Power Enhancement of the Hybrid ElectricOIL Laser

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Recent experiments have led to improvements in the hybrid Electric Oxygen-Iodine Laser (ElectricOIL) system that significantly increased the laser power output. The continuous wave laser operating on the I(2P1/2) \rightarrow I(2P3/2) transition of atomic iodine at 1315 nm was pumped by the production of O2(a1Δ) in a radio-frequency discharge in a He/O2 mixture. Molecular iodine was injected into the flow products 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(2P1/2) state. The outcoupled laser power was used as a diagnostic to analyze the global effects of pressure, primary/secondary flow rates, sensitizer gases, discharge configuration and other system parameters.

I. Introduction

The classical chemical oxygen iodine laser (COIL) system employs a mixture of potassium hydroxide and hydrogen peroxide denoted “basic hydrogen peroxide” (BHP) with chlorine gas and inert diluents to produce the singlet delta electronic state of the oxygen molecule, O2(a1Δ). The singlet delta enjoys a near-resonant energy transfer relationship with iodine, populating the I(2P1/2) state of the iodine atom following the molecule’s dissociation. This laser has been heavily studied, and its history has been reported elsewhere.2 Recently, engineers at CU Aerospace (CUA) and the University of Illinois – Urbana Champaign (UIUC) have engaged in a research effort aimed at supplanting the liquid chemical singlet oxygen generator (SOG) with an electrically excited all gas phase system.3,4 Other research groups are also performing significant investigations of electric discharge SOGs.5-10 Carroll et al. have reported gain11,12 and laser powers up to 520 mW with this Electric Oxygen Iodine Laser (ElectricOIL) system.13,14 In this paper we discuss recent developments in ElectricOIL technology that have produced significant increases in laser power.

II. Experimental Setup

CUA and UIUC’s ElectricOIL research is carried out in a combination glass / quartz flow system utilizing either hollow aluminum electrodes, copper straps, or copper coils to couple in a 13.56-MHz radio frequency discharge to a primary flow comprising oxygen, an inert diluent, and sometimes a sensitizer gas. Iodine is injected through an aluminum annular injector approximately 81.3 cm downstream of the downstream electrode. A supersonic laser cavity with a 5 cm gain length is in place approximately 10.2 cm downstream of the iodine injector. Here, the primary effluent and secondary iodine injection are expanded through a Mach 2 nozzle to lower their temperature. Directly upstream of the cavity is the tertiary injector, where cold nitrogen at a temperature of ≈135 K is injected to raise the cavity inlet pressure, drive the nozzle, and further cool the flow. Pressure measurements in each flow section are achieved via capacitance manometers. Flow rates are monitored by a variety of thermal and

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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 tubing walls. (Field of view corrections must be made in these instances to maintain comparable calibrated yields.)

The block diagram representing the flow schematic for the system is shown as Figure 1. A photograph of the laser cavity is shown in Figure 2, and a photo of the primary discharge region with an active discharge is presented in Figure 3.

### III. Experimental Techniques

As discussed in prior works, the addition of small amounts of NO to the discharge region was found to be very beneficial.\(^{11-15}\) However, a difficulty with the addition of NO is the production of what is commonly referred to as the “air afterglow”, which is a very large broadband emission as evidenced by the yellow-green glow seen throughout the flow tube, Figure 3. This emission is caused by the emission of NO\(_2^*\) from the following reaction sequence:

\[
\begin{align*}
\text{NO} + \text{O} &\rightarrow \text{NO}_2^* \quad (1) \\
\text{NO}_2^* &\rightarrow \text{NO}_2 + \text{hv} \quad (2)
\end{align*}
\]

This broadband emission also shows up as a significant addition to the spectra, Figure 4(a) and (b), and the data must be corrected for this broadband emission to deconvolve the true magnitude of the O\(_2\)(\(a\)) signal. The true signal is deconvolved by first subtracting a slope from the signal, and second by adjusting the height, such that the
Corrected data has a relatively flat portion running through zero counts. To minimize errors, the signal is integrated from 1250-1300 nm. Reduction of all data through a numeric qualifying technique (manifested as a Microsoft Excel macro) enables repeatable and comparable results. Fixed spectrometer entrance slits, repeatable geometry (thanks to fiber optic coupling and rigid mounts), and mechanical data reduction techniques ensure that day-to-day variation from the O2(a1Δ) detection system is minimized. It was found that using kinematic (adjustable) slits on the spectrometer and monitoring only the spectral peak introduced day-to-day variations in the data that were not acceptable.

