Advanced Gas Laser Experiments and Modeling

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Over the last decade new advanced gas lasers have emerged as possible candidates for high power laser systems that may supplant more conventional chemical and gas laser systems. Among these are the electric oxygen-iodine laser (ElectricOIL), the diode pumped alkali laser (DPAL), the exciplex pumped alkali laser (XPAL), and the optically pumped metastable rare gas laser. In this paper we will primarily focus on the ElectricOIL and XPAL systems, and discuss some of the recent experiments and modeling of these systems.

I. Introduction
Gas and chemical lasers have been in existence since the 1960’s. Systems have utilized CO₂, CO, various halide species (HF/DF, HCl, HBr, and HI), a wide variety of excimer species, many metal vapor systems, He-Ne, He-Cd, oxygen-iodine systems, alkali metals, and recently optically pumped metastable rare gases. Some of these systems are well-established and continue in their use, and others have faded in popularity. An excellent collection of topical chapters on gas laser systems was compiled by Endo and Walter [Endo, 2007]. A brief history of high energy laser systems is provided by Carroll [Carroll, 2011]. Books by Perram et al. [Perram, 2010], and Nielsen [Nielsen, 2009] provide a more complete look at high energy laser systems and their effects, respectively. The subject of this paper is to take a closer look at some of the more recent advanced gas laser systems that are receiving attention because of an attractive mix of hybrid electrically driven gas phase characteristics.

II. The Electric Oxygen-Iodine Laser
The electrically driven oxygen-iodine laser (ElectricOIL) that was first demonstrated in 2005 [Carroll, 2004; Carroll, 2005] operates on the electronic transition of the iodine atom at 1315 nm, I(3P₁/₂) → I(3P₃/₂) [denoted hereafter as I* and I respectively]. The lasing state I* is produced by near resonant energy transfer with the singlet oxygen metastable O₂(a’A) [denoted hereafter as O₂(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, 2005; Hicks, 2006] and laser power [Hicks, 2006; Davis, 2008; Bruzese, 2009]. Computational modeling of the discharge and post-discharge kinetics [Stafford, 2004; Palla, 2007] has been an invaluable tool in ElectricOIL development, allowing analysis of the production of various discharge species [O₂(a’Δ), O₂(b’Σ), O atoms, and O₅] and determination of the influence of NO₅ species on system kinetics. Ionin et al. [Ionin, 2007] and Heaven [Heaven, 2010a] provide comprehensive topical reviews of discharge production of O₂(a) and various ElectricOIL studies. The highest gain in an ElectricOIL device reported to date is 0.30 % cm⁻¹ [Benavides, 2012], and the highest output power reported is 538 W [Benavides, 2012].

2.1 Experiments
The ElectricOIL system consists primarily of (i) the flowing gas discharge for creating O₂(a), (ii) the heat exchanger (HX) downstream of the discharge to cool the discharge flow, (iii) the iodine injectors and injection region, (iv) the nozzle and laser cavity extraction region, and (v) the diffuser to more efficiently slow the gas flow down and bring

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the flow to higher pressures. These systems are coupled together in terms of the entire laser system performance, i.e. one element will influence the performance of other elements upstream and downstream throughout the system. Therefore, optimizing any single element on its own does not necessarily optimize it for the entire laser system; however, optimizing individual elements will typically provide rough operating regimes that will help guide the optimization of the whole system. Experiments provide the only reliable way to test these different operating regimes on a component and system level.

Over the past 5 years of research and development of the ElectricOIL device, higher performance and efficiency have been consistently obtained by moving towards higher operating flow rates and pressures. Towards this end, discharge geometries that can simultaneously handle high flow rates and higher pressures while maintaining O₂(a) yields are the most attractive for laser power extraction. Heat exchanger technology that also permits good temperature reduction and atomic oxygen recombination while minimizing any loss of O₂(a) yield is also of great importance. These effects were the focus of recent experiments along with their impact on laser gain in the supersonic flow cavity and are summarized herein.

2.1.1 Cav7 ElectricOIL Apparatus

Figure 1 shows a schematic of the Cav7 ElectricOIL experimental apparatus. The device has three 13.56 MHz RF discharge modules fed by pre-mixed O₂/He/NO. In each module two tubes are concentrically aligned with electrodes on either side of the dielectric barrier gap of 0.75 cm created by the quartz tubes, Fig. 2. The inner brass electrode is cooled internally by water flow. The high-pressure O₂(a) yield performance of this type of discharge compared to other configurations was discussed by Woodard et al. [Woodard, 2011]. The selection of discharge gap used in this device was motivated by a detailed series of work summarized by Braginsky et al. [Braginsky, 2007].
Fig. 2. Photograph and cross-sectional schematic of the concentric tube transverse discharge with high voltage electrodes on the outside of the outer quartz tube and a grounded, water-cooled electrode inside the inner quartz tube. Flow is from left to right in the photograph and into the page in the schematic.

