

Hybrid ElectricOIL Discharge, Gain, and Power Enhancements

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Recent experiments have led to improvements in the hybrid Electric Oxygen-Iodine Laser (ElectricOIL) system that significantly increased the discharge performance, supersonic cavity gain, and laser power output. The continuous wave laser operating on the $I(^2P_{1/2}) \rightarrow I(^2P_{3/2})$ transition of atomic iodine at 1315 nm was pumped by the production of $O_2(a^1\Delta)$ in a radio-frequency discharge in an $O_2/He/NO$ gas mixture. Molecular iodine was injected into the active oxygen flow from the electric discharge and expanded through a supersonic cavity to lower the gas mixture temperature and shift the atomic iodine equilibrium in favor of the $I(^2P_{1/2})$ state. Greater than 30% of the discharge power is now being coupled into the active oxygen gas flow, a gain of $0.067\% \text{ cm}^{-1}$, and a laser power of 4.5 Watts were measured. Modeling with the BLAZE-IV model is in good agreement with data.

I. Introduction

The classical COIL first reported by McDermott [McDermott, 1978] 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 $O_2(a^1\Delta)$ [also denoted hereafter as $O_2(a)$]. Conventionally, a chemical two-phase process is used to produce the $O_2(a)$ at the interface of liquid basic H_2O_2 and Cl_2 gas. Zaleskii [Zaleskii, 1975] and Fournier [Fournier, 1980] made early attempts to use electric discharges for $O_2(a)$ production and transfer energy to iodine for lasing, but did not obtain positive gain. Since then, various groups [Carroll, 2003a; Carroll, 2003b; Schmiedberger, 2001; Hill, 2001; Ionin, 2007; Rakhimova, 2003] have investigated other continuous flowing systems and have measured $O_2(a)$ yields in excess of 15%, a necessary condition for positive gain at room temperature. Carroll et al. [Carroll, 2004] achieved positive gain and reported lasing in the same system in subsequent work [Carroll, 2005a]. One key difference between the traditional chemical excitation route and the electrical one is the presence of atomic oxygen levels on the same order as the $O_2(a)$. Atomic oxygen depletes the upper laser level [Azyazov, 2006a; Azyazov, 2006b], I^* , and must be controlled [Carroll, 2005b; Palla, 2006]. This was accomplished by the use of NO_2 titration made downstream from the discharge or by adding NO to the discharge flow or downstream of the discharge; all of these approaches resulted in the oxygen atoms being depleted to a level such that there were still enough oxygen atoms to dominate the I_2 dissociation process, but low enough such that the power loss through the $I^* + O$ quenching channel is not prohibitively detrimental. Subsequent efforts have demonstrated gain [Rawlins, 2005a; Verdeyen, 2006; Hicks, 2007] and lasing [Verdeyen, 2006; Hicks, 2007] in other ElectricOIL configurations since the first demonstrations [Carroll, 2004; Carroll, 2005a]. ElectricOIL discharge models [Ionin, 2007; Stafford, 2005; Babaeva, 2006; Rakhimova, 2006; Proshina, 2006; Palla, 2007] have been developed and post-discharge kinetics-gain-laser modeling [Carroll, 2003a; Palla, 2006; Palla, 2007] has been performed. For an excellent topical review of

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discharge production of $O_2(a)$ and ElectricOIL studies see Ionin *et al.* [Ionin, 2007]. In the work presented herein we discuss recent developments in ElectricOIL technology that have produced significant increases in discharge performance, supersonic cavity gain, and laser power.

II. Experiments

Use of the well-calibrated gas laser facility at the University of Illinois allowed advanced concepts to be economically implemented, compared directly against less sophisticated methods and examined in detail. This effort leveraged advanced discharge technologies and diagnostics developed over the past several years to produce an efficient intermediate demonstration of our small-scale ElectricOIL laser devices. The ability to implement these critical advanced technologies into the ElectricOIL laser testbed enables the community to understand the highly complex system questions involved and to advance the applicability of this technology.