![Figure 4: Effect of NO flow on the observed O2(a) spectra: (a) O2(a) spectra without NO in the flow, and (b) O2(a) spectra with background afterglow and with data correction (afterglow digitally removed).](image)

Similar correction subtraction techniques are applied to O2(b1Σ) spectra about 761.9 nm and I* spectra centered at 1315.24 nm. The O2(b1Σ) is often quite tedious to separate from the airglow, as it exists in a very strong region of the broadband NO2* emission. However, the I* usually requires little to no baseline correction, however, as the spectra is quite narrow and large compared to the baseline.

Measurements of gain (or absorption) were made using the Iodine-Scan Diagnostic (ISD) developed by PSI, and Davis et al.16 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. The windows on the sides of the cavity when using the gain diagnostic were wedged and anti-reflection coated to minimize etalon effects. A 4-pass “White cell” configuration17 (20 cm path length) was used for the bulk of the gain measurements in the supersonic section. For a few gain measurements a 14-pass “Herriott cell” configuration18 (70 cm path length) was used. There were notable differences between the two configurations. First, the Herriott cell was considerably more difficult to align correctly than the White cell; operationally, this made the White cell a much more attractive option from a day-to-day standpoint. Second, the use of the Herriott cell resulted in gain measurements that were approximately a factor of 4 times smaller in magnitude at 300 W of RF power and approximately a factor of 2 times smaller in magnitude at 400 W of RF power than those obtained using the White cell configuration. We believe that this difference can be attributed to the fact that all 4 passes through the gain medium with the White cell were on the nozzle centerline where the flow velocity is highest, and correspondingly the flow temperature is its lowest, whereas the circular criss-crossing pattern of the Herriott cell samples the entire flow field including warmer boundary layer regions near the top and bottom nozzle walls. Since the gain is effectively proportional to an inverse function of temperature, the gain observed by the White cell configuration is understandably higher than that observed by the Herriott cell. A similar observation was made by Rawlins et al. when comparing a 40-pass Herriott cell with a single pass measurement.9 Figure 5 shows the measured difference between the White cell and Herriott cell configurations as a function of RF power for approximately the same flow rates and pressures. Given the operational simplicity of
setting up the White cell, gain results that we have reported in past work and in this work were taken with a White cell aligned on the centerline of the nozzle.

Scientech Astral model AC2500/AC25H calorimeters with Scientech Vector S310 readouts were used to measure the laser power. All data collected for this work employed a 1.0-micron long-pass filter in front of the power meters to minimize the effects of any stray illumination sources. The laser power meters have proven to be an invaluable diagnostic for quickly assessing the global performance of the laser system in an integrated sense, i.e. the laser power represents the summation of all effects when a change is made to baseline conditions. Hence, by monitoring the laser power it permits us to make flow rate and discharge adjustments and see the global effect on the laser performance in real time. The beam profile was monitored in a qualitative way via IR detection cards from New Focus (model 5842). Mode shifting is seen frequently when adjustments are made to the flow or discharge conditions; however, the beam profile was typically fairly uniform (as observed on the IR card) for conditions that provided maximum power on a given run day.

**IV. Power Enhancement Experiments**

A graphical summary of power increases for ElectricOIL is presented as Figure 6. The history of the power scaling effort is broken up into three major regions: Region 1 - “First Light and Initial Parameter Studies”, Region 2 - “Initial Power Enhancement”, and Region 3 - “Power Enhancement to >1 Watt”.

![Figure 5: Measured gain with a 4-pass White cell at flow centerline and a 14-pass Herriott cell that samples more of the flow field height (both curves represent series with 4 mmol/s O₂, 16 mmol/s He, and a subsonic discharge pressure of 10 Torr)]
The baseline configuration that provided first light was 3 mmol/s Oxygen, 16 mmol/s Helium, and 0.15 mmol/s Nitric Oxide. In these initial tests, NO₂ was admitted in the post-discharge region to titrate out some of the oxygen atoms produced in the discharge.¹¹ This was guided by substantial data and associated numerical studies suggesting that the \( \text{I}^* + \text{O} \rightarrow \text{I} + \text{O} \) quenching reaction¹⁹ was hurting laser power more than the \( \text{I}_2 + \text{O} \rightarrow \text{IO} + \text{I} \) and \( \text{IO} + \text{O} \rightarrow \text{O}_2 + \text{I} \) dissociation process was helping it. However, we quickly found that under higher total flow pressures this was not necessary – with sufficient NO in the primary, atom recombination or losses due to dissociation of the I₂ are sufficient to overcome the deleterious effects of the \( \text{I}^* + \text{O} \rightarrow \text{I} + \text{O} \) reaction.¹³,¹⁴

