Development of an Electric Discharge Oxygen-Iodine Laser

DISSERTATION

Presented in Partial Fulfillment of the Requirements for the Degree Doctor of Philosophy in the Graduate School of The Ohio State University

By

John R. Bruzese, M.S.

Graduate Program in Mechanical Engineering

The Ohio State University

2011

Dissertation Committee:

Dr. Igor V. Adamovich, Advisor

Dr. Walter R. Lempert

Dr. J. William Rich

Dr. Mohammad Samimy
Abstract

An electrically excited oxygen-iodine laser with a scaled-up electric discharge has been designed and operated. Singlet delta oxygen is generated in a capacitively coupled RF discharge sustained in the plenum of a M=3 nozzle. The discharge operates at pressures up to \( p_0 = 120 \) torr, powers up to 4.5 kW, and flow rates up to 0.5 mole/sec of 0-15 % O\(_2\) in He. Singlet delta oxygen yields up to \( Y = 7.5 \) % have been measured using absolute infrared emission spectroscopy. Small signal gain on the \( I'(2P_{1/2},F' = 3) \rightarrow I(2P_{3/2},F'' = 4) \) iodine atom transition in the supersonic flow has been measured by tunable diode laser absorption spectroscopy, up to \( \gamma = 0.209 \) %/cm. The addition of NO to the laser mixture and injection of chilled helium to reduce the laser mixture temperature have been shown to enhance laser performance considerably. Other modifications, including the addition of argon to the laser mixture, chilling the main flow through the electric discharge, and dissociating iodine directly by electron impact in an auxiliary electric discharge have not resulted in further gain increase. Laser action has been achieved using a transverse resonator in the supersonic cavity, with laser output power up to \( P_L = 7.8 \) W.
Acknowledgements

I would like to thank everyone who has helped me reach this point in life and contributed to this research project:

My advisor, Dr. Igor Adamovich, for five years of understanding, guidance, and support.

Dr. Walter Lempert, for all his help and support, especially in the end.

The faculty and staff of the MAE Department for providing such a wonderful place to work and learn.

The Physics Department machinists, without whom I would have accomplished nearly nothing.

Fellow grad students, past and present, for helping with every aspect of my project, graduate school, and life.

The SMART fellowship program, for their support through graduate school and guidance in starting my career.

The Joint Technology Office (JTO) and Air Force Research Laboratory Directed Energy Directorate (AFRL/RD, formerly AFRL/DE) for providing project funding.

Dr. David Carroll and the rest of the UI/CU Aerospace research group for many useful discussions.

My family, especially my mother, for being supportive.

A list of friends too long to name. I certainly would not have survived without you.

Leo, for just being Leo.
Vita

2002..............................................................High School Diploma
   Dover High School
   Dover, OH

2006..............................................................B.S. Aeronautical and Astronautical Engineering
   The Ohio State University
   Columbus, OH

2008..............................................................M.S. Aeronautical and Astronautical Engineering
   The Ohio State University
   Columbus, OH

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Chapter 1: Introduction

1.1 The Airborne Laser

The United States Air Force has a standing interest in innovative new methods to intercept and destroy ballistic missiles and other high velocity airborne targets. Directed energy weapons have the potential to deliver a lethal strike at the speed of light – much faster and more accurately than could be achieved with conventional projectiles. The first major attempt to develop a high-power airborne laser for missile defense was the Airborne Laser Laboratory (ALL), shown in Figure 1.1. This test bed prototype featured a 500 kW CO$_2$ laser mounted on a modified Boeing KC-135 aircraft. The program culminated in 1983 with the destruction of several aerial target drones and AIM-9 Sidewinder air-to-air missiles.

![Airborne Laser Laboratory](image_url)

Figure 1.1: Airborne Laser Laboratory

1 Reproduced from reference (78)
These successful tests provided proof of concept that motivated the more ambitious Airborne Laser Program (ABL), launched in the early 1990s. A comprehensive review of the state of directed energy weapons at the inception of the ABL program can be found in reference (1). The ABL program endeavored to produce a fleet of high-power airborne lasers capable of destroying tactical ballistic missiles (and possibly intercontinental ballistic missiles in their boost stage) at ranges up to 600 km. The YAL-1A aircraft developed under this program, shown in Figure 1.2, is a modified Boeing 747-400 armed with a megawatt class chemical oxygen-iodine laser (COIL) fired out of a nose turret. Only one such aircraft has been produced, which recorded its first successful air-to-air destruction of a missile in February, 2010. Since then, the ABL has been downgraded to a ‘test bed’. Despite this setback to the ABL, interest in directed energy weapons, specifically COIL, remains. It is currently in development for other weapons programs independent of the ABL, including the Advanced Tactical Laser (ATL) program, which is developing a low altitude airborne laser for ground strikes.
1.2 The Chemical Oxygen-Iodine Laser

COIL is a gas laser that emits at $\lambda=1315$ nm and operates on a transition between the $I^*$(2P$_{1/2}$) and I(2P$_{3/2}$) spin-orbit split states of atomic iodine, further denoted simply as $I^*$ and I, respectively. The $I^*$ population inversion is pumped by metastable, electronically excited O$_2$(1$\Delta$) in a near resonant energy transfer reaction (see discussion below). The oxygen-iodine laser was first proposed in 1971$^2$ in the discussion of experiments where O$_2$(1$\Delta$) was generated in an electric discharge and energy transfer to $I^*$ was observed. The chemical oxygen-iodine laser was first demonstrated in 1978$^3$ and has since been developed to achieve powers in excess of a megawatt$^{4,5}$.

The important factors for evaluating a directed energy weapon are detailed in reference (1). The intention of a directed energy weapon is to heat the target surface sufficiently to cause failure. Surface heating on the target is directly proportional to fluence,

---

$^2$ Reproduced from reference (78)
\[ F = \frac{P_L \Delta t}{A} [J/m^2]. \tag{1} \]

where \( P_L \) is laser power, \( \Delta t \) is dwell time on target, and \( A \) is the beam area on target. The radius of the beam spot is the product of the range, \( R \), and the far field beam divergence, which (for a diffraction limited beam) is the wavelength, \( \lambda \), divided by the laser aperture diameter, \( D \). The beam area on target is

\[ A = \frac{\pi R^2 \lambda^2}{D^2}. \tag{2} \]

Substituting into Eq. (1) and rearranging terms, the expression for brightness is derived:

\[ B = \frac{FR^2}{\Delta t} = \frac{P_L D^2}{\pi \lambda^2} [W/sr]. \tag{3} \]

The expression on the left gives brightness in terms of parameters related to engaging a target, which determines the difficulty of destroying that target. The expression on the right gives brightness in terms of laser parameters, which determines the effectiveness of that laser as a directed energy weapon. The advantage of a smaller far field beam divergence due to shorter wavelengths is clear. Of the gas lasers shown to be scalable to high power, including HF (\( \lambda=2.6-3 \mu m \)), DF (\( \lambda=3.7-4 \mu m \)), CO (\( \lambda=5 \mu m \)), and CO\(_2\) (\( \lambda=9.4-10.6 \mu m \)) lasers, COIL has the most
advantageous wavelength of $\lambda=1315$ nm. COIL requires fifty times less power to accomplish the same mission as a CO$_2$ laser (used in the Airborne Laser Laboratory) due to its shorter wavelength. Additionally, COIL exhibits good beam quality at high power, is fiber optic compatible, and emits at a wavelength that avoids major atmospheric transmission losses due to Raleigh scattering and absorption, although it is somewhat limited by water vapor absorption. Long distance transmission of HF and CO lasers is limited by atmospheric water vapor absorption and long distance transmission of DF and CO$_2$ lasers is limited by atmospheric CO$_2$ absorption.

Detailed technical descriptions of the design and operation of COIL can be found in references (1; 3; 6). In COIL, singlet delta oxygen is generated in a chemical reaction summarized by the following equation:

$$\text{Cl}_2 + \text{H}_2\text{O}_2 + 2\text{KOH} \rightarrow \text{O}_2(1\Delta) + 2\text{KCl} + 2\text{H}_2\text{O}, \quad (4)$$

with $\text{O}_2(1\Delta)$ yields greater than 60%$^{(7)}$, where yield is defined as

$$Y = \frac{[\text{O}_2(1\Delta)]}{[\text{O}_2(1\Delta)] + [\text{O}_2(3\Sigma)]}. \quad (5)$$

The detailed two-phase reaction mechanism of $\text{O}_2(1\Delta)$ production is$^{(8)}$: 
with potassium hydroxide, hydrogen peroxide, and chlorine being the main reactants. Water vapor is removed from the flow of reaction products in a condenser and the remaining oxygen, often diluted in helium, is mixed with molecular iodine vapor.

Iodine is dissociated in a net reaction equation,
\begin{equation}
\text{nO}_2(\overset{1}{\Delta}) + \text{I}_2(\overset{1}{\Sigma}) \rightarrow \cdots \rightarrow \text{nO}_2(\overset{3}{\Sigma}) + 2\text{I},
\end{equation}

where \(n=4-6\). The energy of the singlet delta oxygen state is only \(\varepsilon=0.98\) eV, which is not enough to produce \(\text{I}_2\) dissociation (dissociation energy \(D=1.55\) eV) in a single collision. The kinetics of this reaction are discussed in references \(10; 11; 12; 6; 8\) but remain not fully understood. An energy diagram of the relevant excited states of oxygen and iodine is shown in Figure 1.3. One dissociation pathway begins with an energy pooling reaction,

\begin{equation}
2\text{O}_2(\overset{1}{\Delta}) \rightarrow \text{O}_2(\overset{3}{\Sigma}) + \text{O}_2(\overset{1}{\Sigma}),
\end{equation}

where the energy of \(\text{O}_2(\overset{1}{\Sigma})\) is \(\varepsilon=1.64\) eV and thus it can dissociate \(\text{I}_2\) in a single collision; however, the rate coefficient of the energy pooling reaction, \(k_{12}=2.7 \cdot 10^{-17}\) \(\text{cm}^3/\text{sec}\), is too slow to explain the rate of the iodine dissociation reaction detected in the experiment. A more likely pathway involves ground state \(\text{I}_2(\overset{1}{\Sigma})\) being excited to an intermediate state in an initial collision with \(\text{O}_2(\overset{1}{\Delta})\), then being dissociated in a second collision with \(\text{O}_2(\overset{1}{\Delta})\). The lowest electronically excited state of molecular iodine, \(\text{I}_2(A'^3\Pi_2)\), has an energy of \(\varepsilon=1.25\) eV, which is too high to be excited by \(\text{O}_2(\overset{1}{\Delta})\) in a single collision with ground state \(\text{I}_2(\overset{1}{\Sigma})\). Thus, the dissociation process involving intermediate excited states of \(\text{I}_2\) requires vibrational excitation of \(\text{I}_2\) in the ground electronic state,
\[ O_2 (^1\Delta) + I_2 (^1\Sigma) \rightarrow O_2 (^3\Sigma) + I_2 (^1\Sigma, v) \]  

(13)

with a rate constant \( k_{13} = 7.0 \cdot 10^{-15} \) cm\(^3\)/s\(^{\irsch\(8\))\). Vibrationally excited \( I_2 (^1\Sigma, v) \) can then be dissociated by a second collision with \( O_2 (^1\Delta) \),

\[ O_2 (^1\Delta) + I_2 (^1\Sigma, v) \rightarrow O_2 (^3\Sigma) + 2I (^2P_{3/2}). \]  

(14)

which occurs rapidly with a rate constant \( k_{14} = 3.0 \cdot 10^{-10} \) cm\(^3\)/s\(^{\irsch\(8\))\). Once a significant fraction of I atoms have been generated, the rate of vibrational excitation of \( I_2 (^1\Sigma) \) is increased significantly by the reaction,

\[ I^* (^2P_{1/2}) + I_2 (^1\Sigma) \rightarrow I (^2P_{3/2}) + I_2 (^1\Sigma, v). \]  

(15)

with a rate constant \( k_{14} = 3.8 \cdot 10^{-11} \) cm\(^3\)/s\(^{\irsch\(8\))\).
An excited atomic iodine population inversion is created in collisions of ground state I atoms with $O_2(^1\Delta)$,

$$O_2(^1\Delta) + I \leftrightarrow O_2(^3\Sigma) + I^*,$$  \hspace{1cm} (16)\]

with the forward temperature-dependent rate coefficient $k_{16}(T)=7.8 \cdot 10^{-11} \ (295/T) \ \text{cm}^3/\text{s} \ \text{}}{13}$. The forward reaction of Eq. (16) is slightly exothermic (e.g. see Figure 1.3), with energy defect of $\delta/k_B=402$ K, which shifts equilibrium to the products as the temperature is reduced. The reverse reaction has an energy barrier of $\delta/k_B=402$ K, such that its rate is reduced at lower temperatures. Equilibrium in the reaction of Eq. (5) is quickly established, where the concentrations of the species are related by the equilibrium constant,

\footnote{Reproduced from reference (8)}
\[
\frac{[O_2(^3\Sigma)][I^*]}{[O_2(^1\Delta)][I]} = K_{eq}(T) = 0.75 \exp \left(\frac{402}{T}\right). \tag{17}
\]

The \(O_2(^1\Delta)\) threshold yield, obtained from Eqs. (5, 17) and given as

\[
Y_{th} = \frac{1}{1 + 2K_{eq}(T)} \tag{18}
\]

is the minimum \(O_2(^1\Delta)\) yield required to produce an \(I^*\) population inversion, defined as \([I^*]/(0.5[I]) > 1\), when gain is observed on the \(I^* \rightarrow I\) transition. A factor of 0.5 (or 2) appears in the expression for the iodine atom population inversion due to the ratio of statistical weights (degeneracies) of \(I(^2P_{1/2})\) and \(I(^2P_{3/2})\) states, see discussion below. A plot of \([I^*]/(0.5[I])\) vs. temperature for different \(O_2(^1\Delta)\) yields is shown in Figure 1.4. Reducing the temperature from \(T=300\) K to \(T=160\) K reduces the threshold yield from \(Y_{th}=15\%\) to \(Y_{th}=5\%\), which reduces the minimum energy stored in \(O_2(^1\Delta)\) by a factor of 3 and increases the maximum possible efficiency of the laser. It is critical to have high yield and low temperature to achieve \(I^*\) population inversion and positive gain. The \(I^*\) population is controlled primarily by the forward and reverse rates of the reaction of Eq. (16) since the rates of spontaneous emission, \(A=7.7\text{ s}^{-1}\), and collisional quenching of \(I^*\) are typically slow and do not affect equilibrium significantly (see discussion in Section 1.3).
Figure 1.4: I atom population ratio vs. temperature for different yields

Laser action occurs due to stimulated emission on the $I^\ast \rightarrow I$ transition,

\[ I^\ast (^2P_{1/2}) + \nu \rightarrow I(^2P_{3/2}) + 2\nu. \]  \hspace{1cm} (19)

The upper laser state, $I^\ast (^2P_{1/2})$, is split by the nuclear spin-orbital angular momentum interaction (nuclear spin, $I=5/2$) into 2 hyperfine components and the lower laser state, $I(^2P_{3/2})$, is split into 4 hyperfine components. The hyperfine splitting yields a new quantum number, $F$, ranging from $I-S$ to $I+S$. The selection rule $\Delta F=0,\pm 1$ allows 6 magnetic-dipole radiative transitions between the upper and lower states, shown in Figure 1.5.
Small signal gain on each of the hyperfine structure transitions is

$$\gamma_{F',F''}(v) = \sigma_{F',F''}(v) \left( [I^*(F')] - \frac{g_{F'}}{g_{F''}} [I(F'')] \right), \quad (20)$$

where

$$\sigma_{F',F''}(v) = A_{F',F''} \frac{c^2}{8\pi\nu_o^2} \ g(v - \nu_{F',F''}) \quad (21)$$

is the absorption/stimulated emission cross section, $g_{F'}$ is the degeneracy of the F' hyperfine component of the upper state ($I'$), $g_{F''}$ is the degeneracy of the F'' hyperfine component of the lower state ($I$), $A_{F',F''}$ is the Einstein coefficient for

**Figure 1.5: Hyperfine splitting of atomic iodine and allowed radiative transitions**
spontaneous emission, $c$ is the speed of light in vacuum, $\nu_0=227.9$ THz is the emission frequency on the line center, and $g(\nu-\nu_{F',F''})$ is the lineshape function.

The lineshape function has contributions from Doppler (temperature) and collision (pressure) broadening. The Doppler lineshape function is given by the Gaussian function,

$$g_D(\nu - \nu_{F',F''}) = \sqrt{\frac{4 \ln(2)}{\pi}} \frac{1}{\Delta \nu_D} \exp \left\{ -4 \ln(2) \left( \frac{\nu - \nu_{F',F''}}{\Delta \nu_D} \right)^2 \right\}, \quad (22)$$

where

$$\Delta \nu_D = \sqrt{\frac{8 k_B T \ln(2)}{M c^2}} \nu_0 = 14.56 \sqrt{T} \text{[MHz]}, \quad (23)$$

is the Doppler lineshape full width at half maximum (FWHM), $k_B=1.38 \cdot 10^{-23}$ J/K is the Boltzmann constant, $M=126.9$ amu is the mass of an iodine atom, and $T$ is the temperature in Kelvin. The Lorentzian (collision broadened) lineshape function is given as follows,

$$g_C(\nu - \nu_{F',F''}) = \frac{\Delta \nu_C/(2\pi)}{(\Delta \nu_C/2)^2 + (\nu - \nu_{F',F''})^2}, \quad (24)$$

where,
\[ \Delta v_C = \sum_i \Delta_i p_i, \quad (25) \]

is the collision lineshape FWHM, \( \Delta_i \) is the collision broadening parameter, and \( p_i \) is the partial pressure of each constituent gas in the mixture. The collision broadening parameters, determined experimentally for the \( I'(F'=3) \rightarrow I(F''=4) \) transition \(^{(14)}\), are shown in Table 1.1.