2.1 Experimental Setup

CUA and UIUC's ElectricOIL research is carried out with a quartz discharge flow tube utilizing either internal hollow aluminum electrodes or external copper electrodes to couple in a radio frequency (RF) discharge to a primary flow comprising oxygen, an inert diluent (typically helium), and a sensitizer gas (typically nitric oxide, NO). Two supersonic laser cavities were used for this work. The second generation hardware that we will denote as "Cav2" was used in the original ElectricOIL gain and laser demonstrations [Carroll, 2004; Carroll, 2005a]. Third generation hardware, denoted as "Cav3" hereafter, was designed and fabricated to have considerably more flexibility than the Cav2 hardware. In the Cav2 hardware, the locations for iodine and cold tertiary gas injection were somewhat restricted, which has been significantly limiting both gain and laser power enhancements (as will be discussed below). A iodine-helium gas mixture was injected through an aluminum annular injector 63.5 cm downstream from the downstream electrode, followed by injection of cold (≈ 135 K) N_2 gas tertiary flow injected 12.7 cm downstream from the iodine injection plane with the Cav2 hardware. This gas mixture then entered the Cav2 supersonic laser cavity with a 5 cm gain length with the nozzle throat 24.1 cm downstream from the tertiary N_2 injector. In the Cav3 hardware configuration, the iodine and tertiary flows can be injected as in the Cav2 hardware, but can also be injected at 15 discrete positions ranging from 1.5 cm to 38.1 cm upstream from the nozzle throat position. In both the Cav2 and Cav3 configurations, the gas flow is expanded through a Mach 2.1 nozzle (based upon pressure ratio; the nozzle is Mach 2.4 based upon geometric area ratio) to lower the temperature of the gas to favor the iodine population inversion for gain and lasing.

Pressure measurements in each flow section are achieved via capacitance manometers. Flow rates are monitored by a variety of thermal and coriolis effect mass flow meters. An LN_2 -cooled OMA-V InGaAs array detector is used to measure spectra of the $O_2(a \rightarrow X)$ transition at 1268 nm. An electro-thermally-cooled Apogee CCD is used to measure spectra of the $O_2(b \rightarrow X)$ transition about 761.9 nm. Both instruments are fiber-coupled to allow positioning flexibility and repeatability. Most data is taken in a diagnostic duct fitted with purged window ports, but some data is taken directly through the flow tube walls. (Field of view corrections were made in these instances to maintain comparable calibrated yields.) A Physical Sciences Inc. (PSI) IodineScan diagnostic [Rawlins, 2005b] was used for the gain measurements. Scientech Astral model AC5000 calorimeters with Scientech Vector S310 readouts were used to measure the laser power. The beam profile was monitored in a qualitative way via IR detection cards from New Focus (model 5842).

2.2 Transverse Discharge Experiments

In prior work by the CUA-UIUC team, we typically utilized either a longitudinal or an inductive 13.56 MHz RF discharge in our ElectricOIL experiments, but this type of discharge has typically resulted in discharge instabilities that would occur at around 500 W in our small system. In an effort to improve the discharge stability we implemented a "dipolar excitation" (transverse) discharge concept [Carroll, 2006], Figs. 1 and 2. This concept places contoured capacitive electrodes along the length of the discharge section, similar to experiments being performed at Moscow State University [Rakhimova, 2006]. One significant advantage of this technique is that it provides inherent capacitive ballasting that inhibits current constriction and arcing, thus providing better discharge stability. Hicks *et al.* [Hicks, 2007] have also observed that the transverse discharge provides better stability at higher discharge pressures. Experiments performed in our laboratory with a transverse discharge resulted in significant yields and power coupled into the $O_2(a)$ state at pressures of 20 and 30 torr, Fig. 3; these yields were significantly higher (greater than a factor of 1.5 improvement) than we had previously been able to obtain with a longitudinal discharge. Also very importantly we were able to attain larger fractions of the power deposited into the flow going into the desired $O_2(a)$ state at higher pressures, Fig. 4; $>30\%$ at a pressure of 20 Torr and $>25\%$ at 30

torr. This is a significant improvement above the longitudinal discharge with which we were only getting about 18% of the electrical power into the desired $O_2(a)$ state. With the dipolar exciter discharge a gain of 0.027%/cm was obtained at 20 Torr, Fig. 5.

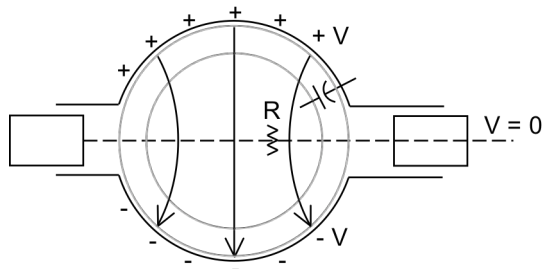


Figure 1. Cross sectional illustration of transverse dipolar discharge showing capacitive ballasting. Gas flow is into the paper.