This higher-pressure series enabled repeatable laser power on demand, and a variety of other systematic investigations occupied the laboratory efforts for some time. Sweeps in iodine carrier flow rate and preliminary feedback from the modeling studies then guided the attempt to raise the secondary flow rates, increasing laser power from ~200 mW to ~480 mW.

The ability to monitor laser power, 1315-nm emission, and small signal gain on a real time basis allowed the research group to quickly parameterize a variety of flow rates, powers, pressures, etc. While previous data suggested that O₂(a) yields would drop with this longitudinal discharge at higher pressure and higher O₂ flow rates in the primary, the atomic Oxygen flow rate was also found to drop significantly with increasing pressure. Thus, it was possible that a small drop in O₂(a) yield might still result in higher gain and laser power due to higher O₂(a) number densities and lower O atom concentrations at higher pressure. As such, a sweep of flow rates and pressures was conducted and the result was a near double in total laser power to a new high of 974 mW.

The last significant increases in laser power originated from a fresh pair of mirrors and a higher I₂ flow rate. With optimum levels established for the 10:33:0.15 case at 16 Torr, mirrors #7 and #8 (see Table 1) yielded a new record ElectricOIL total power of 1.34 W. Later in this same test series, an investigation of higher I₂ flow rates demonstrated a somewhat higher 1.47 W of total laser power. The gain for this last power point was measured to be 0.022 % cm⁻¹.
Table 1: All laser mirrors employed during this reported research.

<table>
<thead>
<tr>
<th>Mirror #</th>
<th>Curvature</th>
<th>Manufacturer</th>
<th>Transmissivity, T</th>
<th>Reflectivity, R</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1m CC</td>
<td>LGR</td>
<td>0.000135</td>
<td>0.999865</td>
</tr>
<tr>
<td>2</td>
<td>1m CC</td>
<td>LGR</td>
<td>0.000143</td>
<td>0.999857</td>
</tr>
<tr>
<td>3</td>
<td>2m CC</td>
<td>LGR</td>
<td>0.000071</td>
<td>0.999929</td>
</tr>
<tr>
<td>4</td>
<td>2m CC</td>
<td>LGR</td>
<td>0.000070</td>
<td>0.999930</td>
</tr>
<tr>
<td>7</td>
<td>2m CC</td>
<td>ATF</td>
<td>0.000053</td>
<td>0.999947</td>
</tr>
<tr>
<td>8</td>
<td>2m CC</td>
<td>ATF</td>
<td>0.000051</td>
<td>0.999949</td>
</tr>
</tbody>
</table>

As noted in previous work\textsuperscript{11-14} we had observed that discharge instabilities occur at higher RF power levels. Early in the program, we found that these instabilities would begin to occur at around 300-400 Watts. One of the characteristics of these instabilities was the visible formation of streamers that looked like diffuse electrical arcs, oscillating around the electrodes inside the plasma. As power increased, the streamers would narrow in diameter and the oscillation frequency would increase. We hypothesized that the phenomenon enabling these streamers was localized hot regions of the discharge gas. In these regions, densities would decrease due to the localized heating. Unfortunately, this lowered the impedance, allowing more current to flow in that region. With more current came more heat, and the process carried forward until arcs were developed streaming from upstream to downstream electrode. We hypothesized that this effect could be minimized or delayed by a simple conditioning of the flow that would both discourage straight electrode-to-electrode current paths and enable localized hot regions to be quickly mixed back into the surrounding flow. To do this, we created multiple types of flow conditioners (augmentors) with angled holes that would "swirl" the gases prior to entering the discharge. The flow conditioner design that was most frequently used consisted of a flat plate with two rings of angled 3/16" holes (an inner ring of 4 holes and an outer ring of 8 holes) drilled through from one side of the plate to the other; these angled holes force the flow "swirl". Figure 7 illustrates the significant improvement in O$_2$(a) production with the flow conditioner (augmentor) in place upstream of the first discharge electrode. The flow conditioner demonstrated improved mixing of the gases, distributed the heat more uniformly in the discharge flow, and prolonged arcing in the discharge region to much higher powers, thereby improving the overall discharge stability. More electrical energy is coupled into the singlet delta state of O$_2$ with improved discharge stability, especially at higher power levels.