<table>
<thead>
<tr>
<th>Gas</th>
<th>( \Delta ) [MHz/Torr]</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>( 3.6 \cdot (296/T)^{0.36} )</td>
</tr>
<tr>
<td>O(_2)</td>
<td>( 5.0 \cdot (296/T)^{0.70} )</td>
</tr>
<tr>
<td>N(_2)</td>
<td>5.5 at ( T=296 ) K</td>
</tr>
</tbody>
</table>

For limiting cases when \( \Delta v_D \gg \Delta v_C \) or \( \Delta v_D \ll \Delta v_C \), the lineshape function may be approximated by the Doppler or collisional lineshapes, respectively. For cases where \( \Delta v_D \sim \Delta v_C \), the lineshape is given by the Voigt function,

\[
g_V(v-v_{F,F''}) = \int_{-\infty}^{\infty} g_D(v'-v_{F,F''})g_C(v-v')dv'. \quad (26)\]

This convolution integral has no known analytical solution and must be solved numerically. For typical values of temperature and pressure in the experiments conducted in the present work, the Doppler broadening is \( \Delta v_D=145.6 \) MHz and the collision broadening is \( \Delta v_C=17.6 \) MHz. The Doppler and Voigt lineshapes are shown in Figure 1.6.
In equilibrium, the hyperfine components are populated statistically according to their degeneracies, $g_F = 2F + 1$. In this case, the population of each hyperfine component can be expressed as a fraction of the total population of the upper and lower states,

\begin{equation}
[I^*(F')] = \frac{g_F}{g_u} [I^*],
\end{equation}

(27)

\begin{equation}
[I(F'')] = \frac{g_F}{g_l} [I],
\end{equation}

(28)
where \( g_u = 12 \) is the total degeneracy of the upper (I') state, and \( g_l = 24 \) is the total degeneracy of the lower (I) state. Small signal gain on each hyperfine transition, given as,

\[
\gamma_{F',F''}(v) = \sigma_{F',F''}(v) \frac{g_{F'}}{g_u} \left( [I'] - \frac{g_l}{g_l} [I] \right) = \sigma_{F',F''}(v) \frac{g_{F'}}{12} \left( [I'] - \frac{1}{2} [I] \right),
\]

is proportional to the degeneracy of the hyperfine component of the upper state (\( g_{F'} \)) and the Einstein coefficient for spontaneous emission (\( A_{F',F''} \)). A complete theoretical hyperfine spectrum is given in reference \(^{(15)}\). The pertinent parameters are given in Table 1.2, where intensity and frequency shift are given relative to the strongest \( \text{I}'(F'=3) \rightarrow \text{I}(F''=4) \) transition.

<table>
<thead>
<tr>
<th>( \text{I}'(F') \rightarrow \text{I}(F'') )</th>
<th>( A_{F',F''} ) (sec(^{-1}))</th>
<th>( g_{u,F'} )</th>
<th>Intensity (%)</th>
<th>( \Delta \nu ) (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 \rightarrow 4</td>
<td>5.0</td>
<td>7</td>
<td>100</td>
<td>0</td>
</tr>
<tr>
<td>3 \rightarrow 3</td>
<td>2.1</td>
<td>7</td>
<td>43.2</td>
<td>4.23</td>
</tr>
<tr>
<td>3 \rightarrow 2</td>
<td>0.6</td>
<td>7</td>
<td>12.3</td>
<td>6.20</td>
</tr>
<tr>
<td>2 \rightarrow 3</td>
<td>2.4</td>
<td>5</td>
<td>34.7</td>
<td>-12.15</td>
</tr>
<tr>
<td>2 \rightarrow 2</td>
<td>3.0</td>
<td>5</td>
<td>43.2</td>
<td>-12.89</td>
</tr>
<tr>
<td>2 \rightarrow 1</td>
<td>2.3</td>
<td>5</td>
<td>33.3</td>
<td>-14.86</td>
</tr>
</tbody>
</table>

The laser resonator is typically placed downstream of a supersonic nozzle where the gas mixture has been expanded to a Mach number of \( M=2-3 \). The supersonic expansion is used to reduce temperature in the inviscid core flow according to the isentropic relation,
where T is the local static temperature, $T_0$ is the stagnation temperature, and $k$ is the specific heat ratio. As discussed above, the temperature reduction shifts the equilibrium in reaction of Eq. (5) toward the products, this favoring formation of I$^*$. The $O_2(^1\Delta)$ threshold yield, obtained from Eqs. (5,17), given as,

$$Y_{th} = \frac{1}{1 + 2K_{eq}(T)},$$

is the minimum $O_2(^1\Delta)$ yield required to produce an iodine atom population inversion when gain is observed on the I$^* \rightarrow$I transition. Again, reducing the temperature from $T=300$ K to $T=160$ K reduces the threshold yield from $Y_{th}=15\%$ to $Y_{th}=5\%$ (see Figure 1.4), which reduces the minimum energy stored in $O_2(^1\Delta)$ by a factor of 3 and increases the maximum possible efficiency of the laser. Reducing the temperature also increases gain due to the increased equilibrium concentration of I$^*$ (see Figure 1.4) and due to the temperature dependence of the Doppler lineshape function, which becomes narrower at low temperatures. This allows energy to be extracted in a more compact resonator.

**1.3 Electric Discharge Excited Oxygen-Iodine Laser (DOIL)**

Safety and logistical concerns about the aggressive reactants used in COIL to generate $O_2(^1\Delta)$, mainly concentrated hydrogen peroxide and chlorine gas, have
prompted a search for alternative methods of generating $\text{O}_2(^1\Delta)$ to power an oxygen-iodine laser. Attention has recently shifted back to the original concept of an electric discharge excited oxygen-iodine laser, where $\text{O}_2(^1\Delta)$ is generated by electron impact in an electric discharge, without the need of hazardous chemical fuels. Although development of a DOIL was attempted in the 1970s and 80s$^{(16; 17; 18)}$, it has not been successfully demonstrated until 2005$^{(19)}$. An electric discharge excited oxygen-iodine laser could also potentially be operated in a closed cycle, where the laser gas mixture is flowing in a closed loop through the laser. This process eliminates the need for refilling and would allow the laser to be used multiple times while deployed at a remote location, requiring only a source of electrical power.

As is well known, kinetics of electric discharge plasmas are controlled primarily by the reduced electric field, $E/N$, where $E$ is the electric field and $N$ is the number density of particles in the plasma. It can be shown that electron temperature (or average electron energy) is approximately proportional to $E/N$.$^{(20)}$ Two commonly used units for the reduced electric field are $10^{-16} \text{ V} \cdot \text{cm}^2$ and $1 \text{Td}=10^{-17} \text{ V} \cdot \text{cm}^2$. The reduced electric field in the plasma, along with the electron energy-dependent cross sections of elastic and inelastic processes occurring in collisions of electrons and heavy species (electron impact collisions) determine the electron energy distribution function (EEDF). Electron number density is determined by a balance of ionization and electron losses due to electron-ion recombination, electron attachment (in electronegative plasmas), and ambipolar
diffusion with subsequent recombination on the walls. The electron impact cross sections and EEDF determine the rate coefficients of electron impact processes in the plasma. Both EEDF and the rates of electron impact processes can be predicted by solving the Boltzmann equation for plasma electrons, in which the cross sections and the reduced electric field are the input parameters \(^{(20)}\). Solution of the Boltzmann equation also predicts discharge input energy partition among excitation of various energy levels of molecules and atoms (rotational, vibrational, and electronic), as well as ionization and electron attachment. The Boltzmann equation in oxygen and oxygen-helium mixtures, typically used in electrically excited oxygen-iodine lasers, has been solved in Refs \((21; 22)\). Figure 1.7 plots cross sections of the most important electron impact processes in oxygen. It can be seen that the \(O_2(^1\Delta)\) excitation cross section has a low energy threshold, approximately 1 eV. Figure 1.8 shows discharge input energy fractions into excitation of various energy levels of \(O_2\), as well as dissociation, ionization, and dissociative attachment. From Figure 1.8, one can see that discharge energy fraction spent on excitation of \(O_2(^1\Delta)\) exceeds 40 % and peaks approximately at \(E/N=10\) Td. In helium-oxygen mixtures, discharge energy fraction into \(O_2(^1\Delta)\) excitation is significantly reduced and peaks at lower reduced electric field values, \(E/N=4-5\) Td (see Figure 1.9). These results demonstrate that an electrically excited oxygen-iodine laser needs to operate at relatively low \(E/N\) values.
Figure 1.7: Energy-dependant electron impact cross sections for $\text{O}_2(^{1}\Sigma)$. Cross sections: 1, momentum transfer; 2, excitation of effective vibrational level; 3, excitation of $\text{O}_2(^{1}\Delta)$; 4, excitation of $\text{O}_2(^{1}\Sigma)$; 5, attachment; 6, excitation of effective electronic level $\text{O}_2^{*}$ with threshold 4.5 eV; 7, dissociation into $\text{O}(^{3}\text{P})$ atoms; 8 dissociation into $\text{O}(^{3}\text{P})$ and $\text{O}(^{1}\text{D})$ atoms; 9 ionization; 10, ionization with dissociation; 11, excitation of molecular rotation.\(^4\)

\(^4\) Reproduced from reference (22)
Figure 1.8: Discharge input energy fractions. Processes: 1, elastic losses and rotational excitation; 2, vibration; 3, excitation of O$_2(^1\Delta)$; 4 excitation of O$_2(^1\Sigma)$; 5, attachment; 6, effective electronic level O$_2$ with threshold 4.5 eV; 7, dissociation into O($^3P$) atoms; 8, dissociation into O($^3P$) and O($^1D$) atoms; 9, ionization.$^5$

More advanced kinetic models, where the Boltzmann equation is coupled to equations for the number densities of O$_2$-He plasma species, including vibrationally and electronically excited oxygen molecules and atoms, excited helium atoms, as

$^5$ Reproduced from reference (22)
$^6$ Reproduced from reference (21)
well as electrons and ions, have been developed recently (23). Their further development to incorporate pulsed discharge waveforms and extension to two spatial dimensions (24; 25) made possible detailed kinetic modeling of low-temperature, nonequilibrium oxygen-helium plasmas and predict SDO yield in a wide range of discharge configurations and operating conditions.

The crucial technical development of DOIL was efficient generation of \( \text{O}_2(1\Delta) \) in electric discharge at yields sufficient to maintain iodine atom population inversion. In the discharge, \( \text{O}_2 \) is typically diluted in a noble gas, such as helium or argon, to mitigate the effect of discharge heating and to improve plasma stability. Since helium has high thermal conductivity and diffusion coefficient, adding it to the laser mixture helps dissipating arc filaments formed at high discharge pressures and power loadings.

The dominant process of SDO generation in the electric discharge plasma is by electron impact excitation of \( \text{O}_2 \) from the ground electronic state,

\[
\text{O}_2 \left( ^3\Sigma \right) + e^- \rightarrow \text{O}_2 \left( ^1\Delta \right) + e. \tag{32}
\]

As can be seen from Figure 1.8 and Figure 1.9, in oxygen-helium plasmas with low initial singlet delta oxygen mole fraction, up to 20-40 % of input discharge energy at \( E/N=5\text{-}10 \) Td is spent on excitation of \( \text{O}_2(1\Delta) \). As the \( \text{O}_2(1\Delta) \) increases, net discharge energy fraction into excitation of singlet delta oxygen decreases, mainly due to reverse process, de-exitation of \( \text{O}_2(1\Delta) \) in superelastic collisions with electrons,
Several attempts have been made to model $O_2(^1\Delta)$ generation in a discharge and the subsequent iodine chemistry \textsuperscript{(26; 27; 28; 29; 30; 31; 32; 33; 21; 34)} \textsuperscript{(5; 6)}; however, there is sufficient discrepancy between the predictions of the models to suggest that more effort in this area is needed to achieve predictive modeling of DOIL kinetics. Specifically, little is known about the electron impact cross sections for many excited species present in $O_2$-He-I\textsubscript{2} plasmas. It is beyond the scope of the current work to explore each of these models in detail; however, some of the model predictions can be used models to guide experimental research.

DOIL operation has several fundamental differences from COIL due to plasma chemical reactions, which result in formation of significant amounts of O atoms, ozone, as well as excited oxygen molecules and atoms,

\begin{equation}
O_2(^3\Sigma) + e^* \rightarrow 2O(^3P) + e
\end{equation}

\begin{equation}
O_2(^3\Sigma) + e^* \rightarrow O(^3P) + O(^1D) + e
\end{equation}

O atoms persist downstream of the discharge due to slow recombination times,
where the rate constants of the three-body recombination are $k_{36}=4.8\cdot10^{-33}$ cm$^6$/s$^5$ and $k_{37}=6.9\cdot10^{-34}$ cm$^6$/s$^5$ (equivalent second-order rate coefficients are on the order of $10^{-14}$ cm$^3$/sec or more at typical discharge pressure of 90 torr at room temperature).

The effect of O atoms on $O_2(^1\Delta)$ concentration is due to O atom recombination, which may increase SDO yield,

$$O + O + M \rightarrow O_2(^1\Delta) + M,$$  \hspace{1cm} (38)

where the rate constants are $k_{38}=2.4\cdot10^{-33}$ and $k_{39}=1.2\cdot10^{-33}$ cm$^6$/sec$^5$. In addition, O atoms can generate $O_2(^1\Delta)$ in surface reactions. Lee, Rawlins, and Davis reported that $O_2(^1\Delta)$ yields in a microwave discharge are increased by a factor of two from $Y=20-25\%$ to $Y=45\%$ when O atom recombination is catalyzed on an oxygen-iodine
film (35). Oxygen atoms can also play a beneficial role in dissociating molecular iodine by chain reactions,

$$O + I_2 \rightarrow IO + I,$$  \hspace{1cm} (40)

$$IO + O \rightarrow O_2 + I,$$ \hspace{1cm} (41)

with rate constants of $k_{40} = k_{41} = 1.4 \cdot 10^{-10} \text{ cm}^3/\text{sec}$ (36). On the other hand, O atoms also rapidly deactivate $I^*$,

$$I^* + O = I + O,$$ \hspace{1cm} (42)

with the rate constant of $k_{42} = 1.2 \cdot 10^{-11} \text{ cm}^3/\text{sec}$ (37), which has a significant detrimental effect on DOIL performance. A more complete list of reactions involving O atoms in oxygen-iodine plasmas can be found in references (5; 37; 38; 39).

It is well known that oxygen atom concentration in the discharge afterglow can be controlled by the addition of NO or NO$_2$ (nitric oxide titration),

$$O + NO + M \rightarrow NO_2^* + M,$$ \hspace{1cm} (43)
\[ O + NO_2 \rightarrow NO + O_2, \quad (44) \]

with \( k_{43}=1 \cdot 10^{-31} \text{ cm}^6/\text{sec} \) (38) and \( k_{44}=1 \cdot 10^{-11} \text{ cm}^3/\text{sec} \) (36). The addition of small amounts of NO to oxygen-helium mixtures has also been shown to increase \( O_2(\Sigma) \) yields \((40; 41)\) in RF discharges. This effect may be due to increased conductivity of the plasma, leading to a greater coupled energy at smaller reduced electric fields when NO is added due to the lower ionization energy of NO, \( \varepsilon_{\text{NO}}=9.6 \text{ eV} \), compared to the ionization energy of \( O_2 \) and He, \( \varepsilon_{O_2}=12.2 \text{ eV} \) and \( \varepsilon_{\text{He}}=24.6 \text{ eV} \), respectively. Modeling calculations have also shown increased \( O_2(\Sigma) \) yields and reduced O atom concentrations in discharges containing trace amounts of NO\((42; 28)\).

Metastable excited state \( O_2(\Sigma) \) is also generated in He-\( O_2 \) discharges. The electron impact cross section for the \( O_2(3\Sigma \rightarrow 1\Sigma) \) transition has energy threshold of \( \varepsilon=1.64 \text{ eV} \) and peaks at about the same electron energy as the \( O_2(3\Sigma \rightarrow 1\Delta) \) transition, albeit at a value about a factor of 5 smaller \((5)\). Singlet sigma oxygen dissociates iodine molecules in a single collision,

\[ O_2(\Sigma) + I_2 \rightarrow O_2(3\Sigma) + 2I, \quad (45) \]

with the rate coefficient \( k_{45}=3.5 \cdot 10^{-11} \text{ cm}^3/\text{s} \) \((10; 12)\). Singlet sigma oxygen is also quenched by iodine into a lower-energy singlet delta state,
with the rate coefficient $k_{46} \approx 2.4 \cdot 10^{-11}$ cm$^3$/s$^{(10; 12)}$.

Heat addition in the discharge may raise the gas temperature significantly, making thermal management more important. Low singlet delta oxygen yields achieved in electric discharge oxygen-iodine lasers$^{(21; 40)}$, typically far below the threshold yield of 17% at $T=300$ K (see Figure 1.10), which requires significant temperature reduction to achieve iodine atom population inversion. Even at $T=160$ K, when threshold yield is $Y_{th}=5\%$, a significant fraction of the energy stored in $O_2(^1\Delta)$ cannot be used for energy transfer to iodine and subsequently for lasing which is a major cause of inefficiency. Flow temperature in the discharge afterglow can be reduced using supersonic expansion, which also reduces density and pressure according to the isentropic relations,

$$
\left(\frac{T}{T_0}\right) = \left(\frac{p}{p_0}\right)^{\frac{k-1}{k}} = \left(\frac{\rho}{\rho_0}\right)^{k-1},
$$

(47)

where $p$ is the pressure and $\rho$ is the density. As can be seen from Eq. (46), in a 16.5% $O_2$-He mixture ($k=1.6$), reducing temperature by a factor of 4 also reduces pressure by a factor of 40 and density by a factor of 10. Because such low densities and pressures are undesirable, additional means of reducing temperature may be employed, such as using heat exchangers and injection of chilled gasses in the
subsonic section. Operating at low pressures would require a large vacuum system, and low density in the resonator would reduce laser power because gain is proportional to iodine atom density. Thus, operating the discharge at high pressures is critical to mitigate these difficulties.