The flows exiting the discharge modules enter independent cross-flow heat-exchanger modules which use Syltherm XLT coolant chilled to -30°C by LN2. Some trade studies used to develop heat-exchangers for ElectricOIL are summarized in [Woodard, 2011] and [Zimmerman, 2009]. The design used in Cav7 experiments is reported in [Benavides, 2012].

The flows exiting the heat-exchanger modules enter a plenum in which iodine vapor (carried by helium) is injected (secondary injection) followed by chilled nitrogen (tertiary injection). The combined flow is then expanded into a 2-D Mach 2.4 nozzle (geometric) to approximately Mach 2. Within the supersonic flow, the temperature is reduced, the iodine population inversion is attained ($\langle I^* \rangle > 0.5 \langle I \rangle$) and power is extracted by means of a resonator formed by high-reflectivity mirrors mounted on either side of the supersonic flow channel. Mounting hardware for a dual resonator combination was fabricated, consisting of two 50.8-mm dia. mirror resonators. The width of the supersonic channel corresponding to the gain length of the resonators is 22.9 cm. Downstream of the supersonic laser cavity region is a supersonic diffuser which transitions the flow to higher pressure subsonic flow prior to the pumping system which consists of four series-staged Roots-type blowers backed by two (parallel) Kinney model KT-850 rotary pumps. The gases used in experiments were laboratory grade oxygen and helium (99.95% pure), high purity nitric oxide (99.99% pure), and gaseous and liquid dry nitrogen were supplied from a liquid nitrogen tank at the facility. The iodine vapor was carried to the experiment by helium passed through a heated chamber containing a bed of solid iodine chips (99.5% pure).

2.1.2 Discharge Experiments

With three concentric discharge tube modules similar to the one applied to Cav6, the Cav7 system is capable of exciting large flow rates of O$_2$(a). Figures 3 and 4 show measurements of the power carried by O$_2$(a) as a function of O$_2$ flow rate into the discharges, holding the ratios of O$_2$, He and NO input flows constant, while also holding the ratio of input power to O$_2$ flow rate (O$_2$ specific power) constant. This scaled-up system can provide approximately 2.5 kW of power carried by the O$_2$(a) flow. The efficiency was constant as total discharge flow rate was increased proportionally to RF power.
Fig. 4.  O$_2$(a) power vs. input oxygen flow rate for four different discharge configurations. The pressure for each range of data is noted. The flow rate ratio was 1:3.33:0.01, O$_2$:He:NO for all cases, and power was 100 W/mmol/s of O$_2$.

2.1.3 Gain experiments

Figure 5 shows the gain measured at the conditions for best laser power in Cav7 with the dual resonator to be 0.184 %-cm$^{-1}$, corresponding to $g_0L = 0.042$ and a temperature of 148 K. For reference, the best gain results for Cav6 (L = 7.62 cm) corresponded to $g_0L = 0.023$. It should be noted that the gain measured in Cav7 is lower than that measured in Cav6 [Benavides, 2012]. We believe the primary reason for this is a mode beat between the use of multiple power supplies for the discharges that results in lower average O$_2$(a) and lower gain.

2.1.4 Power Extraction Measurements

The flow conditions for best power extraction were as follows: 165:570:2.3 mmol/s O$_2$:He:NO through the discharge, 0.35:150 mmol/s I$_2$:He in secondary injection, and 915 mmol/s N$_2$ in tertiary injection. The RF power input per tube was 6 kW. The highest CW total power extraction was 538 W. Figure 6 shows a comparison of outcoupled power versus mirror reflectivity for three different gain length ElectricOIL systems (5.1 cm Cav5, 7.6 cm Cav6, and 22.9 cm Cav7) all having 5 cm diameter mirrors. Note the dramatic superlinear increase in outcoupled power as the gain length is increased; using only 5 cm diameter mirrors, Cav7 shows a 7-fold increase in output power (393 W vs. 55 W) above Cav6 performance for only a 3-fold increase in gain length (22.9 cm vs. 7.6 cm).

Presently the ElectricOIL system is exhibiting superlinear enhancement of power output with $g_0L$ with various versions of ElectricOIL, Fig. 7. The slope of the line shown in Fig. 7 indicates approximately a factor of 30 increase for every factor of 10 increase in $g_0L$. How much further this superlinearity will hold as $g_0L$ increases is uncertain, but the trend is intriguing.
Fig. 6. Laser power versus reflectivity data and Rigrod model for three ElectricOIL laser cavities. The data shown are for 5 cm diameter mirrors in a standard resonator configuration.

Fig. 7. Laser power of various ElectricOIL configurations as a function of \( g_0 L \).