![Figure 7: O$_2$(a) signal vs. RF system power with and without flow conditioning (augmentation) prior to the discharge. Flow conditions were O$_2$:He = 10:40 at a total pressure of 10 Torr.](image-url)
V. Future Systems Enhancements

It has long been understood that control of the E/N parameter is critical to producing high yields of O$_2$(a). E is the electric field in units of V-cm$^{-1}$ and N is the gas number density in units of cm$^{-3}$. Theoretical calculations in Refs. 7 and 22 illustrate the functional dependence of how much electrical power can be deposited into the important O$_2$(a) and O$_2$(b) states as a function of E/N (or equivalently electron temperature). With the longitudinal, capacitive, and inductive RF discharges, we typically operate with an average E/N in the range of 15 to 25 Townsend (Td), dependent upon flow conditions. [Note: 1 Td = 10$^{-17}$ V-cm$^2$.] From Refs. 7 and 22 it is clear that optimal power deposition occurs in the region of 5-10 Td.

A concept being pursued by the CU Aerospace / University of Illinois team$^{14,20}$ and other groups in different forms$^{0,21,22}$ is that of the “Pulser-Sustainer” (PS) discharge. The basis for the idea is that a repeated high power pulse can provide more than the necessary amount of ionization. This pulse is followed by a sustained electric field tuned to operate at the optimal E/N. The concept theorizes that the pulser provides enough electrons such that the value of the electric field, E, can be controlled by the sustainer field, Figure 8. The optimum repetition rate of the pulser is determined by the decay of excess ionization. The other primary advantages that we foresee for the PS are a more stable discharge at higher average power and at higher pressure capability, both of which are critical issues for scaling the system.

![Figure 8: Pulser-Sustainer concept: high voltage pulse that creates an excess of electrons is followed by a sustainer field to control the electric field for optimal E/N. The pulse repetition rate is determined by the decay of the excess ionization. A bipolar pulser-sustainer is illustrated, but it could also be unipolar.]

A local power electronics startup, SmartSpark Energy Systems, built all of the PS systems tested in this paper. The first PS-1 system was un-cooled and failed in preliminary tests. SmartSpark added passive cooling to the second PS-2 system, Figure 9. Initial results were encouraging with the PS-2 unit in the low average power (up to ≈ 250 W) regime. Figure 10 shows how the O$_2$(a) signal was increased by nearly a factor of two as the pulse to sustainer power ratio was increased. A simultaneous drop in the RF coil current of the inductive sustainer was observed. This is significant because it can be shown mathematically that the electric field E drops as the inductive coil current drops, which means that the observed drop in coil current corresponds to a desired drop in E/N.

![Figure 9: Photograph of the early PS-2 unit. PS-2 used an inductive RF sustainer]

![Figure 10: RF coil current and O$_2$(a) signal with the PS-2 unit as a function of the ratio of pulse power to sustainer power. 0.4 ms pulses added to a 50 W inductive sustainer. O$_2$:He flow rates of 3:16 mmol/s at a total pressure of 3.6 Torr.]
Given the positive results with the PS-2 unit, we decided to have SmartSpark continue their work and scale up the system to higher power. The initial version was passively cooled; the PS-3 version was actively water-cooled, Figure 11. Testing with this modified version of the PS took us up to average power levels of \( \approx 700 \) W. Typical V-I characteristics for the PS-3 system are illustrated in Figure 12; of importance is the relative cleanliness of the oscilloscope traces, i.e. the pulses show very little “ringing” that is typical of such electronic systems, and the sustainer current slowly falls as the ionization decreases after the pulse. The PS-3 unit used a DC square-wave sustainer, and hollow cathode electrodes were used for both the pulse and sustainer simultaneously. Figure 13 shows the \( \text{O}_2(a) \) yield vs. total input power (pulser + sustainer) as a function of pulser repetition frequency and for two different sets of flow rates. Most importantly, Figure 13 shows that the higher frequency of 100 kHz provided approximately twice the yield as that with the 50 kHz pulser. The other promising feature from Figure 13 is that the discharge did not develop any significant discharge instabilities as were observed with the RF discharges at around 500 W.