![Graph showing the relationship between temperature and threshold oxygen singlet delta yield.](image)

**Figure 1.10: Threshold oxygen singlet delta yield vs. temperature**

Both small signal gain and laser output power are sensitive to I₂ flow rate. Indeed, increasing the I₂ flow rate leads to higher iodine atom number density in the gain medium, which may increase gain and laser output power. On the other hand, generating a higher I atom number density by reaction of Eq. (11) may come at a significant cost, in terms of O₂(\(^{1}\Delta\)) yield, since several O₂(\(^{1}\Delta\)) molecules are needed to dissociate one iodine molecule. This would reduce the amount of O₂(\(^{1}\Delta\)) remaining in the flow and available for energy transfer to iodine atoms, which may in turn reduce gain. Molecular iodine may also be dissociated directly by electron impact in an auxiliary discharge, prior to mixing with the primary discharge.
products, thus reducing the cost of dissociation. Figure 1.11 shows iodine population inversion, calculated from Eqs. (11, 16, 17) at a temperature of T=125 K and a yield of Y=10 %, vs. I$_2$/O$_2$(^1$\Delta$) flow rate ratio for different numbers of O$_2$(^1$\Delta$) molecules required to dissociate an I$_2$ atom ('dissociation cost), given by Eq. 11. Reducing O$_2$(^1$\Delta$) losses associated with I$_2$ dissociation has the potential to increase gain in a wide range of conditions and may significantly increase gain, in particular, at high I$_2$ flow rates.

![Figure 1.11: Iodine population inversion vs. I$_2$/O$_2$(^1$\Delta$) flow rate ratio calculated from Eqs. (11, 16, 17) for a temperature of T=125 K and a yield of Y=10 %](image)

1.4 Previous DOIL Development

Over the last few years, several different groups have contributed to electric discharge singlet oxygen generator (DSOG) development by implementing a variety of different discharges, including pulsed and continuous DC, microwave, radio frequency, electron beam, and pulser-sustainer (also referred to as controlled
avalanche) discharges. Ionin et al. at P. N. Lebedev Physics Institute in Moscow, Russia reported $O_2(^1\Delta)$ yield up to $Y=10.5\%$ in an electron beam sustained discharge in a $O_2:Ar:CO=1:1:0.05$ mixture at 30 torr \(^{43}\). Schmiedberger at the Academy of Sciences of the Czech Republic and Fujii at the Anan College of Technology in Japan reported $O_2(^1\Delta)$ yield up to $Y=32\%$ in an axial RF discharge in a $O_2:N_2:NO=200:20:10$ mixture at 0.43 torr \(^{44}\). At similar operating conditions, the discharge products were mixed with a chilled $Ar:NO_2$ flow and $I_2$ vapor in a subsonic resonator where $I^*$ emission was observed, but laser action did not occur \(^{44}\). Schmiedberger et al. have proposed a new DOIL concept based on “a fast mixing of Ar hybrid plasma jet of DC electric arc and RF discharge with a neutral oxygen stream” \(^{45}\); however, no results have been published. Rakhimov et al. at Moscow State University have contributed significantly in this area \(^{46; 47; 48; 49; 50; 51; 52; 53; 54; 55}\). Recently they have reported that $O_2(^1\Delta)$ yield may be increased from 9\% in a c.w. RF discharge to 12\% when the RF discharge is pulsed at 1 kHz in 10 torr of $O_2$ with 1-20\% NO added\(^{56}\). However, this extensive body of work also has not resulted in detecting positive gain in mixtures of oxygen excited in an electric discharge with iodine vapor. A detailed overview of all previous DOIL efforts can be found in references \(^{4; 5}\). Further discussion will be limited to an overview of work that resulted in development and operation of electrically excited oxygen lasers, which has been accomplished by only two groups: the collaborative effort of the University of Illinois and CU Aerospace, and the Non-Equilibrium Thermodynamics Laboratory (NETL) of The Ohio State University.
1.4.1 University of Illinois/CU Aerospace

Carroll and Solomon presented their concept of a DOIL (also called ElectriCOIL) as early as 2000 (57). Positive gain measured on the I'(F'=3)→I(F''=4) transition was first reported in 2004 (58; 40) and laser action was first reported in 2005 (19; 59). A schematic of the apparatus used in their work is shown in Figure 1.12. The group tested three different discharge configurations with similar results. First, a longitudinal (axial) capacitively coupled radio frequency (CCRF) discharge was operated in a 5 cm diameter quartz tube between two external, concentric ring electrodes located about 25 cm apart in the flow direction (58; 59; 19; 40). Second, an inductively coupled RF discharge was operated in the same flow tube using a 5-turn inductive coil wrapped around the tube, extending 6.4 cm along the tube in the flow direction (59). Finally, a pulser-sustainer discharge was tested by simultaneously operating the inductively coupled RF discharge described above while applying high voltage, 0.5 µs duration pulses at pulse repetition rate of 100 kHz to the ring electrodes outside the tube (59). Adding nitric oxides (NO or NO2) to the laser mixture was used to enhance O2(^1Δ) yield and mitigate the detrimental effect of O atom generation on small signal gain (see discussion in Section 1.3 above). Flow temperature was reduced by adding chilled diluents to the laser mixture flow, and subsequent expansion through a supersonic nozzle (Mach 2). The group reported maximum yields up to Y=17 %, gain up to γ=0.015 %/cm, and laser output power of P_L=520 mW using 1 inch diameter resonator mirrors (58; 59; 19; 40).
Laser performance continued to improve as further modifications were made to the apparatus. Operating a transverse CCRF discharge between contoured electrodes mounted externally on the top and bottom of the flow tube resulted in further gain increase up to $\gamma = 0.067 \%$/cm and laser output power up to $P_L = 4.5$ W, using 2 inch diameter resonator mirrors (60). Dissociation of I$_2$ in an auxiliary discharge prior to mixing with the primary discharge products increased gain up to $\gamma = 0.10 \%$/cm and output power up to $P_L = 6.2$ W (61). Operating two identical transverse CCRF discharges in smaller 1.9 cm diameter tubes arranged in parallel (a single 5 cm diameter tube had been used previously) resulted in significant increase in discharge power, pressure, and gas flow rate, which improved gain to $\gamma = 0.17 \%$/cm and laser output power to $P_L = 12.3$ W (62). Operation of a single discharge in a quartz channel with a rectangular cross section and the addition of a heat exchanger downstream of the discharge to reduce the flow temperature further improved gain to $\gamma = 0.26 \%$/cm and laser output power to $P_L = 54.8$ W (63). In addition to these performance-enhancing modifications, the feasibility of laser action in a

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7 Reproduced from reference (19)
subsonic flow (64) and in an air-helium mixture (65) was demonstrated with laser output powers of $P_L=540$ mW and $P_L=32$ mW, respectively. These modifications were not pursued further due to decreased performance. Kinetic modeling calculations were conducted using a modified version of the BLAZE gas laser code (42; 28; 29; 66; 67) in tandem with ongoing experiments.

The current E-COIL laser configuration developed by the University of Illinois/CU Aerospace group represents the state of the art in this field (68). In this laser, two separate transverse CCRF discharges are operated in 6 parallel discharge tubes with 1.9 cm outer diameter, producing $O_2(\Sigma \Delta)$ yield up to $Y=12.5\%$. The total flow rate through these tubes is 44 mmole/sec of $O_2$ diluted in 150 mmole/sec of He, with 0.23 mmole/sec of NO added to the flow, at a discharge pressure of $p_0=45$ torr. Downstream of the discharge the flow is mixed with separate flows of 0.3 mmole/sec $I_2$ in 46 mmole/sec He carrier and 312 mmole/sec of $N_2$ chilled to $T\approx100$ K. The mixture is expanded supersonically in an $M=2$ nozzle to a pressure of $p=4$ torr. Gain up to $\gamma=0.26 \%/cm$ has been measured in the supersonic cavity with a total gain path of 7.6 cm. Laser output power of up to $P_L=92$ W has been measured at an electrical efficiency of $\eta=2.5 \%$ in a simple, transverse resonator with two 4 inch diameter mirrors, where electrical efficiency is defined as the ratio of laser output power to input electrical power, $\eta=P_L/P_p$. Finally, laser output power up to $P_L=102.5$ W has been measured at an electrical efficiency of about 2% with four 2 inch diameter mirrors configured in a Z-pass resonator.
1.4.2 NETL of The Ohio State University

Electric discharge excited oxygen-iodine laser development at Ohio State began in 2003, with the first results in singlet delta oxygen reported in 2004 \(^{(69)}\). In this approach, single delta oxygen was generated in a transverse pulser-sustainer discharge with yields up to 4.4% in a flow of 10% \(O_2\) in He at a pressure of \(p_0=120\) torr. Ionization in the discharge was generated by 20-25 kV peak voltage, 10-20 ns duration pulses applied at a pulse repetition rate of 40 kHz to 5 x 5 cm electrodes located in the top and bottom walls of the rectangular flow channel with a 1 x 5 cm cross section. In addition, transverse DC discharge was operated at powers up to \(P_p=1.2\) kW and reduced electric fields of \(E/N=3-7\) Td between 1 x 5 cm electrodes located in the side walls.

Figure 1.13: Schematic of NETL electric discharge oxygen-iodine laser apparatus

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8 Reproduced from reference (21)
Laser action was first achieved in 2006\textsuperscript{(70)}. Singlet delta oxygen was generated in a similar transverse pulser-sustainer discharge with powers up to $P_p=1.5$ kW in a 15\% O\textsubscript{2}-He mixture at a pressure of $p_0=60$ torr, in a 2 x 5 cm cross section discharge flow channel. Downstream of the discharge, I\textsubscript{2} vapor in helium carrier was injected into the flow, which was subsequently expanded supersonically in a $M=3$ nozzle, reducing the temperature to $T=100$ K. Small signal gain up to $\gamma=0.022$ \%/cm and laser output power up to $P_L=0.28$ W were observed.

The addition of NO to the discharge resulted in removal of O atoms, in addition to increasing plasma conductivity, increasing the current, and reducing the voltage of the sustainer discharge\textsuperscript{(41)}. Gain up to $\gamma=0.049$ \%/cm and laser output power up to $P_L=1.24$ W were reported\textsuperscript{(41)}. The addition of an auxiliary discharge in the iodine delivery line effectively dissociated I\textsubscript{2} to dissociation fractions up to $\alpha=50$ \%, although gain improved only modestly, from $\gamma=0.07$ \%/cm with the auxiliary discharge off to $\gamma=0.08$ \%/cm with the auxiliary discharge on\textsuperscript{(71)}. Maximum laser output power up to $P_L=1.4$ W was measured at the best gain conditions.

Recently, a scaled up pulser-sustainer discharge section with a 2 x 10 cm flow cross section and a 10 cm length was designed and tested. In a pulser-DC sustainer discharge sustained in this channel, singlet delta yield up to $Y=3.7$ \% were reported at sustainer discharge powers of up to $P_p=2.7$ kW\textsuperscript{(72)}. At high DC sustainer voltages, the discharge extended downstream to the iodine injector block made of aluminum, limiting the discharge powers that could be tested. In preliminary experiments, gain
up to $\gamma=0.07\ \%$/cm was measured. Further experiments with a scale-up pulser-sustainer discharge were suspended due to the nanosecond pulse plasma generator malfunction. While the pulse generator was being repaired, further SDO yield and small signal gain measurements continued using a capacitively coupled transverse RF (CCRF) discharge sustained between the same electrodes that have been used for the nanosecond pulse discharge. As discussed in the following chapters of the present dissertation, SDO yields measured using these two types of discharges turned out to be fairly close. In addition, transverse RF discharge generates much less electromagnetic noise compared to the pulser-sustainer discharge, and thus does not affect electronics in optical diagnostics used. For these reasons, subsequent DOIL laser experiments reported in the present dissertation have been conducted using the CCRF discharge. The experimental setup used in the present work is described in Chapter 2. The results are discussed in Chapter 3. Finally, Chapter 4 provides a summary and conclusions.
Chapter 2: Experimental Setup

2.1 Experimental Apparatus

A schematic of the experimental apparatus is shown in Figure 2.1. The O₂-He-NO main flow is mixed from the contents of a bank of three He cylinders, a 50% O₂-He mixture cylinder, and a 5% NO-He cylinder in a 1 inch outer diameter delivery line about 5 meters upstream of the laser. Flows from the O₂-He and He cylinders are started by remote-controlled solenoid valves and flow from the NO-He cylinder is started with a manual quarter turn valve. The O₂ and He flow rates are determined from the choked flow equation (73),

\[ \dot{n} = A p_0 \sqrt{\frac{k}{M R_0 T_0}} \left( \frac{2}{k+1} \right)^{\frac{k+1}{k-1}}, \]  

where \( \dot{n} \) is the mole flow rate, \( A \) is the throat area of a supersonic nozzle downstream of the discharge section (see Figure 2.1), \( p_0 \) is the pressure in the discharge region (plenum pressure), \( M \) is the molecular weight, \( R_0 \) is the universal gas constant, \( T_0 \) is the temperature in the discharge section, and \( k \) is the specific heat ratio. Flow rates and mole fractions of the mixture components are determined by recording the plenum pressure increase due to each flow, using the following procedure. For example, at baseline conditions, a He flow at plenum pressure of
$p_0 = 42$ torr is produced first, and, then a 50% O$_2$-He flow is added until the plenum pressure reaches $p_0 = 60$ torr. Using Eq. (48), the mole fraction of oxygen in the mixture, $x_{O_2}$, is calculated from the plenum pressure before and after adding the O$_2$-He mixture using the following equations,

$$p_{0\text{He}} \sqrt{\frac{k_{\text{He}}}{M_{\text{He}} R_0 T_0} \left( \frac{2}{k_{\text{He}} + 1} \right)^{\frac{k_{\text{He}} + 1}{k_{\text{He}} - 1}}} =$$

$$= (1 - 2x_{O_2}) p_{0\text{mix}} \sqrt{\frac{k_{\text{mix}}}{M_{\text{mix}} R_0 T_0} \left( \frac{2}{k_{\text{mix}} + 1} \right)^{\frac{k_{\text{mix}} + 1}{k_{\text{mix}} - 1}}}$$ (49)

where

$$M_{\text{mix}} = (1 - x_{O_2}) M_{\text{He}} + x_{O_2} M_{O_2}$$ (50)

and

$$k_{\text{mix}} = \frac{(1 - x_{O_2}) C_{p\text{He}} + x_{O_2} C_{pO_2}}{(1 - x_{O_2}) C_{v\text{He}} + x_{O_2} C_{vO_2}}$$ (51)

Utilizing the assumption that $k_{\text{mix}} \approx k_{\text{He}}$ for small oxygen fractions ($x_{O_2} \ll 1$), Eq. (49) may be simplified to
Substituting $M_{He} = 4$ and $M_{O_2} = 32$ and rearranging terms, the oxygen mole fraction is:

$$x_{O_2} = \frac{4 + 7 \left( \frac{p_{He}}{p_{mix}} \right)^2 - \sqrt{49 \left( \frac{p_{He}}{p_{mix}} \right)^4 + 72 \left( \frac{p_{He}}{p_{mix}} \right)^2}}{8}.$$  

Solving Eq. (52) for $p_{He}=42$ torr and $p_{mix}=60$ torr gives $x_{O_2} = 7.1\%$. Finally, the NO flow rate through the discharge section is determined using an Omega FMA-A2323-SS mass flow meter.

![Figure 2.1: DOIL Schematic](image)

An optional main flow chiller, which consists of 8 m of 3/8 inch outer diameter copper tubing coiled and immersed in a cold liquid bath, can be installed upstream of the discharge section inlet. A main flow chiller schematic and photograph are shown in Figure 2.2. The cold bath can use liquid nitrogen ($T=77$ K).
or a liquid nitrogen-ethanol ‘slush’ (T=157 K)\(^{(74)}\). The actual cooling bath temperatures were not measured. The liquid nitrogen-ethanol ‘slush’ was created by filling the bath alternately with small amounts of ethanol and liquid nitrogen while stirring the mixture by hand. The main flow chiller was removed when not in use.

Prior to entering the discharge section, the flow passes through an expansion section with a 30° half angle expansion angle to a 2 x 10 cm rectangular flow cross section, shown in Figure 2.3. The expansion section is followed by a flow conditioning section, shown in Figure 2.4, which improves flow uniformity and removes separation regions that would otherwise occur on the walls of the 30° expansion section. Presence of such separation zones was detected using tufts installed in the flow and detecting flow direction. The flow conditioning section consists of a 2.5 cm long, 3 mm cell diameter honeycomb followed by two identical wire mesh screens with 1 mm mesh size. Each component is separated by about 5 cm in the flow direction.

Figure 2.2: Main flow chiller

Figure 2.3

Figure 2.4
The flow conditioning section is followed by an optional upstream iodine vapor injector, shown in Figure 2.5 (‘upstream’ refers to its location upstream of the discharge section). This injector, made of acrylic plastic, has a 2 x 10 cm inlet and exit cross section and a 5 cm length in the streamwise direction. Twenty-eight injection ports with a 1.3 mm diameter are evenly distributed on both the top and bottom walls across the 10 cm width of the flow channel. When in use, a flow of iodine vapor in helium carrier flow, produced in the iodine vapor generation system, described in Section 2.1 below, is delivered to the injector through a heated ¼ inch outer diameter delivery line. The upstream iodine injector was installed to explore the effect of iodine dissociation in the primary electric discharge and remained in place for all subsequent measurements.
The flow then enters two identical, consecutive discharge sections, shown in Figure 2.6, each with a total length of 25.4 cm. A transverse CCRF discharge is operated between 10 x 10 cm electrodes, flush mounted in the top and bottom walls. The electrodes are separated by the 2 cm flow channel height, forming a 200 cm³ discharge region. In each discharge section, the electrodes are separated from the flow by 1.6 mm thick alumina ceramic plates, which prevent secondary emission and increase discharge stability and uniformity. The side walls in the discharge region are protected by flush mounted Teflon inserts and all flow channel walls downstream of the discharge region are covered by alumina ceramic plates. Gaps between the electrodes and the ceramic plates are filled with self-hardening silicone rubber to prevent corona discharge formation in the air pockets. A pressure tap is located 5 cm upstream of the first discharge region to determine pressure in the discharge sections.
Figure 2.6: Primary discharge section
The electrodes are powered by a Dressler 13.56 MHz, 5 kW RF plasma generator with an automatic impedance matching network. High voltage is connected to the top electrodes and the bottom electrodes are grounded, as shown in Figure 2.7. An additional variable inductor, adjusted manually between the runs, is connected in series with the load, between the impedance matching network and the electrodes in the top walls of the discharge sections to improve load impedance matching. Since this inductor is placed outside the grounded matching network case, it may act as an antenna and radiate a fraction of the incident RF power, thereby reducing the power coupled to the plasma. The two discharge sections may be operated individually or simultaneously. During simultaneous operation, the electrode pairs are connected in parallel with each other and powered by the same RF plasma generator. Total coupled RF power for both discharges was measured, but power distribution between the two discharges was not determined. An electrical circuit schematic for simultaneous operation of both discharges is shown in Figure 2.7.