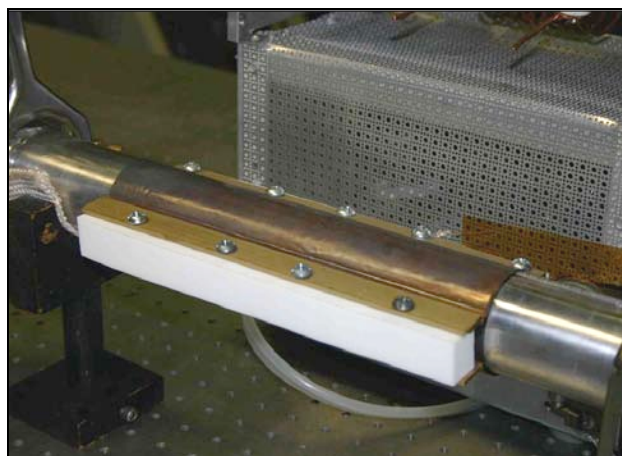


Figure 2. Assembled transverse dipolar discharge utilized in experiments.

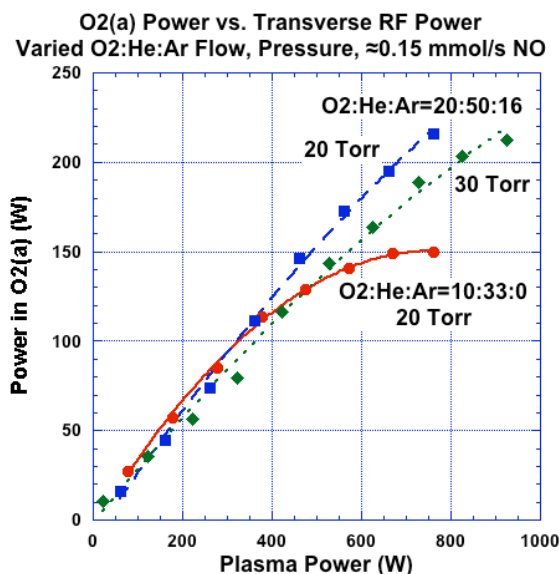


Figure 3. Yield of $O_2(a)$ vs. rf power absorbed by the plasma as a function of flow rate and pressure using an rf transverse discharge.

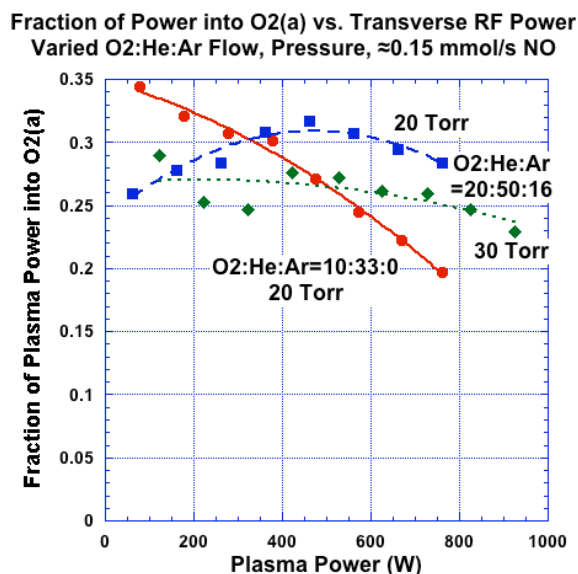


Figure 4. Fraction of plasma power deposited into $O_2(a)$ vs. rf power absorbed by the plasma as a function of flow rate and pressure using an rf transverse discharge.

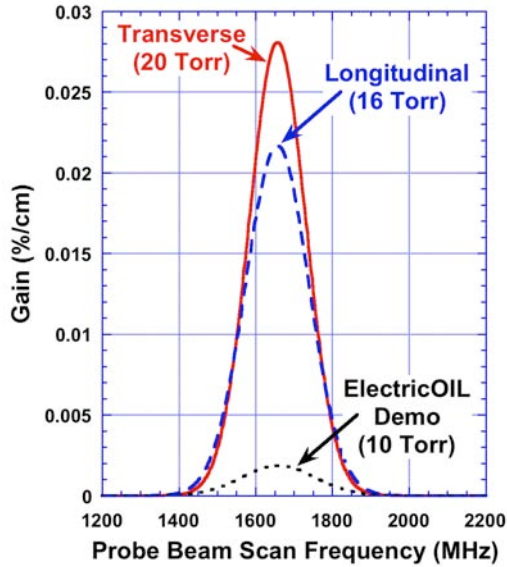


Figure 5. Digitally filtered gain signal in the supersonic cavity as a function of probe beam scan frequency for: (i) $O_2:He=4:10$ mmol/s with $P_{total} = 10$ Torr longitudinal discharge from the initial ElectricOIL demonstration [Carroll, 2004], (ii) $O_2:He=10:33$ mmol/s with $P_{total} = 16$ Torr longitudinal discharge [King, 2006], and (iii) $O_2:He=10:33$ mmol/s with $P_{total} = 20$ Torr transverse dipolar discharge from this work.