\[ V_s = 697 \text{ V}, I_s = 900 \text{ mA} \]
\[ V_s = 166 \text{ V}, V_{dc} = 531 \text{ V} \]
\[ E/N \approx 1.9 \times 10^{-15} \text{ V cm}^2 \text{ cm}^{-2}, P_{p} / P_{s} \approx 5.8 \]

The early PS-2 and PS-3 experiments were very encouraging and have demonstrated three primary advantages with this concept, (i) some control of the critical E/N parameter, (ii) a more stable discharge at higher average power, and (iii) a more stable discharge at higher pressure, all of which are critical issues for scaling the system. We reached the limits of the PS-3 system at \( \approx 700 \) W and 100 kHz. As such, we have commissioned...
SmartSpark to build a more robust, higher power, higher frequency PS-4 system to be tested in the future. Methodologically, we have chosen an approach of testing smaller systems first to learn how they perform under a variety of operating conditions, followed by progressive upgrades in the directions indicated by experiments and theory.

VI. Concluding Remarks

The ElectricOIL power enhancement efforts by CUA and UIUC have resulted in an electrically pumped iodine laser of 1.47W total power; this is the highest power recorded to date in an electrically-pumped oxygen iodine laser. Key technological difficulties encountered during this enhancement effort included spectral deconvolution techniques, gain diagnostic path-methodology pros and cons, discharge instabilities, and assessment of the role played by the oxygen atom. By employing the laser power itself as a diagnostic, a variety of system parameters were identified and optimized. Discharge instabilities in the O₂(a) production have limited our total laser power to ~1.5W, but we believe that new discharge types and implementations, coupled with a more complete understanding of the complex electro-chemical kinetic and plasmodynamic processes will overcome these difficulties.

Considerations for the oxygen atom appear to be a dominating factor in the successful extraction of stimulated emission in the electric oxygen iodine laser. Unfortunately, measurement of the atoms proves difficult in a discharge containing nitric oxide. Both actinometric and titration techniques are being refined in the lab to accommodate the broadband airglow emission. Preliminary results reported by Carroll et al.¹² indicate that the atoms have a deleterious effect on the I* production. This is verified in the present work.

It is also found that the role of NO in the primary helps to mitigate these deleterious effects as well as increase the O₂(a₁Δ) production efficiency in the discharge. Although NO₂ was found to produce lasing when employed as a sensitizer in the primary discharge, it was found to have a lesser effect than NO in that role. NO₂ quickly reacts with any oxygen atoms in the discharge,²¹ breaking down into NO via reaction (3), and it is hypothesized that a significant fraction of the O₂ produced is actually O₂(a).¹⁵

\[ \text{NO}_2 + O \rightarrow \text{NO} + \text{O}_2(X,a,b) \]  

From here, the NO₂, now present as NO, carries on as before, boosting O₂(a) production¹⁵ and continuing to scavenge oxygen atoms via the path in reactions (1) and (2) above. A delicate balance in the desired atom level appears to exist. Over-titration quickly degrades laser power, while under-titration has the same effect. Clearly, the atoms are serving a dual role. On the one hand, they are quenching I* via the methods first described by Heaven and as measured by Azyazov et al.¹⁹ On the other, they are dissociating the molecular iodine, saving a precious singlet delta molecule from this job and reserving it to pump the iodine atom instead. An optimum between these two (and likely other) reactions is located experimentally.

Preliminary pulser-sustainer experiments have been performed in an effort to more efficiently couple electrical power into the oxygen. Note that these early experiments have been performed with limited power pulsers so that we have not been able to adequately expand the operational envelope, but higher power pulsers are presently being built.

Acknowledgments

This work was supported by the Air Force Office of Scientific Research (AFOSR), the Missile Defense Agency (MDA), and the U.S. Army Space and Missile Defense Command (USA/SMDC). The authors would like to acknowledge the contributions of: T. Madden (Air Force Research Laboratory); G. Hager (University of New Mexico); M. Kushner (Iowa State University); S. Davis and T. Rawlins (Physical Sciences Inc.); M. Heaven and K. Morokuma (Emory University); G. Perram (Air Force Institute of Technology); J. Kimball and B. Nee (SmartSpark Energy Systems); M. Berman (AFOSR); J. Mulroy (MDA); B. Otey (USA/SMDC); A. Ionin (P.N. Lebedev Physics Institute); T. Rakhimova and Y. Mankelovich (Lomonosov Moscow State University). We would also like to thank B. Wheaton and A. Roberts for their technical assistance.

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