Figure 2.7: Electrical diagram of simultaneous primary discharge operation
Downstream of the second discharge section, an optional 5 cm long diagnostic section can be placed. A 1 cm diameter aperture with a 2.54 cm thick BK-7 glass window, flush mounted with the inside wall surface, is located in the side wall to allow optical access for emission spectroscopy measurements. A concentric 4.6 cm diameter circular recess, 1 mm deep has been machined in the outer wall to match the outer diameter of a fiber optic link collimator to simplify its alignment. The diagnostic section is removable but remained in place for all present measurements.

![Window and flow diagram](image)

**Figure 2.8: Diagnostic section (side view)**

Molecular iodine vapor in helium carrier is injected downstream of the diagnostic section through one of two interchangeable iodine injectors, as shown in Figure 2.9. Schematics of both iodine injectors are shown in Figure 2.9. Injector 1 is an aluminum block with a 2 x 10 cm inlet cross section, a 0.6 x 10 cm exit cross section, and a 5 cm length in the streamwise direction. The flow cross section in the injector is reduced to increase the flow velocity and enhance mixing of the injection flow and the main flow. The flow Mach number at the injector exit is $M \approx 0.3$, based on the exit cross section to nozzle throat area ratio. Twenty-eight injection ports with a 1.3 mm diameter are evenly distributed on both the top and bottom walls.
across the 10 cm width of the flow channel. Injector 2 is made of acrylic plastic with inlet and exit cross section identical to Injector 1 and a 7.5 cm streamwise length. A pair of 3 mm diameter brass rods insulated from the flow by 4.75 mm outer diameter alumina tubes span the 10 cm width of the flow channel in each of the top and bottom walls of injector 2, as shown in Figure 2.9. These rods serve as electrodes for an auxiliary discharge to dissociate I\textsubscript{2} directly by electron impact. The rods are spaced 1 cm center-to-center in the streamwise direction and are recessed 4.5 mm from the top/bottom flow channel wall. Injection occurs through a 1.6 mm thick alumina ceramic plate with twenty-eight evenly spaced, 1.3 mm diameter injection ports, similar to Injector 1. This plate can be recessed from the top/bottom flow channel wall (Injector 2a) or flush mounted with the top/bottom injector wall (Injector 2b). At typical operating conditions, the injection ports are choked and the pressure in the iodine injector plenum is $p=500$-$1000$ torr, much higher than the main flow pressure of $p=40$-$120$ torr. The high pressure in the injector plenum makes producing breakdown and sustaining a discharge more difficult and decreases the reduced electric field (E/N) in the discharge, thus reducing dissociation efficiency. When Injector 2 is configured as Injector 2a, there is a 1.6 cm long (in the streamwise direction) and 0.76 cm deep recess in each of the top and bottom walls, as shown in Figure 2.9. The electrodes in Injector 2 can be powered by an ENI 13.56 MHz 600 W RF power supply with a MFJ-3000 manual impedance matching network or by an 11 kV, 100 ns pulse width high voltage power supply operating at a pulse repetition rate up to 10 MHz. In both cases, the discharge is
operated between the two electrodes in the same recess (i.e. transverse to the I$_2$-He injection flow). The objective is to confine the discharge to the recess regions to minimize its effect on the main O$_2$-He flow. The two pairs of electrodes are connected in parallel with the power supply.
Figure 2.9: Iodine injectors
The I₂-He flow supplied to the iodine injector is generated by flowing preheated helium carrier gas over heated iodine crystals in two 3.8 cm diameter, 12.7 cm length mixing cells connected in parallel. The helium carrier gas flow is activated by a remote-controlled solenoid valve and delivered to the mixing cells through a ¼ inch outer diameter delivery line. The helium carrier flow rate is measured with an Omega FMA-A2322 mass flow meter. A pair of 3-way valves placed immediately upstream and downstream of the iodine-helium mixing cells can divert the helium flow through an ‘iodine-free’ bypass line, as shown in Figure 2.10. Then, the I₂-He mixture passes through a 3.8 cm diameter, 12.7 cm length optical absorption cell with glass windows on each side. Emission from a blue LED is passed through the cell and an optical band-pass filter centered at λ=488 nm to a photodiode to measure absorption by I₂ molecules. Absorption measurements are made twice, first with the He carrier flow passing through the mixing cells and again when the He is flowing through the bypass line so that I₂ number density can be inferred by Beer’s law,

\[ n_{I_2} = \frac{1}{\sigma_{I_2} L} \ln \left( \frac{I_0}{I} \right), \]  

(54)

where \( n_{I_2} \) is the I₂ number density, \( \sigma_{I_2} \) is the absorption cross section at λ=488 nm, \( L \) is the length of the absorption cell, \( I \) is the measured LED intensity with He flowing through the mixing cells, and \( I_0 \) is the measured LED intensity with He flowing through the bypass line. Pressure in the absorption cell is recorded with He flowing
through the bypass line so that iodine vapor does not damage the pressure gage. Temperature is measured by a thermocouple located between the exterior of the diagnostic cell and the heating tape surrounding it, as shown in Figure 2.10. The \( I_2 \) flow rate is determined by multiplying the He flow rate by the ratio of \( I_2 \) and He number densities in the cell,

\[
\dot{n}_{I_2} = \dot{n}_{He} \frac{n_{I_2}}{n_{He}} = \dot{n}_{He} n_{I_2} \frac{k_B T}{p},
\]

where \( \dot{n}_{I_2} \) is the \( I_2 \) mole flow rate, \( \dot{n}_{He} \) is the He mole flow rate, \( k_B \) is the Boltzmann constant, \( T \) and \( p \) are the temperature and pressure in the absorption cell. The \( I_2 \)-He flow is delivered to the iodine injector through a \( \frac{1}{4} \) outer diameter delivery line. To control the temperature in the iodine crystal cells, all delivery lines and cells are heated with variable power tape heaters. Plumbing components downstream of the mixing cells are heated to temperatures higher than the temperature in the mixing cells to avoid excessive iodine vapor condensation on component walls and windows. Iodine vapor flow rate is controlled primarily by the mixing cell temperature and iodine crystal surface area. For this reason, newly refilled \( I_2 \) crystals tend to produce higher iodine vapor flow rates compared to crystals that have been heated for long periods of time and have agglomerated into a nearly solid mass. Setting the \( I_2 \) flow rate requires varying the tape heater power, followed by a waiting period to verify that the system has reached a steady state temperature before \( I_2 \) flow rate can be measured. This is essentially a trial-and-error procedure,
which may be time consuming. Typically, the temperature in the iodine crystal cells is maintained at \( T = 60-80^\circ C \).

Iodine flow rates reported in the present work are subject to significant uncertainty. Accurate measurement of the iodine flow rate depends on the accuracy of measurements of LED transmission through the optical absorption cell with no iodine present, \( I_0 \) in Eq. (53). Although the ‘iodine-free’ He bypass line is used for this, sufficient iodine residue accumulates on the surfaces of all components of the iodine delivery system such that an “iodine-free” measurement is essentially impossible, leading to a significant underestimation of the iodine vapor flow rate. In fact, this effect is so significant that laser action has been observed while the iodine delivery system was operating on bypass, in which case iodine vapor in the injection flow was produced from iodine residue on plumbing components. This effect has
also been observed by the CU Aerospace/University of Illinois group\(^{(75)}\). Iodine residue also accumulates on the windows of the optical absorption cell such that, after the windows are cleaned, the LED transmission through the cell decreases slightly during each run, until the windows are cleaned again. The iodine residue may block the LED signal completely even after a single run unless the optical absorption cell is heated to significantly higher temperatures than the iodine crystal cells. In this case, the temperature measured on the exterior of the optical absorption cell may be significantly higher than the temperature inside the cell, also leading to an underestimation of the iodine flow rate. Note that the temperature inside the iodine crystal cell cannot be measured directly, because exposure of a thermocouple to iodine vapor would damage it. Also note that a typical run is only 5-10 seconds long, which may be insufficient for the temperature in the iodine crystal cell to reach steady state, so the iodine flow rate may well vary during a run.

Since operation of the primary electric discharge in the nozzle plenum significantly affects the intensity of the LED, all iodine flow rate measurements are made immediately after a run with the electric discharge on. The ‘iodine-free’ absorption measurements are made last as an attempt to reduce iodine residue buildup by flowing helium through the \(I_2\) delivery lines after each run.

As shown in Figure 2.1, downstream of the iodine injector is a chilled flow injector with a 0.6 x 10 cm flow cross section and 5 cm length in the streamwise direction. Similar to injectors discussed above, twenty-eight injection ports with a 1.3 mm diameter are evenly distributed on the top and bottom walls across the 10
cm width of the flow channel. Chilled helium or nitrogen is injected into the core flow to reduce flow temperature prior to supersonic expansion. The flow is delivered to the injector through two 3/8 inch outer diameter copper delivery lines. Each line is connected to a flow chiller consisting of 4 meters of 3/8 inch outer diameter copper tubing coiled and immersed into a liquid nitrogen bath. Two 3/8 inch outer diameter bypass lines can deliver uncooled (room temperature) flow to the injector so the effect of pressure rise due to injection may be studied independently of the effect of temperature reduction. The injection flow rate in the range of 0-275 mmole/sec is measured with an Omega FMA 1844 mass flow meter. A schematic of the chilled flow injection system is shown in Figure 2.11.

As shown in Figure 2.11 and Figure 2.12, the chilled flow injector is followed by an aerodynamically contoured M=3 nozzle with a 0.32 x 10 cm throat cross section. The nozzle is formed by a pair of acrylic plastic inserts which span the 10
cm width of the flow channel on the top and bottom walls. These inserts were designed to be replaceable so that the flow Mach number in the laser cavity could be varied; however, prolonged exposure to iodine led to the inserts being effectively glued in place.

Downstream of the nozzle, the flow enters a supersonic section with top and bottom walls diverging at 1.5° each for boundary layer relief. A step-angle supersonic diffuser was initially located 15.3 cm downstream of the nozzle exit, to improve pressure recovery and increase run time. The diffuser was formed by a pair of inserts that spanned the 10 cm flow channel width and had 8°-10° upstream and 10° downstream step angles (see Figure 2.11 and Figure 2.12). The height of the inserts was iteratively reduced to determine the optimum dimensions of the diffuser (see Section 3.1 for details). It was eventually determined that the diffuser provided little benefit to run time duration but significantly diminished Mach number and gain in the downstream portion of the supersonic cavity due to boundary layer separation, so it was removed completely. A photograph of the entire supersonic section, including the supersonic nozzle, laser cavity, and diffuser, is shown in Figure 2.12.
The diffuser section is followed by a 36 cm long (in the streamwise direction) gradual flow expansion section, with the top and bottom walls diverging at 3°. The expansion section is followed by a 4 inch diameter ball valve and a 6 inch diameter PVC pipe 7 m long, connecting the test section to the vacuum system, consisting of two tanks with volumes of approximately 150 and 600 ft³, respectively. The vacuum system is evacuated by either a United Vacuum 400 series 300 cfm vacuum pump or a Stokes 150 cfm vacuum pump. Due to iodine residue buildup and iodine induces corrosion, the vacuum pumps had to be replaced or repaired every few months.

Several interchangeable 4.5 cm thick aluminum side walls can be installed on the supersonic section. The original pair of side walls had 12.7 mm diameter apertures centered vertically with the flow channel and located 3.2 and 12.7 cm downstream of the nozzle exit, shown in Figure 2.13. A bolt pattern on the exterior side of the walls allowed for flanges to be attached directly to the walls, or for flanges to be attached at the end of 3.8 cm diameter, 12.7 cm long extension arms.
The arms were a part of the original design to allow for a longer transverse laser resonator length and to provide a sufficiently large volume between the laser mirrors attached to the ends of the arms and the core flow. In this design, which could be purged to protect the mirror surfaces from direct contact with iodine vapor. However, our previous experiments have shown that adding a purge flow to the arms affects the main flow through the laser cavity, increasing static pressure and reducing the apparent Mach number. For this reason, the mirror purge has not been used in the present work. Typically, the extension arms were not used since they provided no benefit for the laser power. Flanges with either 1 cm diameter wedged, anti-reflection coated BK-7 glass gain windows or 2.54 cm diameter laser mirrors can be attached at either of the two locations for gain or laser output power measurements, respectively. To map gain distribution in the supersonic region, seven additional (nine total) 12.7 mm diameter apertures were spaced along the flow axis, with the first aperture located at the nozzle exit and the last located 12.7 downstream of the nozzle (x=12.7 cm), as shown in Figure 2.14. The spacing of the additional gain windows is not quite regular, to avoid interference with the O-ring seals of the flanges used with two original apertures. In the new side wall design, wedged, anti-reflection coated BK-7 glass windows are flush mounted with the inside wall in each of the seven new apertures.
Figure 2.13: Original side wall with 2 flange compatible apertures

Figure 2.14: Side wall with 9 apertures

flange compatible apertures

apertures with flush mounted windows
When installed, the laser mirrors can form two separate transverse stable resonators, 3.2 and 12.7 cm downstream of the nozzle exit. Mirrors with reflectivity of 99.99, 99.9, 99, and 98.5 % were used. The reflective mirror surface is pressed against a 2.54 cm outer diameter O-ring, which forms a vacuum seal, surrounding a 1.9 cm diameter aperture in the flange, as shown in Figure 2.15. Three thumb screws distributed evenly around the mirror circumference contact a bushing placed on the back side of the mirror. Tightening the thumb screws applies pressure to the edge of the mirror and compresses the O-ring, allowing the mirror angle to be fine-tuned for resonator alignment. Originally, the bushings had 1.27 cm diameter apertures for laser output, matching the aperture size in the side wall, but significantly smaller than the 1.9 cm aperture on the outer side of the flange. When significant divergence of the laser beam was observed in the experiment, the bushing aperture diameter was increased to 1.9 cm, thus resulting in increased laser output power.

![Figure 2.15: Laser mirror mount flanges and assembly](image)
Prior to an experimental run, laser mirrors are aligned using a He-Ne laser guide beam. For this, the 1.27 cm diameter side wall apertures are plugged with plastic inserts each containing a pinhole aperture centered with the larger aperture. The guide beam is aligned through the resonator apertures by passing it through pinhole apertures, insuring that it passes through the center of each side wall aperture. Laser mirror mounts are installed at the ends of the 12.7 cm extension arms or directly on the side walls, forming a stable, transverse resonator with a 10 cm gain path and a 44.4 cm (with extension arms) or 19 cm (without extension arms) total resonator length, respectively. The laser mirrors are then aligned by directing the guide beam reflection back on a pin-hole aperture located near the laser source, as shown in Figure 2.16. Pressure change in the flow channel noticeably alters the mirror alignment, so alignment is done while the test section is pumped to vacuum. It is likely that even small pressure changes during the run (2-3 torr) detrimentally affect the mirror alignment, thus leading to reduced output power. Attempts to increase laser output power by adjusting mirror alignment during a run were unsuccessful. Since run times of several seconds are separated by 20-25 minutes, using this approach is essentially not feasible.

**Figure 2.16: Laser mirror alignment schematic (top view)**
An additional pair of sidewalls was manufactured to accommodate a Z-pass laser resonator, to be operated with four 50.8 mm diameter laser mirrors, as shown in Figure 2.17. The mirror pairs are located 3.2 and 10.8 cm downstream of the nozzle exit. Mirrors 2 and 3 are mounted at an 11° angle to turn the optical path by 22°. Each mirror is recessed 4.5 cm from the inside surface of the side wall and accesses the flow through an aperture 1.5 cm high and 4.4 cm long (in the streamwise direction). Note that the flow channel height increases in the streamwise direction due to diverging top and bottom walls (for boundary layer relief) such that the height varies from 1 cm at the nozzle exit to 1.5 cm at the downstream edge of the resonator. Mirrors 1 and 4 are concave with a curvature radius of R=2 m and mirrors 2 and 3 are planar. Mirrors 1, 2, and 3 are nominal ‘total reflectors’ (with reflectivity of 99.9%) and mirror 4 is an output coupler. One additional side wall was manufactured with only one aperture to hold a single, 50.8 mm diameter laser mirror 3.2 cm downstream of the nozzle exit, such that a simple transverse, two-mirror resonator could be tested with the large diameter, 50.8 mm laser mirrors. Mirror mounts and alignment procedures for each larger resonator are similar to those of the smaller resonator described above.
2.2 Diagnostics

During the operation, discharge pressure and static pressure in the supersonic section are monitored. Pressure taps are located in the top wall on the channel center line (5 cm from either side wall), as shown in Figure 2.12. Two pressure taps are located in the subsonic region, upstream of the first primary discharge (estimated flow Mach number M=0.1) and immediately upstream of the nozzle entrance (M=0.3). Two additional pressure taps are located in the supersonic region, 3.2 and 12.7 cm downstream of the nozzle exit (M=3.0). The pressure measured using a tap upstream of the discharge section is very close to the flow stagnation pressure. Flow Mach number is estimated from quasi-one-dimensional isentropic flow equations. Pressures are typically measured with a pressure transducer with a digital readout accurate to 0.1 torr. Time-dependent pressure traces shown in Section 3.1 below are taken with an Omega OMB-DAQ-56 USB Data
Acquisition System. Due to electromagnetic interference from the discharge, all pressure measurements are taken with the discharge turned off.