2.2 Modeling

In an effort to better understand the ElectricOIL system and how to further improve performance, we use the BLAZE-IV/V code [Palla, 2007; Palla, 2010b] to simulate the laser system as a whole, including the discharge, heat exchanger, nozzle, and laser cavity sections of the device. Details of BLAZE-IV/V and the complete kinetic package included in the latest version of the model are provided in [Palla, 2010b] and a summary of the more detailed modeling of the Cav6 device (upon which the Cav7 design is based) is provided in [Benavides, 2012].

2.2.1 Cav7 Power Extraction Modeling

The BLAZE IV model was used to simulate the performance of Cav7, investigating the influence of a mechanism that competes with the standard COIL pumping reaction. This competing mechanism is simulated by modifying both the forward and backward rates of the reaction,

\[
O_2(a) + I \rightleftharpoons O_2(X) + I^*
\]

while maintaining the ratio of forward and backward rates (equilibrium constant). The effect of this mechanism is to slow the rate at which power can be extracted from the gain medium.

Figure 10 shows the modeled power output of Cav7 with standard rates and with rates slowed to 31% of standard to simulate a competing (not quenching) mechanism. This reduced rate was sufficient to model the gain recovery measurements reported in [Zimmerman, 2010; Palla, 2010b]. The simulation uses Fabry-Perot optics with \( R_1 = R_2 = 0.997 \), and the cavity flow conditions of 170:556:3.3 mmol/s \( O_2:He:NO \), 55 Torr, 18 kW RF input. These values are characteristic of the flows and power input of ElectricOIL Cav7. As there is a significant level of power carried by the \( O_2(a) \) flow, the extracted power increases substantially as the resonator length is increased along the flow direction. The effect of the slowed pumping rates (competing mechanism) is to significantly reduce the slope of power extraction with resonator length.

In Cav7 experiments (Sect. 2.4), using two 2-inch (50.8-mm) diameter resonators, approximately equivalent to a single 4-inch (101.6-mm) resonator, an extracted power of 538 W was measured; Fig. 8 predicts only 392 W with the forward and backward rates slowed to 31% (based on Cav6 modeling with 7.6-cm gain length) and 911 W for the nominal pumping rate. Via linear interpolation, this suggests that a better modeling match with this larger gain length Cav7 hardware data would be obtained with the forward and backward pumping rates slowed to only 50% of their nominal values. In prior modeling of 5.1 cm gain length hardware [Zimmerman, 2009] the rates had to be slowed to 25% of nominal. Table I shows how reduction in the forward and backward pumping rate that best
matches data changes for different values of \( g_0L \) in these ElectricOIL devices. The implication is that the rate is closer to nominal in ElectricOIL for increased \( g_0L \), though the cause remains unidentified. Whatever this unidentified competing mechanism is, it appears to become less significant as \( g_0L \) increases, leading to the observed superlinear growth in extracted power with increasing \( g_0L \) (Fig. 7).

![Fig. 8. BLAZE modeling of outcoupled laser power as a function of resonator length along the axial flow direction, showing the influence of gain recovery mechanism.](image)

<table>
<thead>
<tr>
<th>Configuration (gain length)</th>
<th>( g_0L )</th>
<th>Rate Reduced To:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cav5 (5.08 cm)</td>
<td>0.011</td>
<td>25% of Nominal</td>
</tr>
<tr>
<td>Cav6 (7.62 cm)</td>
<td>0.023</td>
<td>31% of Nominal</td>
</tr>
<tr>
<td>Cav7 (22.86 cm)</td>
<td>0.042</td>
<td>50% of Nominal</td>
</tr>
</tbody>
</table>

2.3 Discussion and Future Work

Factors that will influence improved ElectricOIL performance in future work include:

- Discharge characteristics that promote high yields of \( O_2(a) \); the primary parameters which influence \( O_2(a) \) production in RF discharge are diluent ratio (He/\( O_2 \)), power-per-\( O_2 \) flow rate, operating pressure, transverse electrode gap, and excitation frequency.
- Scaling the ElectricOIL device in gain length requires a more complex manifold for delivery of cold nitrogen to the system to improve injector temperature uniformity.
- Reduce residence times throughout the system, minimizing impact of kinetic loss mechanisms on the \( O_2(a) \) population.
- Systematic integration of an electrical iodine predissociation technology is predicted to be highly beneficial, such as that implemented by Benavides et al. [Benavides, 2008], Fig. 9.
- Minimize pressure drop throughout the system.
- Improved resonator design to optimize the power extraction in the ElectricOIL system.

![Fig. 9. Schematic of the electrical iodine predissociator concept implemented into the earlier Cav3 hardware.](image)
III. Alkali Lasers

Forms of alkali pumped laser systems have been around for decades, however the more recent variations have the potential for both high electrical-to-optical efficiency and high beam quality for high energy systems. This section will examine the diode pumped alkali laser (DPAL) and its cousin the exciplex pumped alkali laser (XPAL).

