs between the room-temperature and cold tertiary flow conditions as that observed in the experiment (the power output in the room-temperature N<sub>2</sub> case is approximately 50% of the chilled N<sub>2</sub> case), but the predicted powers are still somewhat higher than measured, even when the reduced pumping rate which agrees with the gain recovery measurement is used. It is possible that the non-uniform gain profile due to thermal boundary layer (gain approaches zero at the walls) could be responsible for this smaller discrepancy. However, it is more likely that it is optical losses that are also playing a role in reducing the extracted power [Carroll, 2008].

**Table 1: Measured laser power and predicted (BLAZE-V) laser power using two different pumping rates for two different temperature conditions.**

	Experimental Power Data (W)	Laser Power (W) BLAZE-V $k_f=7.8 \times 10^{-11}$	Laser Power (W) BLAZE-V $k_f=1.9 \times 10^{-11}$
N <sub>2</sub> @ 300 K	0.85	5.2	1.7
N <sub>2</sub> @ 140 K	1.8	9.2	3.6

### 4.3. Analysis and discussion

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

$$\frac{dg}{dx} = \frac{d}{dx} \left\{ \sigma(T) \left( [I^*] - \frac{1}{2} [I] \right) \right\} = \sigma(T) \left\{ \frac{d[I^*]}{dx} - \frac{1}{2} \frac{d[I]}{dx} \right\}, \quad (6)$$

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

$$\frac{d[I^*]}{dx} = \frac{1}{u} \frac{d[I^*]}{dt} = \frac{1}{u} \left( k_f [O_2(^1\Delta)] [I] - k_b [O_2] [I^*] - k_q [O] [I^*] \right) \quad \text{and} \quad \frac{d[I]}{dx} = -\frac{d[I^*]}{dx}, \quad (7)$$

where the backward rate  $k_b = k_r / K_{eq}$ ,  $k_q$  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 (because the O atoms have been reduced to very small concentrations in the supersonic cavity through three-body recombination and consumption in the iodine dissociation process), 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

$$k_{f,eff} = \frac{2uC}{3\sigma(T) \left\{ [O_2(a)] [I] - \frac{1}{K_{eq}} [O_2] [I^*] \right\}}. \quad (8)$$

Using this simple model, and measured values of slope from the gain data in Fig. 6, and other inputs to Eqn. (8) from Table 2, the values of  $k_{f,eff}$  determined in the (a) chilled and (b) room-temperature N<sub>2</sub> tertiary cases are  $1.64 \times 10^{-11}$  molecules<sup>-1</sup> cm<sup>3</sup> sec<sup>-1</sup> and  $1.45 \times 10^{-11}$  molecules<sup>-1</sup> cm<sup>3</sup> sec<sup>-1</sup>, respectively. Table 2 lists the various estimated flow

conditions and specie concentrations that were used for these estimates of the effective forward rate. Thus, this simple analytical model is in reasonable agreement with the assessment from BLAZE-V calculations that there is a competing mechanism which effectively reduces the forward pumping of I\* by O<sub>2</sub>(a).

**Table 2. Measured and predicted conditions from BLAZE-V model for cold and warm N<sub>2</sub> injection cases.**

Conditions at Axial Position 61.5 cm	Cold N <sub>2</sub> Case (T <sub>inj</sub> ≈ 140 K)	Warm N <sub>2</sub> Case (T <sub>inj</sub> ≈ 300 K)
Temperature (K)	115.3	162.7
$K_{eq}$	24.4	8.84
Velocity, $u$ (cm/s)	57,641	73,540
Pressure (Torr)	2.50	2.28
Pressure, Total (Torr)	20	24
$C = dg/dx$ (cm <sup>-2</sup> )	1.897x10 <sup>-4</sup>	5.527x10 <sup>-5</sup>
[O <sub>2</sub> ] (molecules/cm <sup>3</sup> )	9.303x10 <sup>15</sup>	7.335x10 <sup>15</sup>
[O <sub>2</sub> (a)] (molecules/cm <sup>3</sup> )	1.554x10 <sup>15</sup>	1.254x10 <sup>15</sup>
[I] (molecules/cm <sup>3</sup> )	3.009x10 <sup>13</sup>	2.658x10 <sup>13</sup>
[I*] (molecules/cm <sup>3</sup> )	2.671x10 <sup>13</sup>	1.828x10 <sup>13</sup>
[O] (molecules/cm <sup>3</sup> )	4.635x10 <sup>12</sup>	3.678x10 <sup>12</sup>
$k_{f, effective}$ (cm <sup>3</sup> /molecules-s)	<b>1.64x10<sup>-11</sup></b>	<b>1.45x10<sup>-11</sup></b>