Small signal gain measurements at $\lambda=1315$ nm are made by tunable diode laser absorption spectroscopy (TDLAS) using a PSI Iodine Scan probe (76). In this method, a gain probe laser beam is directed across the supersonic laser cavity, transverse to the flow, and scans across the iodine emission/absorption line at a frequency of 100 Hz. The absorption/gain line sampling frequency is 10 Hz, i.e. a line shape is taken and stored every 0.1 seconds. Ten line shapes are averaged and results are displayed on the screen in real time, as well as saved in a spreadsheet. The gain probe scans across a spectral region of about 5,800 MHz (0.2 cm$^{-1}$) and can be tuned to access all 6 allowed hyperfine $I^* \rightarrow I$ transitions (see Section 3.3). The diode laser output beam is split internally into an internal reference beam, an external reference beam, and a probe beam. The probe beam signal is normalized by the intensity of an internal reference beam to reduce the noise generated from small fluctuations in beam intensity. An absorption cell containing iodine vapor, heated to $T=623$ K in a tube furnace, is used to produce measurable concentrations of thermally dissociated iodine atoms. The gain probe is tuned by adjusting the diode laser current and reference absorption is measured in the heated iodine absorption cell. The probe beam is delivered to the experiment through a 1SME-TB2-OFNR(ETL)OFNFT4(CSA) Corning optic cable. Extension cables are employed as needed, so the gain probe electronics can be moved a sufficient distance from the discharge to reduce the effect of the electromagnetic noise. Absolute single-pass
gain is measured and divided by gain path length (10 cm) in order to calculate gain per unit length. Typically, the probe beam is aligned manually through the center of gain windows using an infrared card, except for gain measurements in the boundary layer, discussed in Section 3.8.

Emission spectroscopy measurements are made with a Roper Scientific Optical Multichannel Analyzer (OMA) V with a 512 x 1 pixel InGaAs linear photodiode array (PDA) camera, and a Spectra Pro 0.5 m monochromator with a 600 grooves/mm grating blazed at 1 μm. Signal is collected through one of the optical access windows in the side wall of the test section (e.g. see Figure 2.1) with a 3.3 cm diameter collimator lens and transmitted to the monochromator inlet slit with a 2 m long ORIEL model #77527 optical fiber bundle. The OMA is calibrated using a mercury-argon lamp. The camera and the optical fiber position in front of the monochromator slit are adjusted to maximize the signal strength and resolution of the 1013.975 nm Ar emission line produced by an Hg-Ar lamp. Absolute emission measurements, used to infer O₂(1Δ) yield, are calibrated using an Infrared Technologies Inc. model 563 blackbody source with an Infrared Technologies Inc. model 201 temperature controller. Calibration spectra are taken with the blackbody set to a temperature of T=1073 K through a 1 cm diameter aperture (to match the optical access window aperture in the laser experiment), at a distance of 4.5 cm from the aperture (to match the side wall thickness used in the laser experiment). The collimator is aligned with the blackbody source aperture by hand to maximize
the signal intensity. For $O_2(^1\Delta)$ yield measurements, the monochromator slit is typically set to 0.2-0.4 mm.

To infer $O_2(^1\Delta)$ yield from the emission spectra, the following procedure is used. The number of photons emitted per unit volume per steradian by spontaneous emission on the $O_2(^1\Delta \rightarrow ^3\Sigma)$ transition is given as,

$$E_{O_2(^1\Delta)} = \frac{1}{4\pi} n_{O_2(^1\Delta)} \cdot A_{O_2(^1\Delta \rightarrow ^3\Sigma)} \cdot t_{\text{exp}}\ [m^{-3}\text{sr}^{-1}], \quad (56)$$

where $n_{O_2(^1\Delta)}$ is the number density of $O_2(^1\Delta)$, $A_{O_2(^1\Delta \rightarrow ^3\Sigma)} = 2.22 \cdot 10^{-4}$ sec$^{-1}$ is the Einstein coefficient for spontaneous emission for the $O_2(^1\Delta \rightarrow ^3\Sigma)$ transition$^{(43)}$, and $t_{\text{exp}}$ is the experiment exposure time. For a finite emission source volume with uniform concentration of $O_2(^1\Delta)$, the number of counts recorded by the PDA camera is,

$$S_{O_2(^1\Delta)} = \iiint R \cdot C \cdot E_{O_2(^1\Delta)} \cdot dV = C \cdot E_{O_2(^1\Delta)} \iiint R \cdot dV, \quad (57)$$

where $S_{O_2(^1\Delta)}$ is camera counts, $R$ is the fraction of emitted photons collected by the collimator lens and determined by the solid angle from which the emission signal is collected, and $C$ is the conversion factor from photons collected to detector counts, determined by the collection and spectrometer optics and by the detector efficiency. The number of photons emitted per unit surface area per steradian by a blackbody source over a wavelength range $\Delta \lambda$ is,
\[ E_{BB} = I(\lambda, T) \cdot \frac{1}{\varepsilon} \cdot \Delta \lambda \cdot t_{BB} \left[ m^{-2} sr^{-1} \right], \quad (58) \]

where

\[ I(\lambda, T) = \frac{2hc^2}{\lambda^5} \left\{ \exp \left( \frac{hc}{\lambda k_B T_{BB}} \right) - 1 \right\}^{-1}, \quad (59) \]

is the blackbody emission intensity, \( \varepsilon = hc/\lambda \) is the photon energy, \( c \) is the speed of light in vacuum, and \( T_{BB} \) is the blackbody temperature. During the calibration, \( \Delta \lambda \) was set to 40 nm (1250-1290 nm) to match the spectral bandwidth of the \( O_2(^{1}\Delta, \nu' = 0 \rightarrow ^{3}\Sigma, \nu'' = 0) \) band. The number of counts recorded by the PDA camera is

\[ S_{BB} = \iint R \cdot C \cdot E_{BB} \cdot dA = C \cdot E_{BB} \iint R \cdot dA. \quad (60) \]

In the present experiments, \( R \) is the same for the volumetric SDO source and for the blackbody aperture, since the blackbody aperture diameter was set to 1 cm to match the optical access window diameter. Contributions from all portions of the \( O_2(^{1}\Delta) \) emission collection volume,

\[ V_{exp} = \pi L \frac{D^2}{4} = 0.785 \text{ cm}^3 \quad (61) \]
are the same. This has been verified by mapping the collimator lens signal collection region, for which the collimator was placed at different distances from the blackbody aperture. In Eq. (61), \( L=10 \text{ cm} \) is the flow channel width and \( D=1 \text{ cm} \) is the aperture diameter. These measurements demonstrate that the collected signal intensity remains nearly independent of the distance between the source (i.e. the blackbody aperture) and the collimator lens. Solving for the SDO number density and dividing by the total \( \text{O}_2 \) number density in the flow, SDO yield is given by,

\[
Y = \frac{k_B T}{x_{\text{O}_2} p} \left( \frac{S_{\text{O}_2(1\Delta)}}{t_{\exp} V_{\exp}} \right) \left( \frac{t_{\text{BB}} A_{\text{BB}}}{S_{\text{BB}}} \right) \left( \frac{4\pi \cdot 1(\lambda T_{\text{BB}}) \frac{1-\Delta \lambda}{A_{\text{O}_2(1\Delta-3\Sigma)}}}{A_{\text{O}_2(1\Sigma \rightarrow 3\Sigma)}} \right),
\]

(62)

where \( T \) and \( p \) are the flow temperature and pressure, \( x_{\text{O}_2} \) is the oxygen mole fraction in the \( \text{O}_2\)-He mixture, \( A_{\text{BB}} \) is the blackbody aperture area, and \( V_{\exp} \) given by Eq. (61) is the volume of the flow from which SDO emission is collected by the collimator lens, i.e. window cross-sectional area times the flow channel width. Rotational temperature of the flow is inferred from the \( \text{O}_2(1\Sigma \rightarrow 3\Sigma) \) visible emission spectra using a synthetic spectrum \(^{21} \). At the present experimental conditions (\( P=50\text{-}100 \text{ torr} \)), rotational and translational temperatures are essentially the same.

Laser output power measurements are made with a 2 inch aperture Scientech AC5000 thermopile calorimeter connected to Scientech S310 power meter. Short laser run times (5-10 seconds) precluded the thermopile from reaching steady state, which occurs within about 30 seconds. An internal algorithm in the power meter estimates power based on the time-dependent voltage output of the
thermopile calorimeter during the transient regime; however, this procedure was determined to be incorrect, such that the peak power displayed by the digital readout overestimated the actual power by about 50%. In the present work, the laser power was determined by calibrating the calorimeter, while operating it in the transient regime, using a c.w. CO laser. For this, the calorimeter was suddenly exposed to the laser radiation and its time-resolved voltage output was recorded with an oscilloscope. The CO laser power was determined from a power meter reading after the calorimeter reached steady state. The calibration results are summarized with the following equation,

\[ U = 0.07P_L\{1 - \exp(-0.13t)\} \]  \hspace{1cm} (63)

where \( U \) is the recorded voltage output of the calorimeter, \( P_L \) is the laser output power in Watts, and \( t \) is time in seconds. The calibration curve fits the voltage output well for up to 10 seconds for the full range of CO laser power available, up to 12 W. Figure 2.18 shows calorimeter output voltage traces for different c.w. CO laser powers, along with the calibration curve fit.
Figure 2.18: Power meter calibration
Chapter 3: Results

The primary objective of the present work is development of a electrically excited oxygen-iodine laser scalable to high powers. During the laser development, several significant modifications of the experimental apparatus have been made. The results of experiments using several different laser modifications are presented approximately in chronological order. A schematic of the baseline apparatus configuration is shown in Figure 3.1. Listing the components in the direction of the flow, the initial laser configuration was as follows: 1 inch diameter main flow delivery line, flow expansion section discussed in Section 2.1 and shown in Figure 2.3, 1 primary electric discharge section shown in Figure 2.6, emission spectroscopy diagnostic section shown in Figure 2.8, iodine injector 1 shown in Figure 2.9, M=3 supersonic nozzle with supersonic test section shown in Figure 2.12 (using side walls with only two 1.27 cm diameter apertures located 3.2 and 12.7 cm downstream of nozzle exit), supersonic diffuser shown in Figure 2.12, gradual flow expansion section, and vacuum system.
3.1 Diffuser Modification

As discussed in Section 2.1, the supersonic diffuser was located 15.3 cm downstream of the nozzle exit (see Figure 3.1 and Figure 2.12). Initially, the diffuser inserts had upstream and downstream step angles of 10° each, and the diffuser throat height had an intentionally restrictive area ratio (i.e. the ratio of diffuser height over the test section channel height) of $A_D/A_{TS}=0.563$. Since low-pressure diffuser performance is difficult to predict, the diffuser throat height was gradually increased using the iterative process described below. During the diffuser test, the supersonic test section was operated at the following flow conditions: $p_0=120$ torr, 14 % $O_2$-He mixture (118 mmole/sec $O_2$, 706 mmole/sec He). Static pressure was recorded in the supersonic test section 3.2 cm and 12.7 cm downstream of the nozzle exit ($p_1$ and $p_2$, respectively). Mach number in the supersonic section was inferred from the static-to-plenum pressure ratio using quasi-one-dimensional isentropic flow equations,
The diffuser inserts were then removed and milled to increase the diffuser height, after which the modified diffuser was tested again at the same flow conditions. During this iterative process, the leading edge step angle was also reduced to 8°. The process continued until further increase in the diffuser height no longer affected the flow Mach number upstream of the diffuser. The results of these measurement are summarized Table 3.1. Note that the design flow Mach number of M=3.0 was not achieved at the downstream pressure tap location, 12.7 cm downstream of the nozzle exit and 2.6 cm upstream of the diffuser insert (see Figure 2.12). The increased pressure at this location, compared to pressure at the upstream pressure tap location (note $p_2/p_0 > p_1/p_0$ in Table 3.1) is likely due to flow separation regions upstream of the diffuser step angle. Although increasing the diffuser throat height continued to increase the apparent Mach number at the downstream location, as can be seen from Table 3.1, the process was stopped at $A_D/A_{TS}=0.823$. It was assumed that achieving the design M=3 flow in the upstream region of the test section would be sufficient for laser experiments, while further diffuser modification might detrimentally affect the flow and reduce the run time.

\[
\frac{p}{p_0} = \left(\frac{T}{T_0}\right)^{\frac{k}{k-1}} = \left(1 + \frac{k-1}{2} M^2\right)^{\frac{k}{k-1}}. \tag{64}
\]
Thus, after the last iteration, the diffuser had 8° upstream and 10° downstream step angles with the area ratio of $A_D/A_{TS}=0.823$. The diffuser performance at a variety of plenum pressures and $O_2$ mole fractions in the flow is summarized in Table 3.2. The baseline flow conditions at which highest gain was produced in a small-scale DOIL apparatus used in previous work at Ohio State\(^{(71)}\) are as follows: $p_0=60$ torr, 7% $O_2$-He flow (34 mmole/sec $O_2$, 446 mmole/sec He). Figure 3.2 shows time-resolved pressure traces at the baseline main flow rates with and without injection of additional He flow of 78 mmole/sec through the iodine injector (to match the previous optimum DOIL operation conditions)\(^{(71)}\). During these measurements, initial back pressure was below 1 torr for each case. The run time, measured from the moment when the pressures in the supersonic cavity stabilized until the back pressure increased sufficiently to affect the flow in the supersonic cavity (observed as the onset of a time dependent static pressure rise), was about 5-6 seconds.

<table>
<thead>
<tr>
<th>$A_D/A_{TS}$</th>
<th>$p_1/p_0$</th>
<th>$p_2/p_0$</th>
<th>$M_1$</th>
<th>$M_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.563</td>
<td>0.244</td>
<td>0.268</td>
<td>1.52</td>
<td>1.46</td>
</tr>
<tr>
<td>0.672</td>
<td>0.126</td>
<td>0.165</td>
<td>1.98</td>
<td>1.79</td>
</tr>
<tr>
<td>0.727</td>
<td>0.0355</td>
<td>0.0565</td>
<td>2.89</td>
<td>2.54</td>
</tr>
<tr>
<td>0.751</td>
<td>0.0314</td>
<td>0.0540</td>
<td>2.98</td>
<td>2.57</td>
</tr>
<tr>
<td>0.774</td>
<td>0.0308</td>
<td>0.0486</td>
<td>3.00</td>
<td>2.65</td>
</tr>
<tr>
<td>0.774*</td>
<td>0.0303</td>
<td>0.0486</td>
<td>3.01</td>
<td>2.65</td>
</tr>
<tr>
<td>0.798*</td>
<td>0.0308</td>
<td>0.0455</td>
<td>3.00</td>
<td>2.70</td>
</tr>
<tr>
<td>0.823*</td>
<td>0.0312</td>
<td>0.0424</td>
<td>2.99</td>
<td>2.75</td>
</tr>
</tbody>
</table>

* Indicates an upstream step angle of 8°
Table 3.2: Diffuser Performance

<table>
<thead>
<tr>
<th>Mixture</th>
<th>$p_0$</th>
<th>$p_1/p_0$</th>
<th>$p_2/p_0$</th>
<th>$M_1$</th>
<th>$M_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.3 % O₂-He</td>
<td>60.0</td>
<td>0.0500</td>
<td>0.0700</td>
<td>2.63</td>
<td>2.39</td>
</tr>
<tr>
<td>7.0 % O₂-He</td>
<td>60.3</td>
<td>0.0481</td>
<td>0.0697</td>
<td>2.66</td>
<td>2.39</td>
</tr>
<tr>
<td>10.3 % O₂-He</td>
<td>60.0</td>
<td>0.0400</td>
<td>0.0533</td>
<td>2.79</td>
<td>2.58</td>
</tr>
<tr>
<td>7.0 % O₂-He</td>
<td>86.0</td>
<td>0.0372</td>
<td>0.0593</td>
<td>2.85</td>
<td>2.51</td>
</tr>
<tr>
<td>14.3 % O₂-He</td>
<td>125.0</td>
<td>0.0312</td>
<td>0.0424</td>
<td>2.97</td>
<td>2.75</td>
</tr>
</tbody>
</table>

Figure 3.2: Time-resolved pressure traces (with diffuser). 7 % O₂-He; 34 mmole/sec O₂, 446 mmole/sec He through the discharge section, (A) $p_0$=60 torr; (B) $p_0$=86 torr, 78 mmole/sec additional He injection; upstream and downstream locations 3.2 and 12.7 cm downstream of nozzle exit, see Figure 2.12

3.2 Temperature and $O_2(^1\Delta)$ Yield Measurements

Absolute infrared emission spectroscopy measurements with black body calibration were made in order to measure the $O_2(^1\Delta)$ yield produced in the transverse 13.56 MHz CCRF primary electric discharge in the plenum, as discussed in Section 2.2. The discharge was diffuse and stable in the entire range of flow conditions and discharge powers used. Typical camera exposure time was $t_{SDO}=2-4$ seconds for $O_2(^1\Delta)$ emission spectra and $t_{BB}=0.02$ seconds for blackbody emission spectra. Flow temperature downstream of the discharge is inferred from a
$O_2(^3\Sigma \rightarrow ^3\Sigma)$ visible emission spectra using best fit synthetic spectra \(^{(21)}\). A typical experimental spectrum and the best fit synthetic spectrum are shown in Figure 3.3. The translational-rotational temperature inferred at these conditions is $T=380\pm10$ K. Flow temperature vs. primary discharge power is shown in Figure 3.4. As can be seen from Figure 3.4, the temperature increases nearly linearly with the discharge power, at a rate of about 25 K/kW. The linear flow temperature rise was assumed in the $O_2(^1\Delta)$ yield data reduction procedure for all present experimental conditions.