3.1 Diode Pumped Alkali Laser (DPAL)

Approximately nine years ago, Krupke et al. [Krupke, 2003] demonstrated an optically-pumped atomic Rb laser operating on the resonance line at 795 nm. An equivalent pictorial representation for the Cs version of DPAL is illustrated in Fig. 10. Several other important studies followed the initial DPAL demonstration including theoretical modeling [Beach, 2004; Hager, 2010; Komashko, 2010], demonstration of lasing with other alkalis [Beach, 2004; Zhdanov, 2007a], lasing with helium as the \( ^3P_{3/2} \rightarrow ^2P_{1/2} \) relaxer gas rather than ethane [Zhdanov, 2007b; Zweiback, 2009], a multi-diode pump scheme has produced 48 W of output power at 894 nm [Zhdanov, 2008], and a high power diode stack has produced an average output power of 145 W at 794 nm [Zweiback, 2010]. A flowing DPAL with an output power of \( \approx 1 \) kW and an optical-to-optical electrical conversion efficiency of \( \approx 48\% \) was recently reported [Bogachev, 2012]. An excellent topical review of DPAL is provided by Krupke [Krupke, 2012].

The level of interest in the laser community has risen because it appears that this laser may offer a route to extremely high power levels. The primary reason for the interest is that it allows one to use high power semiconductor laser diodes as the pump source to drive a gas laser. The disadvantage of the basic DPAL pumping scheme is that it must be pumped in a spectrally very narrow (= 10 GHz, or equivalently \( \approx 0.02 \) nm) range such that only a limited portion of the semiconductor laser power will be absorbed by the alkali vapor because common semiconductor lasers, which typically emit with spectral widths of \( > 1000 \) GHz (roughly 2 nm). The ways to address this issue are to add high pressures (> 10 atm) of He gas to broaden the linewidth of the transition [Krupke, 2004], to narrow the linewidth of the pump laser, or a combination of pressure broadening and narrow linewidth diodes. While significant advances have been made in the area of narrow linewidth diode stacks [Podvyaznyy, 2010], this approach dramatically increases the cost, and scaling of such narrow linewidth diode stacks to very high power is still uncertain.

A sampling of available DPAL data are provided in Fig. 11 showing average output power as a function of average pump power for a variety of pulsed, quasi-CW, and CW configurations. The pulsed data all show excellent potential in terms of having very good optical-to-optical efficiencies \( > 50\% \) [Beach, 2004; Ehrenreich, 2005; Zweiback, 2011]. The quasi-CW data of Zhdanov et al. [Zhdanov, 2008] having a 10% duty cycle also show good slope efficiency, but then some different behaviors begin to emerge as one goes to higher average powers above 20 W, i.e. efficiency starts to drop significantly in the static cells [Zhdanov, 2008; Zweiback, 2010]; even the quasi-CW data above 100 W average pump power show an optical-to-optical efficiency of \( < 20\% \) [Zweiback, 2010]. Several groups believe that this roll off in performance in the static cells is due to thermal gradient issues and other effects such as “laser snow” (a degradation of the buffer gas due to the alkali and the lasing process), to which at least a partial solution is the utilization of a flowing DPAL system. The first report of a flowing DPAL occurred in 2010 [Rodriguez, 2010], and the recent demonstration in Russia [Bogachev, 2012] of a flowing DPAL having an output...
power of ≈ 1 kW and an optical-to-optical efficiency of 48% supports the hypothesis that the answer to scaling a DPAL system is to flow the gas medium. Much is yet to be determined regarding flowing DPAL systems, including the logistics of handling progressively larger quantities of the alkali metal in a closed-loop system as the device is scaled.

3.2 Exciplex Pumped Alkali Laser (XPAL)
The XPAL system approach takes a different course to this problem by invoking a molecular interaction to allow one to pump away from the atomic resonance in a broadband absorption blue satellite created by naturally occurring collision pairs such as Cs-Ar. XPAL was demonstrated by Readle et al. [Readle 2008, 2009a-b, 2010a-b] in mixtures of Cs vapor, Ar, and ethane, by pumping Cs-Ar atomic collision pairs and subsequent dissociation of diatomic, electronically-excited CsAr molecules (exciplexes or excimers). The four- and five-level variations of the pulsed XPAL system have also been modeled by Palla et al. [Palla, 2010a; Palla, 2011]. A more detailed discussion of XPAL, as well as differences and similarities to DPAL systems may be found in [Readle 2008, 2009a-b, 2010a-b; Heaven, 2010b; Galbally-Kinney, 2010].

For conceptual clarity, Cs-Ar interaction potentials including illustrations of the two Cs-Ar XPAL system pumping pathways and the four- and five-level laser operation mechanisms are shown in Fig. 12. The four- and five-level laser operation mechanisms are further illustrated in Fig. 13.