2.3 Variable Nozzle Mach Number

The ElectricOIL kinetics stretch the gain zone further downstream than occurs in a classic COIL system. This has the advantage of reducing the intensity loading on the laser mirrors; however, it also requires modifications to efficiently extract the energy optically and new scaling parameters need to be established. The nozzle geometry affects the gain zone and the temperature of the flow. The trade-off between shorter gain zone (mode width) and flow temperature was explored with new geometric Mach 2.67 and 3.0 nozzles and compared with the baseline Mach 2.4 nozzle, Fig. 6. A higher Mach number nozzle will provide lower temperature, but will stretch the gain zone by the increase in flow velocity.

Pressure and gain testing were performed with the baseline Mach 2.4 nozzle and the new Mach 2.67 and 3.0 nozzles using the Cav2 hardware. Comparison of the results indicates that the old Mach 2.4 nozzle gave higher gain, Fig. 7, than did the higher Mach number nozzles when the injected N_2 gas was cooled. We believe there are three primary reasons for this: 1) the Mach 2.4 nozzle has lower flow speed and higher density in the cavity, 2) since the gas is already being cooled from the cold gas injection, the higher Mach number nozzles have little temperature advantage over the Mach 2.4 nozzle, and 3) for total pressures between 10-16 Torr, the cavity pressures never fell below about 0.75 Torr regardless of nozzle, therefore we believe that our pumping system is not sufficient to properly pump the Mach 2.67 or Mach 3.0 nozzles at the lower total pressures. As we drive the total pressure to higher values the Mach 2.67 and Mach 3.0 nozzles will perform closer to their design point because it is a low pressure limitation that we are encountering, not a flow rate limitation. Our conclusion from this testing was that we should continue to use the Mach 2.4 nozzle for gain and laser testing with the Cav3 hardware, Section 2.4. However, based upon these results we recommend testing a Mach 1.5-2.0 nozzle in future work with our Cav2 or Cav3 hardware for comparison.

It is also interesting to note that Hicks *et al.* [Hicks, 2007] have found their best performance with a Mach 3.0 nozzle and that they recommend moving towards a Mach 4.0 nozzle. For their flow and discharge conditions, this may be necessary because of their very low yields (5-6%), but as yields are improved we do not recommend such a high Mach number nozzle. As was pointed out by Hicks *et al.*, and in agreement with theoretical analysis of COIL flows by Hager *et al.* [Hager, 1996], the residence time in the laser cavity decreases with flow velocity, and a significant competition occurs between residence time in the resonator and the time required to repopulate the upper laser level, and thus it becomes difficult to efficiently extract the flow energy optically. For our flow conditions with much higher yields, we find that a lower Mach number nozzle is preferable.



Figure 6. Photographs of the different nozzles tested to examine the effect of Mach number on the gain performance of the system: geometric Mach numbers of (a) 2.4, (b) 2.67, and (c) 3.0.

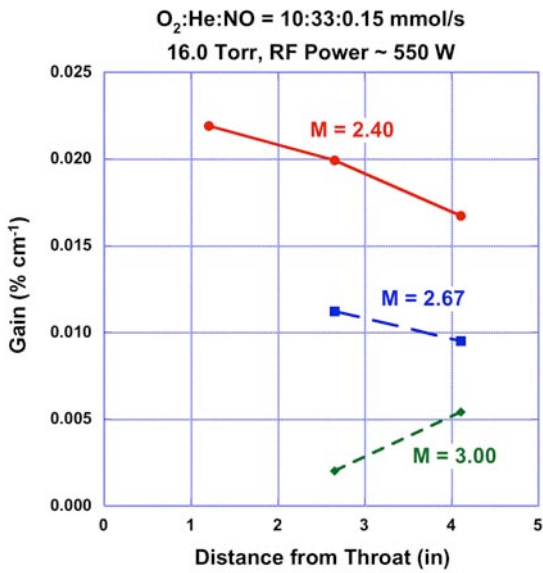


Figure 7. Gain vs. distance from the throat (inches) for three different nozzles having geometric Mach numbers of (a) 2.4, (b) 2.67, and (c) 3.0.

2.4 Power Enhancement of ElectricOIL to 5 W

Building upon results from the previous Sections 2.1-2.3, we implemented a logical progression of knowledge into a 3rd generation "Cav3" ElectricOIL system to increase the laser power output level to approximately 5 Watts. The hardware for the more flexible Cav3 device geometry was completed and all of the aluminum pieces were anodized to minimize chemical interactions with the Cav3 housing, while at the same time providing a solid structure with which to perform adaptive experiments, Fig. 8. Vacuum integrity testing was performed and the new cavity held vacuum as well as the old cavity. Based upon results of Section 2.2, a transverse RF sustainer was chosen for the discharge. Based upon results of Section 2.3, a geometric Mach number of 2.4 was chosen for the supersonic nozzle. In future experiments we plan to test a transverse RF sustainer run at > 27 MHz and smaller diameter discharge tubes with the Cav3 hardware.