Figure 3.3: Typical $O_2(^3\Sigma \rightarrow ^3\Sigma)$ emission spectrum shown with best fit synthetic spectrum. 7 % O2-He; 34 mmole/sec O2, 446 mmole/sec He, 375 µmole/sec NO through the discharge section, $p_0=60$, inferred temperature $T=380\pm10$ K
From the data of Figure 3.4, the estimated fraction of the discharge power converted to heat is

$$F_{th} = \frac{\Delta T}{P_p} \sum nC_p \sim 27\%,$$

(65)

where $P_p$ is the discharge power. A typical $O_2(^1\Delta \rightarrow ^3\Sigma)$ emission spectrum is shown together with a sample blackbody source spectrum in Figure 3.5. The signal integration domain for both spectra, used in Eq. (61), is $\lambda=1250-1290$ nm. A background spectrum is taken with the discharge turned off and subtracted from each experimental spectrum. However, the $O_2(^1\Delta \rightarrow ^3\Sigma)$ emission spectrum has an offset caused by both electromagnetic noise generated by the discharge and broadband chemiluminescence emission from electronically excited $NO_2^*$ generated.
in reaction of Eq. (42) (see Section 2.2). To compensate, a constant offset was subtracted from the $O_2(^1\Delta \rightarrow ^3\Sigma)$ spectrum, equal to the average of the signal from two spectral regions, $\lambda=1240-1250$ nm and $\lambda=1290-1300$ nm. The spectrum in Figure 3.5 is shown with the offset subtracted.

![Figure 3.5: Typical $O_2(^1\Delta \rightarrow ^3\Sigma)$ emission spectrum and blackbody emission spectrum. 7 % $O_2$-He; 34 mmole/sec $O_2$, 446 mmole/sec He, 375 µmole/sec NO through the discharge section; $p_0=83$; 85 mmole/sec He injected downstream of discharge section](image)

$O_2(^1\Delta)$ yield vs. the discharge power, measured at different main flow rates (and discharge pressures) is shown in Figure 3.6. Yield measured at different discharge pressures, but at the same main flow rate, is plotted in Figure 3.7. In Figure 3.6, the 33 % plenum pressure increase from $p_0=60$ torr to $p_0=80$ torr is due to a 33 % main $O_2$-He flow rate increase, from 479 mmole/sec to 639 mmole/sec. As can be seen from Figure 3.6, the yield is much lower at $p_0=80$ torr, due to a higher flow rate resulting in reduced specific energy loading (i.e. primary discharge energy per $O_2$ molecule). On the other hand, In Figure 3.7, the pressure increase from $p_0=60$
to $p_0=83$ torr is due to an additional 85 mmole/sec flow of He injected downstream of the discharge. The injection increases the plenum pressure and residence time of the O$_2$-He flow in the discharge, without affecting the main flow rate. Although in this case slightly higher yields are observed at the lower pressure of $p_0=60$ torr, the difference is not as significant as in the first case (see Figure 3.6). As can be seen from these results, at these conditions O$_2(^1\Delta)$ yield is only moderately affected by discharge pressure, but is significantly affected by specific energy loading. The estimated fraction of discharge power spent on O$_2(^1\Delta)$ generation is

$$F_{O_2(^1\Delta)} = \frac{Y\dot{n}_{O_2}\varepsilon_{O_2(^1\Delta)}}{P_p},$$

where $Y$ is the yield, $\dot{n}_{O_2}$ is the O$_2$ mole flow rate, $\varepsilon_{O_2(^1\Delta)} = 0.98$ eV is the excitation energy of O$_2(^1\Delta)$, and $P_p$ is the primary discharge power. With O$_2(^1\Delta)$ yields leveling off at $Y=7.5$ %, $P_p=3.0$ kW, and O$_2$ flow rate of $\dot{n}_{O_2}=34$ mmole/sec, the primary discharge power fraction spent on O$_2(^1\Delta)$ excitation is $F_{O_2(^1\Delta)}=8$ %.
Figure 3.6: Yield vs. primary discharge power at different flow rates. (A) 7% O₂-He; 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section; \(P_0=60\) torr. (B) 7% O₂-He 45 mmole/sec O₂, 594 mmole/sec He, 375 µmole/sec NO flow through the discharge section, \(P_0=80\) torr.

Figure 3.7: Yield vs. discharge power at different pressures. (A) 7% O₂-He 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section; \(P_0=60\) torr. (B) 7% O₂-He 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section; 85 mmole/sec additional He injection, \(P_0=83\) torr.
Yield measurements were taken at a moderate primary discharge power of $P_p = 1 \text{ kW}$, to demonstrate yield dependence on flow rate and $O_2$ mole fraction in the flow. The results are shown in Figure 3.8 and Figure 3.9, respectively. It can be seen that the addition of trace small amounts of NO, up to 1600 ppm, increases $O_2(^1\Delta)$ yield by up to 20%, consistent with previously reported data\textsuperscript{[40; 41]}. On the other hand, yield decreases with $O_2$ mole fraction, due to reduced power loading at higher $O_2$ mole fractions, a trend similar to what the one observed in Figure 3.6 and Figure 3.7.

![Figure 3.8: $O_2(^1\Delta)$ yield vs. NO flow rate. 7 \% $O_2$-He; 34 mmole/sec $O_2$, 446 mmole/sec He flow through the discharge section; $p_0=60$ torr; $P_p=1 \text{ kW}$](image-url)
Infrared emission spectroscopy measurements have also been attempted in the supersonic test section, when iodine vapor was injected into the flow. The primary objective of these measurements was to measure the \( O_2(^1\Delta) \) amount remaining in the flow after the loss due to iodine dissociation by reaction of Eq. (11), and also to measure iodine dissociation fraction by determining the total iodine atom density using a combination of small signal gain and emission spectroscopy measurements. Typical emission spectra obtained at these conditions are shown in Figure 3.10. It can be seen that the \( I_2(3\Pi_1, v' \rightarrow 1\Sigma, v'') \) emission bands completely obscure the \( O_2(1\Delta \rightarrow 3\Sigma) \) emission, thus making impossible \( O_2(1\Delta) \) concentration measurements in the supersonic cavity by this method. Electronically excited iodine, \( I_2(3\Pi_1, v') \) is generated in the flow by collisional energy transfer from \( O_2(1\Delta) \) and \( O_2(1\Sigma) \) \(^{12; 10; 11}\), as well as by iodine atom recombination. As discussed in Section 2.1,
iodine vapor is present in the supersonic test section even at conditions when iodine is not injected into the flow, due to iodine accumulation on the flow channel walls. This effect also precluded O$_2(^1\Delta)$ measurements in the supersonic test section, even without iodine injection.

3.3 Gain Measurements on I$^*$→I Hyperfine Structure Components

Gain measured on all six allowed transitions between the two hyperfine split states of I$'(^2P_{1/2})$ and the four hyperfine split states of I($^2P_{3/2}$) is shown in Figure 3.11. Gain spectrum shown in Figure 3.11 was taken during 3 separate runs since the limited spectral range of the gain probe laser could not access all six gain/absorption transitions in the same run. Spectrum 1 shows (F$'=$3→F$''=$4) and (F$'=$3→F$''=$3) hyperfine transitions. Spectrum 2 shows (F$'=$3→F$''=$3) and
(F'=3→F''=2) transitions, partially overlapping with spectrum 1. Finally, spectrum 3 shows (F'=2→F''=3), (F'=2→F''=2), and (F'=2→F''=1) transitions. Gain on each of these transitions has been normalized by gain on a (F'=3→F''=4) transition, such that gain in Figure 3.11 is shown in relative units. Gain/absorption line frequency is shown relative to the frequency of the (F'=3→F''=4) transition. Also shown in Figure 3.11 are the theoretical gain values, calculated from Eq. (29) assuming equilibrium among the hyperfine split components (see Table 1.2), as well as the experimental values obtained from a luminescence spectrum measured during photodissociation of C₃F₇I (15). The results of the present measurements show the populations of the hyperfine split states to be in equilibrium. The measured energy difference between the four hyperfine split states of the ground electronic state of I atom are in good agreement with theory. The measured energy difference between the two hyperfine split states of the excited electronic state I*(2P₁/₂) is slightly greater than predicted by theory. All subsequent gain measurements reported in the present work are made for the (F'=3→F''=4) transition.
3.4 Initial Gain Measurements

Gain measurements for a baseline laser configuration (see Figure 3.1) were made 3.2 cm downstream of the nozzle exit through the upstream pair of optical access windows (see Figure 3.1). Typical run conditions were as follows: 7% O\textsubscript{2}-He mixture; 34 mmole/sec O\textsubscript{2}, 446 mmole/sec He, 375 \textmu m mole/sec NO flow through the discharge section, \( P_0 = 80-86 \) torr after injecting 100-150 \textmu m mole/sec I\textsubscript{2} in 80 mmole/sec He carrier downstream of the discharge section, discharge power \( P_p = 0-3.5 \) kW. Gain measured at these conditions is plotted vs. NO flow rate in Figure 3.12. As can be seen, gain increases drastically with NO flow rate from, weak absorption without NO, \( \gamma = -0.012 \%/\text{cm} \), to positive gain up to \( \gamma = 0.116 \%/\text{cm} \) at an optimal NO flow rate of 375 \textmu m mole/sec. As discussed in Section 1.3, the dominant effect of NO on plasma kinetics (or chemistry) is removal of O atoms (by reactions of Eqs. 43 and 44), which rapidly deactivate I\textsuperscript{*} (by reaction of Eq. 41). Also, a modest O\textsubscript{2}(\textsuperscript{1}\Delta) yield
increase due to NO addition (shown in Figure 3.8) may contribute to this effect. As can be seen from these results, the O atoms generated in the discharge are so detrimental that an iodine atom population inversion is not achieved at the baseline conditions, unless NO is added to the flow. Figure 3.12 also shows that gain starts to drop off at high NO flow rates, greater than 400 µmole/sec, suggesting that complete removal of O atoms may not be optimal, since small amounts may be beneficial in the I₂ dissociation process (by reaction of Eqs. 40 and 41). It is also possible that NO acts as an I' quencher,

\[ \text{NO} + \text{I}' \rightarrow \text{NO} + \text{I}, \quad (67) \]

with a rate constant \( k_67 = 1.2 \cdot 10^{-13} \text{ cm}^3/\text{sec}^{(40)} \). However, Carroll et al. concluded that NO is ineffective as an I' quencher \(^{(40)}\) at their DOIL operating conditions.
Gain vs. I$_2$ flow rate is shown in Figure 3.13, for the discharge power of P$_p$=3.0 kW. Controlling the I$_2$ flow rate is of critical importance for maximizing gain in a DOIL, however it is difficult to control and measure, as discussed in detail in Section 2.1. From Figure 3.13, it can be seen that gain peaks near $\gamma$=0.12 %/cm for the optimum flow I$_2$ flow rate of 150-200 µmole/sec. At lower I$_2$ flow rates, gain is limited by the total atomic iodine number density available in the flow. Clearly, gain should go to zero as iodine flow rate approaches zero. Gain also falls off at high I$_2$ flow rates, most likely due to O$_2$(^1\Delta) depletion from I$_2$ dissociation in reaction of Eq. (11). In the range of conditions tested, I$_2$ flow rate could be increased until absorption is measured in the supersonic cavity. As will be seen in subsequent sections, “optimum gain” conditions in the present laser apparatus typically occur at somewhat lower I$_2$ flow rates of 100-150 µmole/sec.
Optimal I\(_2\) flow rate is controlled by the O\(_2\)\(^{1}\Delta\) flow rate, which in turn depends on the discharge power. Gain vs. discharge power measured at different I\(_2\) flow rates is shown in Figure 3.14. Given the difficulty in varying the I\(_2\) flow rate systematically (see Section 2.1), it was easier to set the I\(_2\) flow rate to a constant value and vary the primary discharge power. From Figure 3.14, one can see that for each I\(_2\) flow rate, gain initially increases with the primary discharge power, due to greater O\(_2\)\(^{1}\Delta\) yield at higher powers. Eventually, the effect of flow heating in the discharge (with the flow temperature increasing approximately linearly with power, see Figure 3.4) offsets the yield increase, which levels off at about 2.5-3 kW, see Figure 3.7 and gain begins to fall with power. It can also be seen that the optimum I\(_2\) flow rate increases with discharge power, as expected, since higher O\(_2\)\(^{1}\Delta\) flow rate
at higher discharge power make the laser mixture less susceptible to \( \text{O}_2(\text{I}^\Delta) \) loss to iodine dissociation (see Eq. (11)).

![Graph showing gain vs. discharge power at different I\(_2\) flow rates](image)

**Figure 3.14**: Gain vs. discharge power at different I\(_2\) flow rates. 7 % O\(_2\)-He; 34 mmole/sec O\(_2\), 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section; \( p_0 = 86 \text{ torr} \) after injection of 85 mmole/sec; \( x = 3.2 \text{ cm} \)

Time-resolved gain is plotted in Figure 3.15 for the optimum baseline gain measurement conditions, as follows: 7 % O\(_2\)-He mixture; 34 mmole/sec O\(_2\), 446 mmole/sec He, and 375 µmole/sec of NO flow through the discharge section, \( p_0 = 86 \text{ torr} \) after injection of 140 µmole/sec I\(_2\) in 85 mmole/sec He; \( P_p = 3.0 \text{ kW} \). One can see that gain peak value is \( \gamma = 0.116 \text{ %/cm} \), and gain remains steady in time, with variation within 10 %, for over 7 seconds, until back pressure rise adversely affects the supersonic flow field in the laser cavity.
Figure 3.15: Time resolved gain measurements. 7 % O₂-He; 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow in the discharge section; p₀=86 torr after injection of 140 µmole/sec I₂ in 85 mmole/sec He; Pₚ=3.0 kW; x=3.2 cm

3.5 Baseline Laser Output Power Measurements

Laser output power measurements for the baseline laser configuration (see Figure 3.1) were made at the optimum gain conditions: 7 % O₂-He mixture; 34 mmole/sec O₂, 446 mmole/sec He, and 375 µmole/sec of NO flow through the discharge section; p₀=81 torr after injection of 130 µmole/sec I₂ in 85 mmole/sec He; Pₚ=2.5 kW. The laser resonator operated using two 1.27 cm diameter apertures located 3.2 cm downstream of the nozzle exit (see Figure 3.1). Concave (R=1 m) laser mirrors were installed at the ends of the 12.7 cm extension arms, forming a stable, transverse resonator with a 10 cm gain path and a 44.5 cm total resonator length. Gain measurements made at the resonator location immediately prior to laser mirror installation showed gain of about γ=0.1 %/cm (2 % double pass gain).
In this series of laser output power measurements, a 1.27 cm diameter aperture mirror bushings on non-reflective sides of the mirrors (see Figure 2.15) were used.

Laser action was achieved for two different mirror combinations, a pair of identical 99.9 % reflectivity mirrors and a 99.9 % reflectivity mirror with a 99 % reflectivity output coupler. The measured double pass gain of 2 % greatly exceeds the gain threshold for laser action for these mirror combinations, $\gamma_{th}=0.2 \%$ and $\gamma_{th}=1.1 \%$, respectively. For the 99.9 %-99.9 % mirror combination, a total laser output power of 1.7 W was measured, with approximately 50 % of power coupled out on each side of the resonator. For the 99-99.9 % mirror combination, a laser output power of 1.6 W was coupled out on the 99 % mirror side, with less than 0.1 W coupled out on the 99.9 % mirror side. The laser output power measured in these experiments, was much lower than expected, since a similar laser output power of 1.4 W had been achieved in the previous DOIL apparatus with a double-pass gain of only 0.4 % (i.e. a factor of 5 lower than the double-pass gain in the present baseline configuration) (71). The total power stored in $O_2(^1\Delta)$ flow through the laser cavity and available for lasing,

$$Q = \dot{n}_{O_2}(Y-Y_{th})N_A \varepsilon_{O_2(^1\Delta)}$$

is approximately 160 W at the present conditions. This demonstrates that only a small fraction of energy available for lasing is coupled out. The low $O_2(^1\Delta)$ energy fraction coupled out as laser power, measured in the experiments, suggested that
additional power can be coupled out using a second transverse resonator placed downstream of the first resonator. To determine feasibility of such two-resonator operation, laser gain was measured at several axial locations in the supersonic cavity.

3.6 Axial Gain Measurements and Diffuser Removal

First, gain measurements were made for the optimum baseline gain conditions at two axial locations, 3.2 and 12.7 cm downstream of the nozzle exit, using two pairs of 1.27 cm apertures, as shown in Figure 3.1. The results of these measurements are shown in Figure 3.16. Gain lineshapes, approximated with a Doppler lineshape (see Section 1.3), also shown in Figure 3.16, were used to infer translational temperature in the cavity. As can be seen, gain is reduced by a factor of 4 at the downstream location, in spite of a slightly lower temperature, T=120 K vs. T=125 K at the upstream location. The lower temperature measured at the downstream was unexpected, since the higher static pressure measured at this location (see Table 3.2 and Figure 3.2) indicates a lower flow Mach number based on a quasi-one-dimensional isentropic flow equation, which corresponds to a higher static temperature. This suggests that the flow in the supersonic section may well be non-one-dimensional and non-isentropic, likely due to the effect of angle step diffuser causing flow separation upstream. The uncertainty of the temperature inference using the Doppler lineshape fit is ±5 K.
To provide additional insight into gain reduction along the supersonic cavity, seven additional apertures with gain windows were made, distributed along the flow axis as described in Section 2.1. The results of gain measurements vs. axial distance are shown in Figure 3.17. Lower gain at the nozzle exit (x=0) is observed most likely because the iodine atom population inversion did not have sufficient time to reach equilibrium with excited oxygen after rapid supersonic expansion to low temperatures. It can be seen that gain reaches a maximum 3.2 cm downstream of the nozzle exit, and then rapidly falls off. Boundary layer separation due to shock-boundary layer interaction upstream of angle step supersonic diffuser inserts was assumed to be the main cause of gain fall-off. The diffuser could not be removed because one of the diffuser inserts had been effectively glued in place by iodine.
residue. Therefore, one of the diffuser inserts was milled flush with the cavity wall, and the other was left without a change, after which gain measurements have been repeated.