Figure 12. Interaction potentials of Cs-Ar. The arrows indicate the various pumping pathways, with two variations of the XPAL scheme shown. Laser action at 852.1 nm and 894.3 nm correspond to four- and five-level operation respectively.

Figure 13. Pictorial representations of four- and five-level XPAL operation respectively using either pumping pathway.

In this paper we present a simple theoretical model to make preliminary predictions of continuous-wave (CW) laser performance and to investigate the expected conditions to reach optical transparency in different gas mixtures and temperatures.

3.3 XPAL Theoretical Model

3.3.1 Theoretical Model and Assumptions
To enable quick studies for CW performance, a simple analytic model was created for the four-level XPAL system. Assumptions in the theory include:

(i) that the equilibrium between the Cs(6^2S_{1/2}) and CsAr(X^2Σ^+_{1/2}) states is maintained in steady-state lasing,

(ii) that the equilibrium between the Cs(6^2P_{3/2}) and CsAr(B^2Σ^+_{1/2}) states is maintained in steady-state lasing,

(iii) the CsAr(A^2Π_{1/2}) and CsAr(A^2Π_{3/2}) states do not play a significant role and are not included (assumed to be negligible),

(iv) the Cs(6^2P_{1/2}) state does not play a significant role in the four-level system,

(v) and spontaneous emission from the CsAr(B^2Σ^+_{1/2}) state is neglected.

The species in the model are denoted as: \( N_0 \) for Cs(6^2S_{1/2}), \( N_f \) for CsAr(X^2Σ^+_{1/2}), \( N_2 \) for CsAr(B^2Σ^+_{1/2}), \( N_3 \) for Cs(6^2P_{3/2}), and \( M \) for Ar, Fig. 14. Note that the precise species of alkali and rare gas can be replaced by other alkalis or rare gases, e.g., one could use Rb in place of Cs, and Kr in place of Ar. First we define the equilibrium fractions \( f_{10} \) and \( f_{23} \), which provide relations between \( N_f \) and \( N_0 \), and \( N_2 \) and \( N_3 \):

\[
f_{10} = \frac{N_1}{N_0} = \frac{[CsAr(X^2Σ^+_{1/2})]}{[Cs(6^2S_{1/2})]} = \frac{g_1}{g_0} 4\pi R_0^2 \Delta R \exp \left( \frac{-\Delta E_{10}}{k_b T} \right) [M] \tag{E.1}
\]

\[
f_{23} = \frac{N_2}{N_3} = \frac{[CsAr(B^2Σ^+_{1/2})]}{[Cs(6^2P_{3/2})]} = \frac{g_2}{g_3} 2\pi R_0^2 \Delta R \exp \left( \frac{-\Delta E_{23}}{k_b T} \right) [M] \tag{E.2}
\]

where \( k_b \) is the Boltzmann constant, \( R_0 \) is the optimal internuclear separation (4.5 Å for Cs-Ar) for the blue satellite, \( \Delta R \) is the range of distances over which the resonance absorption condition is maintained (1 Å), and \([M]\) is the rare gas concentration. Note that \( R_0 \) and \( \Delta R \) are implicitly dependent on the bandwidth of the excitation source. These expressions come from quasi-static approximations for the collision pairs as done by [Hedges, 1972] and [Eden,1976]. The factor of 2 difference in Eq.2 arises because collisions between Cs(6^2P_{3/2}) and Ar sample both the B^2Σ^+_{1/2} and A^2Π_{1/2} potential energy curves [Palla, 2010a]. At the peak of the blue satellite (4.5 Å), the CsAr(X^2Σ^+_{1/2}) potential is very slightly repulsive, \( \Delta E_{10} = 10 \text{ cm}^{-1} \) [Merritt, 2009], and the CsAr(B^2Σ^+_{1/2}) potential is entirely repulsive, such that \( \Delta E_{23} = 249 \text{ cm}^{-1} \) at \( R_0 = 4.5 \text{ Å} \).

The sum of the Cs containing species must obey conservation, therefore

\[
N_T = N_0 + N_1 + N_2 + N_3 \tag{E.3}
\]

Under lasing conditions the threshold gain \( g_{th} \) is defined by the gain-equals-loss relation

\[
g_{th} = -\frac{1}{2L_g} \ln(r_2) = \sigma_30 \left[ N_3 - 2N_0 \right] \tag{E.4}
\]

where \( L_g \) is the gain length, \( r_2 \) are the mirror reflectivities, and the factor of 2 multiplier to \( N_0 \) is a result of the degeneracy ratio \( g_2/g_0 \) between the upper and lower states of the four-level system. The system of equations E.1 – E.4 can be solved to find the number densities of the species in the CW threshold lasing case to be:

\[
N_0 = \frac{N_T - (1 + f_{23}) g_{th} \sigma_30}{3 + f_{10} + 2 f_{23}} \tag{E.5}
\]