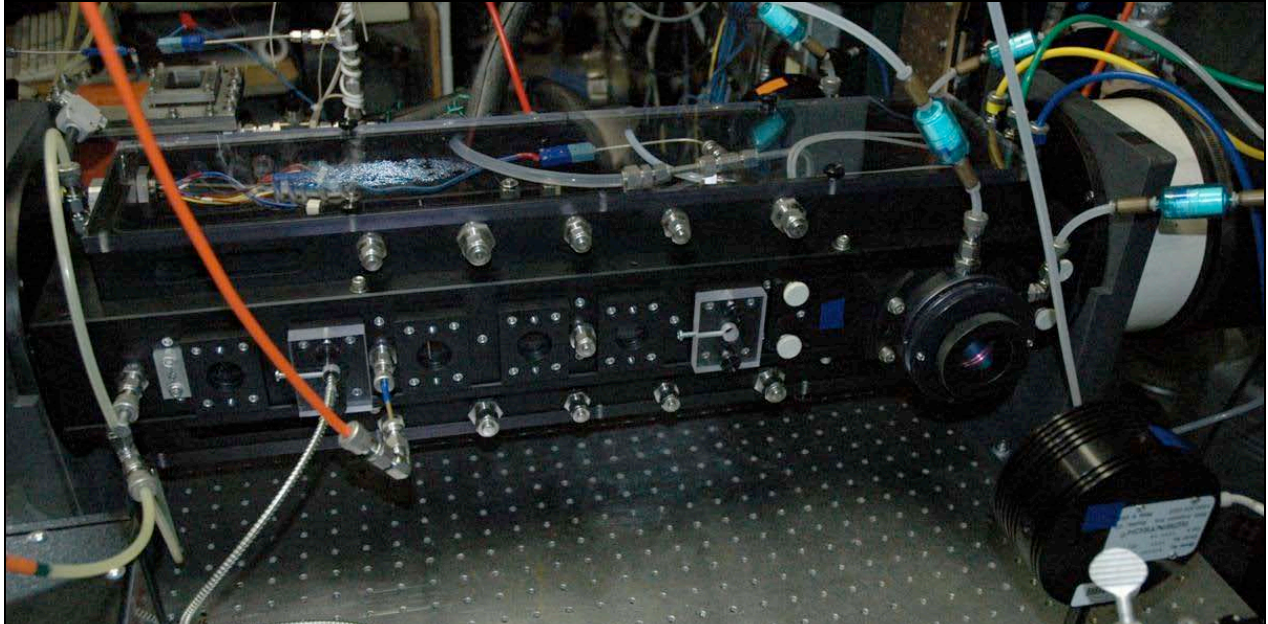


Figure 8. New more flexible and robust 3rd generation Cav3 ElectricOIL diagnostic-laser housing. The Cav3 housing has more diagnostic ports and flexible placement of injection positions for both I_2 and N_2 (if needed). The current internal flow channel is 5 cm wide (gain length), but the Cav3 housing is capable of expansion to a longer gain length (up to 15 cm) and multiple discharge tubes for future scaling experiments.

The Cav3 ElectricOIL was tested and the hardware available was optimized for gain and laser performance. One of the key improvements with the Cav3 hardware was the capability to place the I_2 and cold N_2 injection points at different positions. Previous modeling with the BLAZE-II code [Sentman, 1977; Carroll, 1995; Palla, 2006] indicated that it was important to have more flexibility in the placement of these injectors to optimize performance. The older Cav2 hardware did not have this capability and the I_2 injectors were essentially fixed at a position 36.8 cm upstream from the nozzle throat and the N_2 injectors fixed at a position 24.1 cm upstream from the nozzle throat. Figure 9 shows the dramatic improvement obtained by moving these injectors much closer to the throat. For these cases the cold N_2 injectors were always placed downstream of the I_2 injectors. We have found in the past that injecting the I_2 downstream of the cold N_2 tends to result in a large amount iodine build up inside the cavity, which is of course not desirable, and also always resulted in lower gain.