Figure 3.17: Gain vs. axial distance (with both diffuser inserts in place). 7 % O₂-He; 34 mmole/sec O₂, 446 mmole/sec He, 375 μmole/sec NO flow through the discharge section; p₀=86 torr after injection of 130 μmole/sec I₂ in 85 mmole/sec He; P₀=2.5 kW

The results are shown in Figure 3.18. Comparing Figure 3.17 and Figure 3.18, it can be seen that, although gain still decreases in the downstream region after one insert has been removed, it remains nearly constant over about 10 cm before the fall-off. Time-resolved static pressure traces, shown in Figure 3.19, show that when one diffuser insert is used, static pressure at the downstream location decreases compared to operation with both diffuser inserts, see Figure 3.1. On the other hand, removing one of the inserts did not significantly reduce steady-state run time nor affected static pressure at the upstream location.
Figure 3.18: Gain vs. axial distance (one diffuser insert used). 7% O₂-He; 34 mmole/sec O₂, 446 mmole/sec He, 375 μmole/sec NO flow through the discharge section; $p_0=80$ torr after injection of 130 μmole/sec I₂ in 85 mmole/sec He; $P_p=2.5$ kW

Since removal of one diffuser inserts had positive effect on axial gain distribution, the remaining insert was also milled flush with the supersonic cavity.
wall. Axial gain distribution measurements and time-resolved static pressure traces are shown in Figure 3.20 and Figure 3.21, respectively. It can be seen that, after diffuser removal, gain remains nearly constant, varying within 10% throughout the entire supersonic section, except at the nozzle exit where gain always remains slightly lower, as discussed above. After diffuser removal, static pressures measured at the upstream and the downstream locations have become nearly the same, \( p_1 = p_2 = 2.9 \) torr, indicating a flow Mach number of \( M = 2.87 \) at both locations. Run time at steady-state flow conditions remains approximately 5-7 seconds. Thus, the step angle diffuser did not help to increase the run time but significantly degraded flow quality in the supersonic section, most likely due to boundary layer separation in a low-pressure flow, thus increasing static pressure and reducing flow Mach number and gain. The apparatus have been operated without a diffuser for all subsequent experiments discussed in the present work.
Figure 3.20: Gain vs. axial flow distance (without diffuser). 7 % O₂-He; 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section, p₀=80 torr after injection of 150 µmole/sec I₂ in 78 mmole/sec He; Pₚ=2.75 kW

Figure 3.21: Time-resolved pressure traces (without diffuser). 7 % O₂-He; 34 mmole/sec O₂, 446 mmole/sec He flow through the discharge section, p₀=80 torr after injection of 78 mmole/sec He
3.7 Laser Output Power Measurements Without Diffuser

After the flow quality in the supersonic cavity was improved by the removal of the diffuser, laser output power measurements were repeated. During these measurements, significant divergence of the laser beam was detected. To provide access to a larger cavity volume, the aperture of the mirror bushing (on the non-reflective side of each mirror) was increased from a 1.27 cm to a 1.9 cm diameter, as discussed in greater detail in Section 2.1. Also, flanges with laser mirror mounts were attached directly to the cavity sidewalls, without the use of the extension arms, as shown in Figure 2.16. Laser power measurements were again made at the optimum gain conditions: 7 % O₂-He mixture; 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section; p₀=80 torr after injection of 150 µmole/sec I₂ in 78 mmole/sec He, Pₚ=2.75 kW. As before (see Section 3.5), laser action was achieved at the axial location 3.2 cm downstream of the nozzle exit, for the same 99.9%-99.9 % and 99 %–99.9 % mirror combinations used in the baseline power measurements in Section 3.5. Laser action was attempted, but not achieved, for a 99 %–99 % mirror combination. For each case, gain was measured at the resonator location immediately prior to mirror installation. Gain was also measured at several locations downstream of the resonator during lasing. The results of gain and laser output power measurements without diffuser are shown in Table 3.3.
Table 3.3: Laser Output Power Measurements Without Diffuser

<table>
<thead>
<tr>
<th>Mirror Combination</th>
<th>Gain prior to mirror installation (x=3.2) cm, %/cm</th>
<th>Laser output power</th>
<th>Gain during laser action, %/cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>99.9 %-99.9 %</td>
<td>0.112</td>
<td>2.5 W</td>
<td>0.085 0.090 0.082</td>
</tr>
<tr>
<td>99 %-99.9 %</td>
<td>0.106</td>
<td>3.1 W</td>
<td>0.100 0.100 -</td>
</tr>
</tbody>
</table>

For the 99.9-99.9 \% mirror combination, power coupled out on both sides of the resonator were approximately equal (1.2-1.3 W), while power was coupled almost entirely out of the side of a 99 \% mirror for the 99-99.9 \% mirror combination. The increased laser output power recorded during these measurements, compared to the baseline laser output power measurements (discussed in Section 3.5) was attributed to a larger aperture in the laser mirror bushings. Again, insignificant gain reduction downstream of the laser resonator during lasing, compared to gain at the resonator location without lasing, demonstrates that only a small fraction of the power stored in \(O_2(\text{^1}\Delta)\) and available for lasing is coupled out. High gain measured downstream of the upstream resonator during lasing shows significant potential for achieving laser action with a second, downstream resonator for each of the mirror combinations used. Gain measured downstream of the resonator during lasing is slightly lower for the 99.9 \%-99.9 \% mirror combination than for the 99-99.9 \% combination, in spite of achieving a lower laser output power (see Table 3.3). This suggests that a resonator with a higher reflectivity output coupler is extracting more power from the flow, but coupling less out in the output laser beam due to higher resonator losses. This is
expected, because power loss in the resonator is proportional to the intra-cavity radiation intensity, which is much greater for a resonator with a higher reflectivity output coupler. Intraresonator radiation intensity can be estimated as

\[ I_i = \frac{I_o}{1 - R}, \]  

(69)

where \( I_i \) is the intraresonator field intensity, \( I_o \) is the power density of the laser output beam from a particular mirror, and \( R \) is the mirror reflectivity. Using this approximation, the intraresonator radiation intensity is about a factor of 4 greater for the resonator using a 99.9 % output coupler, compared to the resonator using two 99.99% output couplers.

Typical time-resolved thermopile calorimeter voltage outputs at the conditions of Table 3.3 are shown in Figure 3.22, along with power calibration fits. From Figure 3.22, it can be seen that output power for the 99.9-99.9 % mirror combination remains constant for more than 10 seconds (consistent with time-resolved gain measurements, shown in Figure 3.15). On the other hand, output power for the 99-99.9 % mirror combination remains constant for only 2-3 seconds, after which it begins to decrease. The time dependence of laser output power for each mirror combination was reproducible over multiple runs conducted over several days. The laser power falloff for the lower reflectivity mirror combination remains not fully understood.
Figure 3.22: Calorimeter voltage output and laser power calibration fits at the conditions of Table 3.3. Best fits indicate 3.1 W laser power output from 99 % output coupler using 99-99.9 % mirror combination, and 1.1 W laser power output from 99.9 % output coupler using 99.9-99.9 % mirror combination.

Finally, to study possibility of laser power extraction using multiple resonators operating simultaneously, laser power was measured in two resonators. The resonators were set to operate simultaneously using 1.27 cm diameter apertures located 3.2 and 12.7 cm downstream of the nozzle exit. Each resonator used a 99-99.9 % mirror combination. Simultaneous laser action in both resonators was achieved at the following flow and discharge conditions: 7 % O₂-He mixture; 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section; p₀=80 torr after injection of 150 µmole/sec I₂ in 78 mmole/sec He, Pₚ=2.75 kW. At these conditions, 2.9 and 0.9 W were extracted from the upstream and downstream resonators, respectively, with a combined laser output power of 3.8 W. Gain measurements made 9.6 cm downstream of the nozzle exit (i.e. between the
two resonators), showed gain of \( \gamma = 0.1 \% / \text{cm} \) while lasing. It can be seen that significantly lower power was extracted from the downstream resonator, in spite of similar gain measured at both locations (see Figure 3.20). It was suspected that boundary layer growth on the top and bottom walls of the supersonic cavity may reduce the thickness of the low-temperature inviscid core flow. This effect would reduce the size of a high-gain region due to a higher temperature in the boundary layers, even at the conditions when gain on the flow centerline remains nearly constant.

### 3.8 Gain Mapping and CFD

To understand better the effect of boundary layers on laser performance, further gain mapping experiments were conducted in conjunction with CFD modeling of the flow in the supersonic cavity. The flow modeling was conducted using a 3-D compressible Navier-Stokes flow code described in detail in reference (77). In the present work, one quarter of the rectangular cross section supersonic test section was modeled, from the nozzle throat to the end of the test section, 14.4 cm downstream of the nozzle exit, using a 101 x 61 x 51 grid clustered near the walls for greater boundary layer resolution. The conditions modeled were 12 \% \( \text{O}_2 \) mole fraction in \( \text{He} \), at a stagnation pressure and temperature of \( p_0 = 80 \) torr and \( T_0 = 380 \) K, respectively, i.e. close to baseline experimental conditions. The upstream boundary condition at the nozzle throat assumed a uniform \( M = 1 \) flow. The downstream boundary conditions assumed zero temperature, pressure, and velocity gradients at the supersonic section exit. The boundary conditions at the
planes of symmetry of the flow channel assumed zero temperature, pressure, and velocity gradients. The boundary conditions at the flow channel walls were zero temperature and pressure gradient (adiabatic wall and negligible pressure gradient across the boundary layer), and zero velocity (no-slip condition).

Static pressure on the top/bottom walls of the test section, predicted by the model, is plotted in Figure 3.23, along with experimental pressure measurements. It can be seen that the model somewhat overpredicts the pressure in the upstream portion of the test section. Mach number contours are shown in Figure 3.24. Note that the top and bottom walls diverging at 1.5° each provides adequate relief for the growing boundary layers, such that the core flow height remains nearly constant, \( h \approx 6 \) mm. The Mach number in the core flow increases modestly with axial distance, correlating with the static pressure reduction. The predicted centerline flow Mach number is \( M=2.66 \) and \( M=2.86 \) at \( x=3.2 \) cm and \( x=12.7 \) cm, in good agreement with the valued inferred from the static pressure measurements, \( M=2.87 \) and \( M=2.87 \), respectively.
Figure 3.23: Static pressure distribution along the top/bottom walls of the supersonic test section, compared with experimental static pressure data. Experimental data points are taken for following flow conditions: 7% O₂-He; 34 mmole/sec O₂ and 446 mmole/sec He flow through the discharge section; \( p_0 = 81 \) torr after injection of 85 mmole/sec He; \( T_0 = 300 \) K (discharge off).

Figure 3.24: Mach number contours in the supersonic cavity. Axial distance given in mm from the nozzle throat. Nozzle exit is 32 mm downstream of the nozzle throat. 12% O₂-He, \( p_0 = 80 \) torr, \( T_0 = 380 \) K.

Gain mapping measurements were taken across the vertical span of the optical access windows located 3.2 and 12.7 cm downstream of the nozzle exit, as
shown in Figure 3.25. Flow temperature inferred from the gain lineshapes using a Doppler lineshape fit, together with the static temperature predicted by the flow code on the center plane of symmetry, is shown in Figure 3.26. Since the temperature in the boundary layers is significantly higher than in the supersonic core flow, gain falls off and changes to absorption in these regions, as expected. However, note that the gain in the boundary layers decreases much faster compared to the temperature rise, indicating that gain reduction cannot be explained solely by the effect of temperature on the equilibrium constant of reaction of Eq. (16). Also note that the positive gain region is much thicker at the upstream location, partially explaining the higher laser output power measured in the upstream resonator (see Table 3.3). The modeling calculations somewhat underestimate the temperature throughout the supersonic channel but capture the trend of moderate temperature reduction at the downstream location (see Figure 3.26).
Figure 3.25: Gain distribution in the supersonic cavity along the vertical coordinate, measured at two different axial locations. 7% O$_2$-He; 34 mmole/sec O$_2$, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section; $p_0=80$ torr after injection of 150 µmole/sec I$_2$ in 78 mmole/sec He; $P_p=2.75$ kW

Figure 3.26: Experimental and calculated temperature distributions in the supersonic cavity at two different axial locations, at the conditions of Figure 3.25
3.9 Temperature Inference from Gain Lineshape

In the present work, flow temperature in the supersonic test section is inferred from the experimental gain lineshape, such as shown in Figure 3.16, approximated with a Doppler lineshape, given by Eq. (22).

The effect of pressure broadening on gain lineshape was not incorporated in the temperature inference fit. As discussed in Section 1.2, the combined effect of Doppler and collision (pressure) broadening results in gain/absorption lineshape given by the Voigt profile (Eq. (26)). The difference between the Doppler and Voigt lineshape at the present conditions is illustrated in Figure 1.6. The Voigt lineshape plotted in Figure 1.6 has been calculated for a 10 % O₂-He mixture at a temperature of T=100 K and a pressure of p=3 torr. At these conditions, the collisional broadening is Δνᵣ=17.6 MHz, the Doppler broadening is Δνᵣ=145.6 MHz, and the Voigt broadening is Δνᵣ=155.2 MHz (see Eqs. (22-26)). The Voigt lineshape shown in Figure 3.27 is approximated with a “best fit” Doppler lineshape. The ‘best fit’ temperature inferred using this approach is T=112 K, i.e. twelve degrees higher than the temperature of T=100 K used to calculated the Voigt profile. As can be seen, at the present conditions the effect of pressure broadening on temperature inference from the gain lineshape is relatively modest, about 12 %, slightly exceeding the estimated uncertainty in temperature inference using a ‘best fit’ Doppler lineshape, ± 5 K. The result is expected since the ratio of pressure broadening to Doppler broadening at the present conditions is fairly small, Δνᵣ/Δνᵣ=0.12.
Figure 3.27: Voigt gain lineshape with 'best fit' Doppler lineshape. Voigt lineshape calculated for 10 \% O\textsubscript{2} in He at a temperature and pressure of T=100 K and p=3 torr (\(\Delta\nu_{c}=17.6\) MHz, \(\Delta\nu_{D}=145.6\) MHz, \(\Delta\nu_{V}=155.2\) MHz). A temperature of T=112 K is inferred using a Doppler lineshape fit.

Note that the measured lineshape function is spatially averaged over the line of sight, i.e. the entire 10 cm width of the supersonic flow channel, including boundary layers on the side walls. Higher temperature supersonic boundary layers, in which gain switches to absorption, as shown for the top and bottom wall boundary layers in Figure 3.25 and Figure 3.26, may well affect the inferred temperature value. Similar boundary layers also develop on the side walls, thus affecting all experimental lineshapes. To estimate the boundary layer effect on the temperature inferred from gain lineshapes, we assume a 0.5 cm thick boundary layer on each side wall with a temperature of T=325 K and a gain (absorption) of \(\gamma=-0.04\) \%/cm, i.e. similar to the results shown in Figure 3.25 and Figure 3.26. The supersonic core is assumed to be 9 cm wide with a static temperature of T=130 K and gain of \(\gamma=0.12\) \%/cm, again similar to the results of Figure 3.25 and Figure 3.26.
Doppler lineshape contributions from each of these two flow regions, along with a superposition of these contributions weighted proportional to their total single-pass gain/absorption, values as well as a Doppler fit to the superposition lineshape, are shown in Figure 3.28. As can be seen from Figure 3.28, the temperature inferred from the Doppler lineshape fit to the superposition lineshape is T=126 K, i.e. several degrees lower than the core flow temperature. This is caused by the negative contributions of the boundary layers into the lineshape, because gain switches to absorption in the boundary layers. Thus, “apparent” gain for the conditions of Figure 3.28 would be γ=0.104 %/cm, approximately 15 % less than the gain of γ=0.12 %/cm in the supersonic core. Also note that, due to this effect, the “apparent” (i.e. line of sight averaged) gain is lower than actual gain in the supersonic core flow. However, the boundary layer effects at the present conditions are relatively minor, of the order of ~10 %.
Uncertainties in flow temperature and small signal gain can be reduced by using “bank blower” nozzles producing helium “curtain” flow near the side walls of the laser cavity, such as have been used in our previous work. Indeed, helium boundary layers formed in the curtain flows would not affect the overall gain. Compared to our previous, small scale DOIL apparatus, boundary layer effects play a less significant role in the scaled DOIL with a longer gain path.

3.10 Gain Measurements Using Two Consecutive Discharge Sections

For these measurements, a second transverse capacitively coupled RF discharge section, identical to the first test section discussed in Section 2.1, was manufactured and installed between the flow conditioning section and the first discharge section, as shown in Figure 2.1. The two discharge sections are referred to as the upstream and downstream discharges. Adding the second discharge section
allowed power loading over a larger discharge volume, thus reducing the current
density (due to larger electrode surface area) and presumably reducing the reduced
electric field at the same discharge power. Lower values of the reduced electric field
were expected to increase the O\textsubscript{2}(^1\Delta) generation efficiency, e.g. see Figure 1.9. When
operating simultaneously, the two discharges were connected in parallel and
powered by the same RF power supply, as described in Section 2.1 and shown in
Figure 2.7.

Infrared emission spectroscopy measurements conducted for three different
discharge configurations (upstream alone, downstream alone, and simultaneous
operation) showed O\textsubscript{2}(^1\Delta) yield to be similar for all of them. The results of gain
measurements during operation of the downstream discharge alone and during
simultaneous operation of both discharges are shown in Figure 3.29. It can be seen
that simultaneous operation of both discharges leads to somewhat reduced gain in
the entire range of discharge powers. From Figure 3.29, it can be seen that during
simultaneous operation of both discharge sections, gain fall-off begins at lower RF
discharge powers (P\textsubscript{p}=3.0 kW for two discharge sections compared to P\textsubscript{p}=3.5 kW for
one section), and peak gain is somewhat reduced (γ=0.1 %/cm compared to γ=0.11
%/cm). Doppler fits to gain lineshapes in the supersonic cavity indicate higher flow
temperatures attained during simultaneous discharge operation, consistent with
lower gain measured at these conditions in spite of similar O\textsubscript{2}(^1\Delta) yields for both
discharge configurations. Gain was not measured while operating only the upstream
discharge section.