\[
N_3 = \frac{g_{th} \sigma_30}{\sigma_30} + 2N_0 \tag{E.6}
\]

\[
N_1 = f_{10} N_0 \tag{E.7}
\]

\[
N_2 = f_{23} N_3 \tag{E.8}
\]
At conditions for optical transparency (OT), the gain is zero and therefore Eqs. E.5 and E.6 reduce to,

\[ N_{0,OT} = \frac{N_f}{3 + f_{10} + 2f_{23}} \] \hspace{1cm} E.9

\[ N_{3,OT} = 2N_{0,OT} \] \hspace{1cm} E.10

An analytical rate equation approach can be used to better understand the expected laser output as a function of pump input, see Carroll and Verdeyen [Carroll, 2012] for details. These rate equations can be solved for various parameters and laser performance predictions. The cross-section for the pump beam \( \sigma_p \) is determined via knowledge of the absorption \( \alpha \) and the reduced absorption coefficient \( k \):

\[ \alpha = k[N_0][M] = \sigma_p[N_1] \quad \Rightarrow \quad \sigma_p = k\frac{N_0}{N_1}[M] = k\frac{M}{f_{10}} \] \hspace{1cm} E.11

It is interesting to note that \( \sigma_p \) becomes a simple function of the rare gas concentration \( M \), the reduced absorption coefficient \( k \), and the ratio of \( N_f \) to \( N_p \). Chen and Phelps [Chen, 1973] established a solid value of the reduced absorption coefficient of \( 1.3 \times 10^{-38} \) cm\(^2\) at 837 nm for CsAr and this was confirmed by measurements using modern instrumentation by Readle [Readle, 2010c]. For a Cs-Ar gas mixture with 500 Torr of Ar at 200°C, \( \sigma_p = 6.5 \times 10^{-15} \) cm\(^2\).

The pump intensity for optical transparency (OT, gain=0) inside the gain volume:

\[ I_{p,OT} = \frac{2\hbar \nu_p}{\sigma_p \tau_{30}(f_{10} - 2f_{23})}, \quad I_{p,OT} = \frac{2\hbar \nu_p}{kM \tau_{30}} \left( \frac{f_{10}}{f_{10} - 2f_{23}} \right) \] \hspace{1cm} E.12

For the lasing case we combine terms to simplify the expressions by letting:

\[ P = \frac{\sigma_p I_p}{\hbar \nu_p} \tau_{30} \quad \text{and} \quad L = \frac{\sigma_L I_L}{\hbar \nu_L} \tau_{30} \] \hspace{1cm} E.13

where \( P \) is a non-dimensional “pumping” term, \( L \) is a non-dimensional “lasing” term, \( \sigma_p \) is the cross-section for the pump beam, \( I_p \) is the intensity of the pump beam in the cell volume, \( \nu_p \) is the frequency of the pump beam, \( \sigma_L \) is the cross-section for the lasing beam, \( I_L \) is the intensity of the lasing beam in the cell volume, \( \nu_L \) is the frequency of the laser, and \( \tau_{30} \) is the spontaneous emission lifetime of the upper state \( N_3 \).

Computing the gain gives

\[ g = \sigma_L (N_3 - 2N_0) = \sigma_L N_0 \left[ \frac{P(f_{10} - 2f_{23}) - 2}{Pf_{23} + L + 1} \right] \] \hspace{1cm} E.14

Note that setting \( L \) and \( g \) equal to zero for optical transparency results in the recovery of E.12 above for \( I_{p,OT} \). As the lasing term \( L \to 0 \), we have the small signal gain regime and \( g \to g_0 \), thus we can now define the small signal gain coefficient for a saturation law to be

\[ g_0 = \sigma_L N_0 \left[ \frac{P(f_{10} - 2f_{23}) - 2}{Pf_{23} + 1} \right] \] \hspace{1cm} E.15

The corresponding gain saturation law is then

\[ g = g_0 \left( \frac{1 + \frac{L}{Pf_{23} + 1}}{Pf_{23} + 1} \right) \] \hspace{1cm} E.16

When \( g \to g_0 / 2 \) we define the saturation parameter \( L_{sat} \) and the saturation intensity \( I_{L,sat} \) to be

\[ L_{sat} = Pf_{23} + 1 \] \hspace{1cm} E.17

\[ I_{L,sat} = \frac{\hbar \nu_L}{\sigma_L \tau_{30}} \left( 1 + \frac{\sigma_p \tau_{30}}{\hbar \nu_p} f_{23} I_p \right) \] \hspace{1cm} E.18

Analogous to the classic Rigrod derivation [Rigrod, 1965], \( I/I_{L,sat} = L/L_{sat} = (L^+ + L)/L_{sat} = \beta^+ + \beta^- \), and one can utilize Rigrod’s formulation to obtain a first order model of laser output power. Because of the presence of
intracavity optical elements in the XPAL systems to date (windows on the alkali cell and a polarizing beam splitter for the pump beam), we will implement a modified version of Rigrod’s formulation that includes intracavity optical losses (which can be significant) [Carroll and Verdeyen, 2009]. (Note that Carroll and Verdeyen originally considered small intracavity diffraction losses, but the formulation is appropriate for any discrete intracavity optical elements that create losses inside the resonator.)