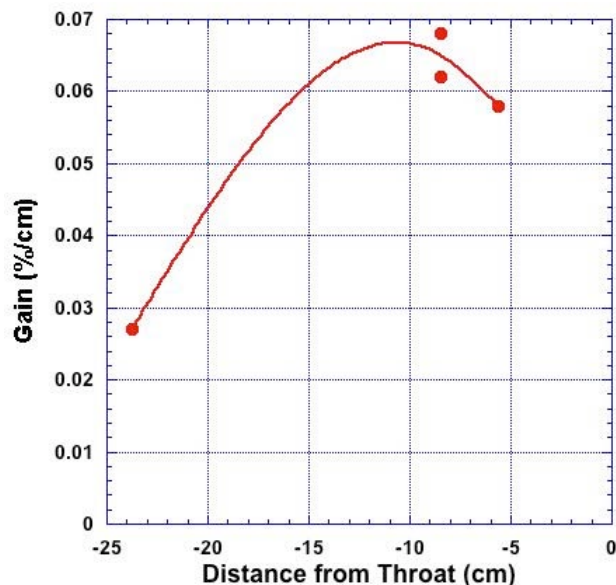


Figure 9. Gain vs. position of iodine injector holes relative to the nozzle throat (negative distance indicates distance "upstream" from the nozzle throat). Data taken with the Cav3 hardware and cold N₂ injectors downstream from the I₂ injectors. Flow conditions were O₂:He=10:33 at 20 Torr pressure and 1000 W RF power.

A gain of 0.067%/cm was achieved, Figs. 9 and 10, with the aforementioned hardware improvements. 2" diameter laser mirrors from AT Films with a reflectivity of approximately 99.997% were used and resulted in laser power of 4.52 W with the laser cavity gain length held fixed at 5 cm. Note that if mirror/absorption scattering losses were accounted for (which typically account for greater than a 20% loss in such low gain stable resonators), the power optically extracted from the flow would be > 5 W. A photograph of the elliptically shaped beam is shown in Fig. 11. The width of the long axis of the beam profile is 3.5 cm at the mirror. ***These results represent approximately a three-fold improvement in gain and laser power that has been achieved.*** We anticipate that continued work will result in a steady progression in performance enhancement of gain and laser power.

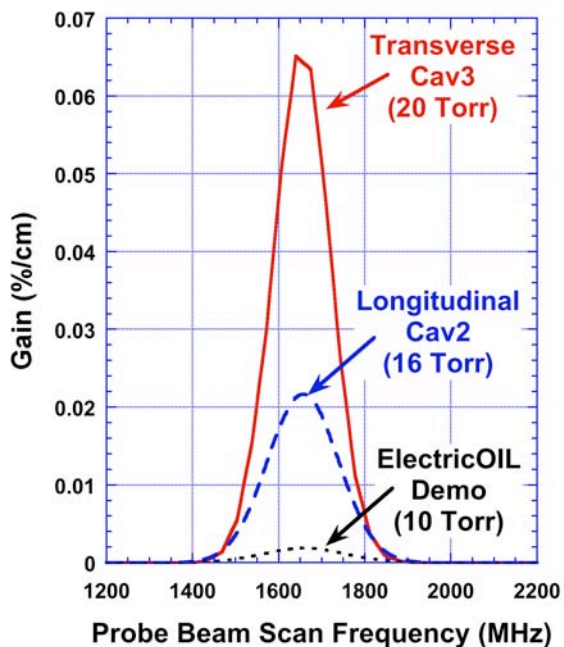


Figure 10. Digitally filtered gain signal in the supersonic cavity as a function of probe beam scan frequency for: (i) O₂:He=4:10 mmol/s with P_{total} = 10 Torr longitudinal discharge from the initial ElectricOIL demonstration with Cav2 hardware [Carroll, 2004], (ii) O₂:He=10:33 mmol/s with P_{total} = 16 Torr longitudinal discharge from prior work with Cav2 hardware [King, 2006], and (iii) O₂:He=10:33 mmol/s with P_{total} = 20 Torr transverse dipolar discharge resulting from the Cav3 ElectricOIL hardware. The temperature indicated by the linewidth of the 0.067% cm⁻¹ measurement is 112 K.



Figure 11. Photograph showing the 3.5 cm wide, elliptically-shaped 4.5 W laser beam on a thermal image card.

III. Modeling

To help guide and understand experiments, the new advanced BLAZE-IV electrodynamics-fluids-laser model [Palla, 2007] was employed. BLAZE-IV is a one-dimensional, fluid-dynamic, chemical-kinetic, non-equilibrium plasma-dynamic, flow model that was developed to investigate the physics critical to the production of $O_2(a)$ in ElectricOIL devices and the downstream kinetics and predictions of gain and lasing. BLAZE-IV is a new model based partially on BLAZE-II [Sentman, 1977; Carroll, 1995; Palla, 2006] the one-dimensional, fluid-dynamic, chemical-kinetic model from which the non-mixing, fluid-dynamic equations used in BLAZE-IV were derived. BLAZE-IV was developed using the C++ programming language in order to simplify the coding of complex physics by taking advantage of object-oriented programming techniques and appropriate data encapsulation. BLAZE-IV utilizes the one-dimensional momentum, continuity, energy, and species concentration equations (neutral and charged) and an implicit Newton iteration scheme. For more details on the BLAZE-IV model see Palla *et al.* [Palla, 2007], in which the model was validated and baselined to other experimental data.