A more rigorous analytical approach can be taken by writing out the rate equations for the species, dropping certain terms as negligible, and assuming homogeneous and particular solutions for the change in iodine concentrations as a function of distance from the end of the optical resonator. For brevity we will only provide the significant results of this more detailed analysis. The gain can be expressed as a function of flow distance  $x$  to be

$$g(x)_{dr} = \frac{\sigma([I] + [I^*])}{2} \left\{ \frac{\left( \frac{Y_{dr}}{Y_{OT}} \right) - 1}{(K_{eq} - 1)Y_{dr} + 1} \right\} \left( 1 - \exp\left(-\frac{x}{L}\right) \right), \quad (9)$$

where  $L$  is the characteristic gain recovery distance

$$L = \frac{u}{(k_f[O_2(a)]_{dr} + k_b[O_2]_{dr})} \quad (10)$$

and the notation “dr” represents flow conditions just downstream of the resonator after some of the O<sub>2</sub>(a), or yield, has been used in the lasing process. Eqn. (9) provides an analytic expression for how fast the gain should recover with distance, and in the limit of  $x \rightarrow \infty$  the value of gain estimated by Eqn. (9) is within 10% of that predicted by the detailed kinetic BLAZE-V model, Fig. 6. This analysis results in a modified expression for the effective forward rate as:

$$k_{f, eff} = \frac{2K_{eq}uC}{3\sigma(T)([I] + [I^*])([O_2] + [O_2(a)]) \left\{ \frac{Y}{Y_{OT}} - 1 \right\}}. \quad (11)$$

Using Eqn. (11) one calculates values of the effective forward rate to be within 2% of those calculated using Eqn. (8). Note that Eqn. (11) reduces to Eqn. (8) with the use of some reasonable approximations, therefore it is not surprising that the two relations give very similar results.

The behavior seen in Fig. 6 appears to be a significant finding for ElectricOIL system development. In both data sets the gain recovers to the expected level (a desired result), but 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 O<sub>2</sub>(<sup>1</sup>Δ) 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 this competing process, or (ii) the use of a longer resonator in the flow direction (or possibly a folded resonator concept).

## 5. CONCLUSION AND FINAL REMARKS

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  $O_2(a)$ ? In this paper, two factors that could help answer this question were investigated. First, spatial measurements of the gain within the resonator volume along the flow axis and nozzle height axis were made, showing a gain profile which is uniform along the flow axis, but influenced by a thermal boundary layer at the nozzle walls. However, this thermal boundary layer is typical for COIL flows and we therefore do not believe that it is playing a significant role in the ability to extract power from an ElectricOIL system. These 2-axis measurements were taken for two flow input conditions resulting in two cavity centerline temperatures of  $\sim 160$  K and  $\sim 110$  K. Second, measurements of gain recovery rate reported here indicate that there is an ElectricOIL-specific kinetic process that is competing with the classic COIL pumping reaction. We are not suggesting that the usual forward pumping rate is in error, even though BLAZE-V simulations show that when the forward rate is reduced by a factor of  $\sim 4$  to fit the data, the modeled gain recovery rate agrees well with the measured recovery rate at both flow temperatures, and the predicted laser output powers are reduced, achieving better agreement with the experimental results. 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  $O_2(a)$  is largely due to a competing kinetic reaction with the pumping reaction and not some error in the pumping rate. Some future work should be guided towards defining the mechanisms responsible for the reduced gain recovery rate determined here. Also, the capability of modeling the 2D effects of thermal boundary layer on the gain profile, and their influence on laser power output would be of interest to ElectricOIL development.

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