\[ I_{out} = \frac{(1-\delta_1)t_1\sqrt{(1-\delta_2)r_2} + (1-\delta_2)r_2\sqrt{(1-\delta_1)t_1}}{(1-\delta_1)\eta_1 + (1-\delta_2)\eta_2} \left[ g_o L_g + \ln(1-\delta_1)(1-\delta_2) \eta_2 r_2 \right] \quad E.19 \]

where \( L_g \) is the gain length, \( r_i \) are the mirror reflectivities, \( t_i \) are the mirror transmissivities, \( \delta_i \) are the loss terms from intracavity elements on either side of the gain medium, and \( I_{out} \) is the total outcoupled intensity. Note that Rigrod’s original formulation only included mirror absorption losses, but not losses from intracavity optical elements. Also observe that a simple substitution of \( R_i = (1-\delta_i) r_i \) results in the recovery of Rigrod’s expressions.

A critical realization that must be included into the model is to recognize that the pump beam will be absorbed as it traverses through the gain medium and hence reduce the pump intensity. The reduction in pump intensity becomes progressively more pronounced with higher temperature and/or longer gain length and ultimately leads to an optically thick gain medium that cannot be lased for reasonable power inputs. To treat this aspect of XPAL, let us consider a two-pass pump beam through the gain medium, and make the simplifying assumption that the integrated average pump intensity through the gain medium will be representative of the true intracavity intensity for the purpose of computing the laser performance. Therefore, \( E.13 \) becomes

\[ \overline{P} = \frac{\sigma_p \overline{I}_p}{h\nu_p} \tau_{s0} \quad E.20 \]

and we must use these integrated numbers in equations \( E.14 - E.19 \). Figure 15 illustrates the geometry for computing the average pump intensity.

The integrated average intracavity pump intensity is computed by Carroll and Verdeyen [Carroll, 2012] to be

\[ \overline{I}_p = I_{p,inp} \eta_{del} t_{w1} \left[ \frac{1}{\sigma_p (N_1-N_2) L_g} \right] \left[ 1 - e^{-\sigma_p (N_1-N_2) L_g} \right] \left[ 1 + t_{w2}^2 e^{-\sigma_p (N_1-N_2) L_g} \right] \quad E.21 \]

where \( \eta_{del} \) is the fraction of the input beam that is delivered to the front face of the alkali cell via the beam train of optics, the \( t_{w} \) are the alkali cell window transmissivities, and \( I_{p,inp} \) is the pump intensity input into the optical system.

### 3.3.2 XPAL Predictions from CW Theoretical Model for CW Performance

One of the critical questions for XPAL is whether or not it can be scaled to have good optical-to-optical efficiency. Further, one of the known challenges for XPAL will be to properly absorb the broadband pump radiation on the \( N_{x} \leftrightarrow N_{1} \) (\( B \leftrightarrow X \)) transition; two ways of accomplishing this are to increase the absorption per cm via an increase in temperature (thereby raising the alkali number density) or an increase in the rare gas number density, or to increase the gain length. If we consider a double-pass system with excellent optical elements having low losses (for example, a high quality polarizing beam splitter and Brewster angle cell windows that are anti-reflection coated), then the values for the \( \delta_i \) loss terms are small (\( \delta_i \) of 0.059 and 0.02 were used in these calculations). Figures 16 and 17 show predicted CW performance of a 4-level XPAL system as a function of temperature and pump intensity with a 10 cm Cs-Ar cell having \( 2.5 \times 10^{19} \) cm\(^{-3} \) of Ar. The model predicts that an optical-to-optical efficiency in the range of 40-50% can be achieved for XPAL. Figure 17 shows that there is an optimum temperature for the available pump intensity, and that in general higher pump intensity should lead to higher XPAL performance.
An alternative to raising cell temperature that also accomplishes the goal of increasing absorption of the pump is to increase the gain length of the alkali cell. Figure 18 shows the predicted CW performance of a 4-level XPAL system as a function of alkali cell length and pump intensity for a 473 K cell temperature having $2.5 \times 10^{19}$ cm$^{-3}$ of Ar. The model predicts that an optical-to-optical efficiency in the range of 40-50% can be achieved for XPAL. Figure 18 shows that there is an optimum cell length for the available pump intensity, and that in general higher pump intensity should lead to higher XPAL performance.

It is clear that such high pump intensities are not desired, and very hard (if not impossible) to achieve in CW systems. However, there are some options to help alleviate the high pump intensity:

1) Some alternate alkali and rare gas mixtures have better blue wing absorption characteristics than Cs-Ar [Readle, 2010b; Carroll, 2012].