For these studies we wanted to demonstrate the unique capability of the BLAZE-IV model to simulate the discharge region, carry the flow into and through the nozzle, and predict gain in the supersonic region of the flow. Figure 12 illustrates a single complete calculation that demonstrates these unique capabilities our new BLAZE-IV code. For this simulation the yield was predicted to be approximately 17.5% for this 20 Torr condition; experiments indicate a slightly lower yield of around 16.5%, but this is quite reasonable agreement for the 1D BLAZE-IV model. As has been observed in the past, there is a yield loss once the I_2 is injected (axial position 90 cm), but because the Cav3 hardware permits injection much closer to the throat we are able to minimize the loss of yield before the optical extraction region that starts at an axial location of around 100 cm. In previous BLAZE-II modeling [Palla, 2006] we were predicting as much as a 60% drop in yield by the time that iodine was in the laser cavity region because of the much larger distance between iodine injection point and laser cavity in the less flexible Cav2 hardware. The predicted temperatures are also in very reasonable agreement with experimental data, e.g., the predicted temperature in the laser cavity region is approximately 110 K, which is in excellent agreement with the temperature from the gain linewidth measurement, Fig. 10.

Figure 13 shows that the gain prediction for the 20 Torr flow conditions is in very good agreement with the measurements. Interestingly, there is a stronger gain region of 0.10-0.15% cm^{-1} in the subsonic plenum between the cold gas N_2 injector and the nozzle throat. Interesting experiments that could be performed in the future would be to (i) measure gain in this region, and (ii) place the optical resonator around this region and measure laser power; these experiments would require some hardware modifications, but should be possible.

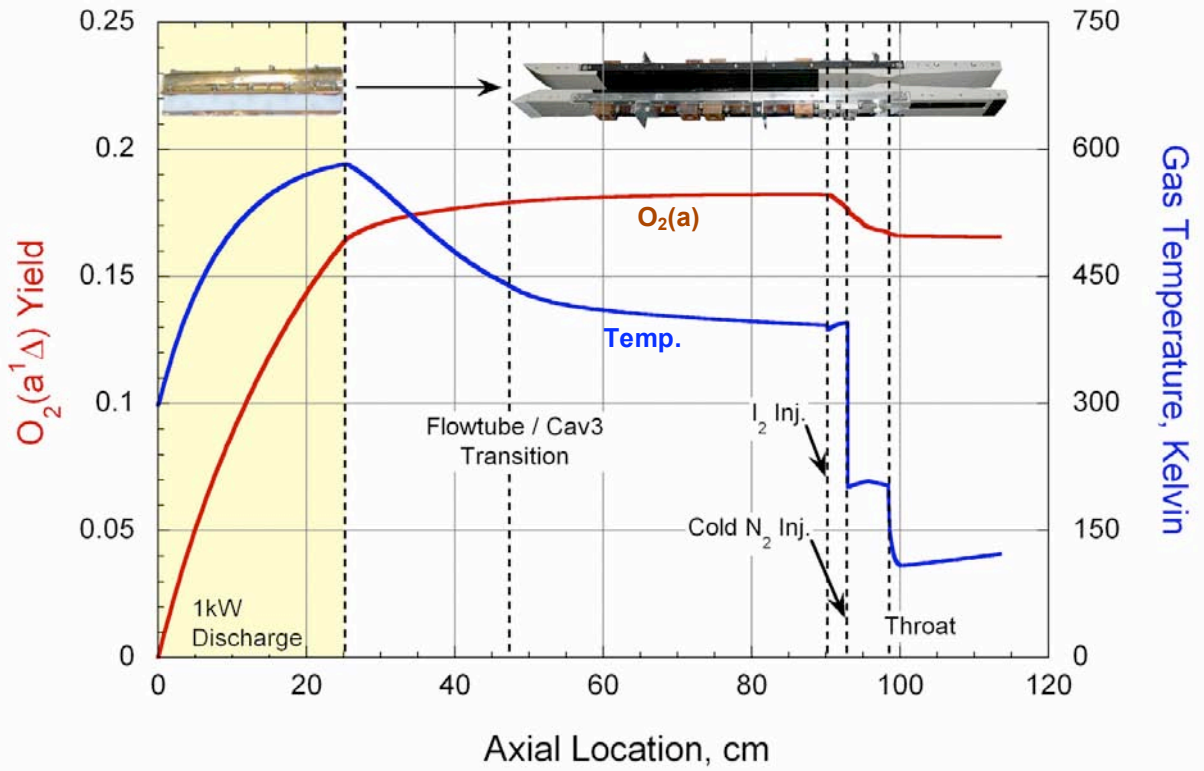


Figure 12. BLAZE-IV simulations of the more flexible and robust Cav3 ElectricOIL 5 cm gain length diagnostic-laser nozzle. The transverse discharge section and the Cav3 nozzle are shown for clarity.