2) The intensity required to reach optical transparency is inversely proportional to rare gas pressure, $E_{12}$, therefore higher rare gas pressure could be utilized to increase absorption and decrease intensity.

3) Multi-pass optical arrangements can be used to increase the integrated average intracavity intensity that drives the gain medium.

Through a combination of the above options, one may be able to push the pump intensities significantly lower.

IV. Optically Pumped Rare Gas Lasers

Lasers produced from excited rare gas species have been around for decades, but these have all been the result of some form of electric discharge or e-beam excitation with wavelengths typically in the visible (He-Ne, or He-Cd) or in the ultraviolet (various excimer lasers). A more recent development has been the optical pumping of metastable rare gas species to enable lasing on lines in the infrared, Fig. 19. In this configuration, an electric discharge is used
to create the rare gas metastable states s[3/2]2, followed by laser pumping to the p[5/2]1 state, relaxation to the p[1/2]1 state and subsequent lasing back to the metastable state. The system is analogous to the alkali metal laser systems such as DPAL. Studies of the relaxation rates in different rare gas mixtures established critical fundamental collisional rates, and that He and Ne were suitable collisional partners for Kr [Kabir and Heaven, 2012].

The recent demonstration of this system in which Ar*, Kr*, and Xe* were all lased [Han and Heaven, 2012] has intriguing possibilities because these all-rare gas systems eliminate many of the standard logistic problems faced by other chemical and gas laser systems. Further, the system operates at room temperature and pressures on the order of 1 atm. Electrical efficiency still needs to be improved for scaling purposes, but given that this laser is in its infancy, it seems likely that significant improvements will occur over the coming several years.

V. Concluding Remarks

Over the past decade, several advanced gas laser systems have been demonstrated that are revolutionizing this area, ElectricOIL, DPAL, XPAL, and the optically pumped metastable rare gas laser. These new systems take advantage of recent advancements in the implementation of different ways to electrically excite the gases to states that can be lased. In particular, the utilization of advances in electric gas discharge technology (as in the case of ElectricOIL), or the implementation of high efficiency laser diodes that utilize an alkali vapor to convert the relatively broadband diode radiation into a narrow linewidth beam (as in the case of DPAL and XPAL), present an attractive hybrid mix of laser technologies.

Systematic development of the electric oxygen-iodine laser has resulted in a peak total out-coupled power of 538 W. These results presently indicate superlinear enhancement of ElectricOIL power output with the product of gain and gain length g0L. The improvement in performance with increased g0L seems to indicate that the unidentified mechanism found in previous investigations which competes with the standard pumping reaction becomes less significant as g0L is increased, leading to more efficient power extraction. This interesting result, along with progress in development of various system aspects such as O2(a) production, heat exchanger performance, flow injection design, the application of iodine pre-dissociator discharge, and resonator design should enable continued enhancement of ElectricOIL performance. The recent demonstrations showing superlinear scaling with g0L indicate that a fully scaled ElectricOIL system is viable with good electrical efficiency.

DPAL appears to be a very intriguing system, but based upon experimental issues at CW power levels above 50 W there are questions about how well the system will scale, i.e., the electrical-to-optical efficiency has been decreasing with power for the static cell CW systems. The current DPAL theories that are baselined to lower power data suggest excellent scaling, but there are likely some missing factors (possibly photo-ionization at high intensity) since the static cell experiments are not scaling linearly at higher average powers. Recent experiments with a flowing DPAL show considerable promise for scaling to larger systems.

While initial pulsed XPAL experiments and modeling look intriguing, quasi-CW and CW experiments need to be performed to prove out the viability of XPAL as a scalable system. The largest concern for XPAL is that the absorption cross-section is small enough to require very high pump intensities to drive the system; it may be possible to accommodate this through the best choice of the alkali-rare gas mixture, and optimized pump and gain volume design. Early modeling indicates that XPAL is a more efficient large system than a small system [Palla, 2010]. A preliminary CW XPAL theory was developed and predicts that an optical-to-optical efficiency in the range of 40-50% can be achieved for XPAL, however high pump intensities (≈1x107 W-cm−2) appear to be required for high efficiency operation.
The recent demonstration of an optically pumped metastable rare gas laser in which Ar*, Kr*, and Xe* were all lased [Han and Heaven, 2012] has intriguing possibilities because these all rare gas systems eliminate many of the standard logistic problems faced by other chemical and gas laser systems.

Other all gas phase iodine lasers have been created [Henshaw, 2000; Masuda, 2010], but the chemistry involved (HN3 or NC1) makes these unlikely candidates for scaling. Interesting developments in DPAL systems recently produced blue laser beams through multi-photon absorption [Pitz, 2010], but it is unclear if this process can be made efficient enough to develop into a high energy system.

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