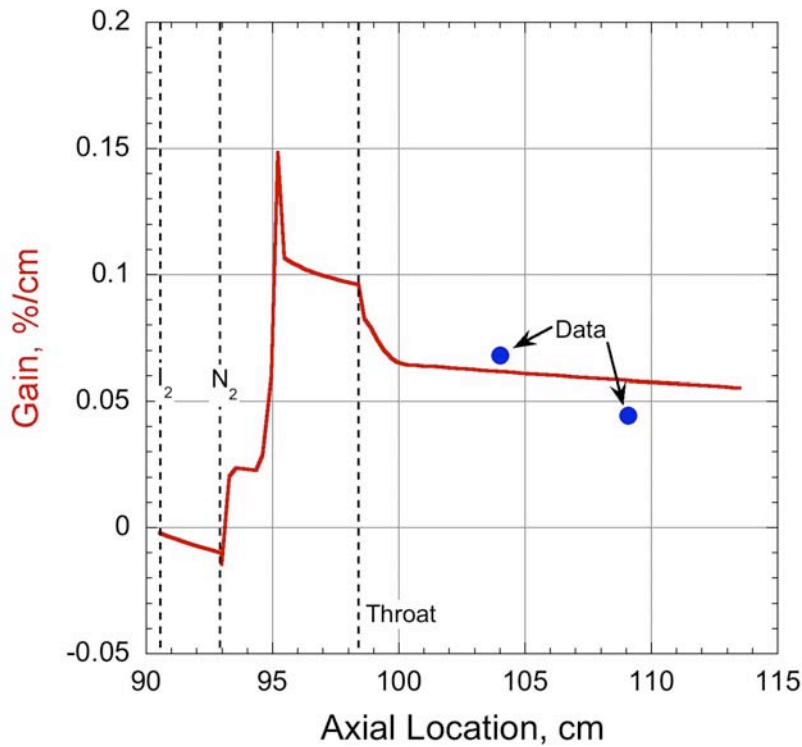


Figure 13. BLAZE-IV predictions of gain vs. axial position through the Cav3 supersonic nozzle. Data taken with the Cav3 hardware and cold N₂ injectors downstream from the I₂ injectors. Flow conditions were O₂:He=10:33 at 20 Torr pressure and 1000 W RF sustainer power.

VI. Concluding Remarks

Over the past three years of research and development, continual improvements in gain and lasing power have been obtained. $O_2(a^1\Delta)$ yields greater than 20% have been demonstrated along with positive gain and cw laser power in both supersonic and subsonic flow systems. *The gain has risen from the initial demonstration of 0.002% cm^{-1} by more than a factor of 30 \times to 0.067% cm^{-1} , and similarly the outcoupled laser power has risen from 0.16 W to 4.5 W (with a 5 cm gain length cavity). We are now obtaining $\approx 30\%$ energy coupling (and for the higher rf power cases more than 200 W of the power) into the desired $O_2(a)$ state, but significant improvements in understanding the role of components of plasma generated "active oxygen" still need to be made in regards to laser extraction of this energy. While O atoms permit rapid dissociation of the I_2 molecule, they appear to be major problem for energy extraction (as they also act as a quencher) and alternate I_2 dissociation schemes need be investigated.*

Significant results arising from this work are summarized as follows:

- Significant improvements in RF sustainer performance were obtained with a transverse "dipolar" discharge and when operating at higher RF frequencies. With the RF discharge operating at 13.56 MHz, approximately 30% of the discharge power was deposited into the desired $O_2(a)$ state at 20 Torr.
- The original geometric Mach number 2.4 nozzle had higher gain than was measured with either a Mach 2.67 or a Mach 3.0 nozzle.
- Flexible 3rd generation laser cavity hardware "Cav3" was designed, fabricated and tested. Using the transverse discharge and optimizing the location of the iodine injection relative to the supersonic nozzle throat and a geometric Mach # 2.4 nozzle, the small signal gain was improved to 0.067 % cm^{-1} .
- Modeling with the BLAZE-IV simulation code showed very good agreement with species, gas temperature, and gain data.
- Laser power output of 4.5 W was demonstrated with a 5 cm gain length cavity.

Acknowledgments

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