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Figure 3.29: Gain vs. power for one and two RF discharge sections in operation. 7% O₂-He; 34 mmole/sec O₂, 446 mmole/sec He, 375 µmole/sec NO flow through the discharge section, p₀=80 torr after injection of 130 µmole/sec I₂ in 80 mmole/sec He; gain measured x=3.2 cm downstream of the nozzle exit

3.11 Gain and Infrared Emission Spectroscopy Measurements in O₂-He-Ar Mixtures

A series of experiments were undertaken when argon was added to the main O₂-He-NO flow mixture. During these measurements, flow from an argon cylinder was started by a remotely controlled solenoid valve and mixed with the main flow in the 1 inch diameter delivery line about 5 meters upstream of the apparatus discharge section (nozzle plenum). While the main O₂ flow rate was held constant at 34 mmole/sec and the plenum pressure was held constant at p₀=80 torr, helium flow rate was gradually reduced and replaced with argon. Due to its greater molecular weight, argon flow increases plenum pressure more significantly, compared to an equal mole flow rate of helium. Because of this, with both the
oxygen flow rate and the plenum pressure remaining constant, the total flow rate through the discharge section dropped (from 480 to 250 mmole/sec) and the $O_2$ mole fraction in the mixture increased (from 7 % to 13.5 %), as the Ar mole fraction in the mixture increased. During these experiments, gain was measured in the supersonic cavity 3.2 cm downstream of the nozzle exit. Gain vs. Ar mole fraction in the mixture, measured at the discharge power of 2.75 kW, is shown in Figure 3.30. During these measurements, only the downstream discharge section was powered.

![Gain vs. Ar mole fraction in the flow. 34 mmole/sec $O_2$, 117-446 mmole/sec He, 0-98 mmole/sec Ar, 375 µmole/sec NO flow through the discharge section, $p_0$=80 torr after injection of 130 µmole/sec I$_2$ in 75 mmole/sec He; $P_p$=2.75 kW (downstream primary discharge), x=3.2 cm](image)

As can be seen, adding argon to the $O_2$-He-NO flow results in dramatic gain reduction until it changes to absorption when argon mole fraction exceeds 20 %. Further measurements demonstrated that the drastic gain reduction when argon is added to the flow is due to reduced $O_2(^1\Delta)$ yield. To illustrate this, infrared emission spectra near 1.3 µm are shown in Figure 3.31 for Ar mole fractions of 0 and 39 %. It
can be seen that adding argon to the flow results in dramatic reduction of \( \text{O}_2(\Delta \rightarrow \Sigma, 0 \rightarrow 0) \) band emission intensity. At the same time, multiple argon emission lines appear in the spectra, with some of them identified in the figure. This result is somewhat unexpected, since kinetic modeling suggests that a significantly higher discharge energy fraction is spent on \( \text{O}_2(\Delta) \) generation in \( \text{O}_2\text{-Ar} \) mixtures compared with \( \text{O}_2\text{-He} \) mixtures (22), as shown in Figure 3.32. However, presence of several Ar emission transitions nearly isoenergetic with that of \( \text{O}_2(\Delta \rightarrow \Sigma) \) suggest rapid collisional energy transfer from \( \text{O}_2(\Delta) \) state to excited Ar atoms, thereby depleting it. Presence of Ar emission lines in \( \text{O}_2\text{-He} \) mixture without argon indicates a leak in the solenoid valve in the Ar line, which was subsequently removed.

![Figure 3.31: Infrared emission spectra taken with and without adding argon to the main \( \text{O}_2\text{-He} \) flow. \( \text{O}_2(\Delta \rightarrow \Sigma, 0 \rightarrow 0) \) emission band and Ar emission lines are labeled. 34 mmole/sec \( \text{O}_2 \), 117 or 446 mmole/sec \( \text{He} \), no Ar flow or 98 mmole/sec \( \text{Ar} \), 375 \( \mu \)mole/sec NO flow through the discharge section, \( p_0=80 \) torr after injection of 75 mmole/sec \( \text{He} \); \( P_p=2.75 \) kW (downstream primary discharge)](image)
Visible emission spectra showing the $O_2(1\Sigma \rightarrow 3\Sigma, 0\rightarrow 0)$ emission band are plotted in Figure 3.33 for Ar mole fractions of 5% and 35%. As in Figure 3.31, it can be seen that with 35% Ar in the flow, $O_2(1\Sigma \rightarrow 3\Sigma)$ band emission is barely detectable. Again, adding more argon to the flow significantly increases Ar emission line intensities (identified in the figure, along with an O atom emission line at $\lambda=777$ nm). Note that strong absorption detected at high Ar mole fractions in the flow (see Figure 3.30) indicates significant iodine dissociation fraction without significant amounts of singlet oxygen present in the flow, so $I_2$ dissociation at these conditions must occur by another kinetic mechanism, likely involving O atoms generated in the discharge.

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9 Reproduced from reference (22)
Figure 3.33: Visible emission spectra taken for two different mole fractions of argon in the O₂-He-NO flow. O₂(1Σ→3Σ,0→0) emission band as well as Ar and O atom emission lines are labeled. 34 mmole/sec O₂, 117 or 350 mmole/sec He, 22 or 98 mmole/sec Ar, 375 µmole/sec NO flow through the discharge section, p₀=80 torr after injection of 75 mmole/sec He; Pₚ=2.75 kW (downstream primary discharge)

3.12 Gain Measurements with Chilled Flow Injection

The results of gain measurements taken up to this point suggest that one of the main factors controlling gain in the supersonic cavity is the flow temperature. To reduce it, a chilled flow injector was installed between the iodine injector and the supersonic nozzle, as shown in Figure 3.35. Briefly, helium or nitrogen flow was chilled in a liquid nitrogen bath and injected into the main flow in the subsonic section to reduce the flow temperature. The precooled flow injection has an additional effect of increasing plenum pressure due to a higher flow rate through the laser.

An estimate of injection flow temperature exiting the flow chiller can be made using equations for a fully developed thermal boundary layer in a circular
tube L=4 m long, with an inner diameter of D=6.35 mm and a constant wall temperature of T_s=77 K. Assuming a helium flow rate of \( \dot{n} = 260 \) mmole/sec at a chiller inlet temperature of T_i=300 K and pressure of p=500 torr, the Reynolds number and Prandtl are Re_D=1.05 \cdot 10^4 and \( \text{Pr} = 0.68 \), respectively. Assuming a fully developed turbulent flow for Re_D \geq 10^4 and L/D \sim 600, the Nusselt number is given by the Dittus-Boelter correlation,

\[
\text{Nu}_D = \frac{hD}{k} = 0.023 \text{Re}_D^{0.8} \text{Pr}^{0.3} = 33.8,
\tag{70}
\]

where \( h \) is the convection heat transfer coefficient, and \( k \) is the thermal conductivity of helium. The temperature of the flow exiting the chiller is

\[
T_e = T_s + (T_i - T_s) \exp \left( \frac{-\pi DLh}{hC_p} \right)
\tag{71}
\]

where \( h \) is evaluated using the Nusselt number given in Eq. (70). The average thermal conductivity of helium, \( \bar{k} \), used in Eq. (70) must be evaluated at the log mean temperature

\[
T_{\text{lm}} = T_s + \frac{T_i - T_e}{\ln \left( \frac{T_i - T_e}{T_e - T_s} \right)}.
\tag{72}
\]

For the given conditions, the log mean temperature is about \( T_{\text{lm}} = 100 \) K, the average thermal conductivity is \( \bar{k} = 73 \cdot 10^{-3} \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1} \), the average convective heat transfer
coefficient is \( \bar{h} = 388 \text{ W} \cdot \text{m}^{-2} \cdot \text{K}^{-1} \), and the exit temperature is \( T_e = 77.8 \text{ K} \), essentially the same as the temperature of liquid nitrogen.

Main flow temperature reduction can be estimated using a simple energy balance equation,

\[
\frac{T_2}{T_1} = \frac{1 + \chi T_1}{1 + \chi T_1} \tag{73}
\]

where

\[
\chi = \frac{\dot{n}_1 C_{p_1}}{\dot{n}_{\text{He}C_{p,\text{He}}} + \dot{n}_{\text{O}_2} C_{p\text{O}_2}} \tag{74}
\]

is the ratio of heat capacities of the two flows. In Eqs. (73,74) \( T_1 \) is the \( \text{O}_2\text{-He} \) flow temperature without precooled flow injection, \( T_2 \) is the flow temperature after injection, \( T_i \) is the temperature of the precooled flow, \( \dot{n}_1 \) is its mole flow rate, \( C_{p_1} \) is the precooled gas specific heat at constant pressure, \( \dot{n}_{\text{He}} \) and \( \dot{n}_{\text{O}_2} \) are \( \text{He} \) and \( \text{O}_2 \) mole flow rates in the main flow at the precooled flow injection point (including the \( \text{He} \) flow injected through the iodine vapor injector, see Figure 3.35), respectively, and \( C_{p,\text{He}} = 20.79 \text{ [J/(mole \cdot K)]} \) and \( C_{p,\text{O}_2} = 29.38 \text{ [J/(mole \cdot K)]} \) are \( \text{He} \) and \( \text{O}_2 \) specific heats, respectively. Note that nitrogen has a higher specific heat at constant pressure compared to helium, \( C_{p,N_2} = 29.12 \text{ [J/(mole \cdot K)]} \), leading to a more significant main flow temperature reduction at the same precooled flow injection.
flow rate. On the other hand, $N_2$ injection in the subsonic section increases plenum pressure more significantly compared to He injection, due to its higher molecular weight. Nitrogen injection also somewhat reduces the Mach number in the supersonic cavity due to its lower specific heat ratio. At the optimum gain conditions, 7% $O_2$-He, 34 mmole/sec $O_2$, 446 mmole/sec He flow through the discharge section, 80 mmole/sec He injected through the iodine vapor injector, at an estimated main flow temperature of $T_1=380$ K, an injection flow of 250 mmole/sec of He chilled to liquid nitrogen temperature of $T=77$ K would reduce the main flow temperature in the subsonic section by 24%, to $T_2=288$ K, thus offsetting heating by the discharge. This would also reduce the static temperature in the supersonic flow in the laser cavity from an estimated $T=125$ K to $T=95$ K.

![Figure 3.34: DOIL Schematic](image-url)
Gain in the supersonic cavity vs. chilled He injection flow rate is shown in Figure 3.36 for the optimum gain conditions achieved without cooling the flow (see Section 3.10). Note that full scale injection increases the plenum pressure by 44 %, from $p_0=82$ torr to $p_0=118$ torr and decreases the supersonic flow temperature by 21 %, from $T=120$ K to $T=95$ K, consistent with the temperature reduction estimate above. This is also accompanied by gain increase by almost 60 %, from $\gamma=0.093$ %/cm to $\gamma=0.147$ %/cm (see Figure 3.36).
This result demonstrates that injecting cold helium into the laser mixture both significantly reduces the flow temperature and substantially increases gain. After this, the discharge parameters were re-optimized to achieve optimum gain while operating with the chilled flow injector. Gain vs. the discharge power for four different plenum pressures is shown in Figure 3.37, and gain vs. O₂ mole fraction in the mixture is shown in Figure 3.38. Based on the results shown in Figure 3.37 and Figure 3.38, optimum gain, \( \gamma = 0.188 \% / \text{cm} \), was achieved at the following flow and discharge conditions: 7 \% O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 \( \mu \text{mole/sec} \) NO flow through the discharge section, \( p_0 = 93 \) torr after injection of 110 \( \mu \text{mole/sec} I_2 \) in 40 mmole/sec He and 260 mmole/sec chilled He, \( P_p = 2.75 \text{ kW} \). In these experiments, gain was measured 3.2 cm downstream of the nozzle exit.
Figure 3.37: Gain vs. discharge power at different main flow rates. 7% O$_2$-He; 17-34 mmole/sec O$_2$, 223-446 mmole/sec He, 375 µmole/sec NO flow through the discharge section, $p_0=85$-114 torr after injection of 110 µmole/sec I$_2$ in 40 mmole/sec He and 260 mmole/sec chilled He; (downstream primary discharge); $x=3.2$ cm

Figure 3.38: Gain vs. O$_2$ mole fraction in the flow through the discharge. 2.5-14 % O$_2$-He (14-59 mmole/sec O$_2$, 353-543 mmole/sec He, 375 µmole/sec NO flow through the discharge section, $p_0=93$ torr after injection of 110 µmole/sec I$_2$ in 40 mmole/sec He and 260 mmole/sec chilled He; $P_p=2.75$ kW (downstream primary discharge); $x=3.2$ cm
Gain measurements at different locations in the supersonic cavity have shown that gain varies moderately along the supersonic cavity (see Figure 3.17), with the highest gain of $\gamma=0.209 \%$/cm measured at $x=9.6$ cm downstream of the nozzle exit. Figure 3.39 shows time-resolved gain measured at this location. As can be seen, nearly steady gain of $\gamma=0.16-0.20 \%$/cm is maintained for about 8 seconds.

![Time-resolved gain in the supersonic test section](image)

Figure 3.39: Time-resolved gain in the supersonic test section measured 9.6 cm downstream of the nozzle exit (with chilled He flow injection). 7 % O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section, $p₀=93$ torr after injection of 140 µmole/sec I₂ in 40 mmole/sec He and 260 mmole/sec chilled He; $P₀=3.0$ kW (downstream primary discharge); $x=9.6$ cm

Attempts to increase gain by replacing chilled He injection with chilled N₂ injection were unsuccessful. A series of gain measurements while injecting precooled or room temperature He or N₂ was made at the following baseline flow conditions: 7 % O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; 95 µmole/sec I₂ in 45 mmole/sec He injection, $P₀=3.0$ kW. The actual discharge pressure after injection, $p₀$, varied depending on
which flow was added through the chilled flow injector. Injection rates of (a) 260 mmole/sec of chilled or room temperature He and (b) 200 mmole/sec of chilled or room temperature N₂ were tested, as well as a control case with no extra injection. First, measurements were made with the downstream primary discharge section powered, with the results shown in Table 3.4. As expected, injection of precooled flows (either He or N₂) increases gain significantly compared to injection of room temperature He or N₂, respectively. Cold helium injection results in significantly higher gain compared to cold nitrogen injection in spite of more significant flow temperature reduction estimated for cold N₂ flow addition (see Eqs. (73,74)). Finally, nearly zero gain was measured without any injection through the precooled flow injector. These results demonstrate that gain is controlled not only by the flow temperature, but may also be affected by flow mixing in the subsonic section.

Table 3.4: Gain in the Supersonic Section for Different Flows Injected Through the Chilled Flow Injector (with the Downstream Discharge Section Powered)

<table>
<thead>
<tr>
<th>Injection</th>
<th>Gain, %/cm</th>
<th>p₀, torr (after injection)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold He</td>
<td>0.160</td>
<td>93</td>
</tr>
<tr>
<td>Room Temperature He</td>
<td>0.077</td>
<td>118</td>
</tr>
<tr>
<td>Cold N₂</td>
<td>0.074</td>
<td>108</td>
</tr>
<tr>
<td>Room Temperature N₂</td>
<td>0.011</td>
<td>136</td>
</tr>
<tr>
<td>No Injection</td>
<td>-0.007</td>
<td>50</td>
</tr>
</tbody>
</table>

Table 3.5: Gain in the Supersonic Section for Different Flows Injected Through the Chilled Flow Injector (with the Upstream Discharge Section Powered)

<table>
<thead>
<tr>
<th>Injection</th>
<th>Gain, %/cm</th>
<th>p₀, torr (after injection)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold He</td>
<td>0.088</td>
<td>93</td>
</tr>
<tr>
<td>Room Temperature He</td>
<td>0.031</td>
<td>118</td>
</tr>
<tr>
<td>Cold N₂</td>
<td>0.052</td>
<td>108</td>
</tr>
<tr>
<td>Room Temperature N₂</td>
<td>0.018</td>
<td>136</td>
</tr>
<tr>
<td>No Injection</td>
<td>0.075</td>
<td>50</td>
</tr>
</tbody>
</table>
Indeed, significant injection flow rates (on the order of the flow rate of the main flow) may well create a complicated flow field in the subsonic section, affecting the iodine vapor mixing with the main flow and possibly creating flow separation/recirculation regions where the injection flows penetrate upstream into the discharge. Repeating these measurements with the upstream discharge section powered, with the results summarized in Table 3.5, further complicated the analysis. These results show much higher gain without precooled or room temperature buffer flow injection (compare with Table 3.4), but much lower gain when adding precooled or room temperature buffer flow. Higher gain measured with the downstream discharge section powered suggest possible interaction between the discharge and the I$_2$-He flow injected through an aluminum injector between the discharge section and the buffer (precooled or room temperature) flow injector (see Figure 3.34). However, this hypothesis remains fairly speculative and requires additional measurements to be confirmed or refuted. Until additional data is obtained on the effect of buffer flow injection, as well as discharge and injector geometry on gain in a supersonic cavity, the decision was made to proceed with laser power measurements at the conditions when highest gain is achieved, i.e. with precooled He flow injection, with the downstream discharge section powered.

### 3.13 Laser Output Power Measurements Following Chilled Flow Injection

Laser output power was measured at the new optimum gain conditions determined in Section 3.12: 7 % O$_2$-He; 22 mmole/sec O$_2$, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section, p$_0$=93 torr after adding 110
µmole/sec I₂ in 40 mmole/sec He carrier and 260 mmole/sec chilled He, \( P_p = 3.0 \text{ kW} \).

The laser resonator operated 3.2 cm downstream of the nozzle exit. The laser mirror mount flanges were attached directly to the supersonic section side walls, without the use of extension arms, as shown in Figure 2.16. As before (see Section 3.7), gain was measured at the resonator location immediately before laser mirror mount installation, to verify that flow and discharge parameters were optimized. To estimate gain recovery, gain was also measured 9.6 cm downstream of the nozzle exit (i.e. 6.4 cm downstream of the laser resonator) while laser action occurred, and without laser action (for this, the resonator was deliberately misaligned). The results of the laser output power and gain recovery measurements are shown in Table 3.6. Note that the 99 % and 99.9 % mirrors are concave, with the curvature radius of 1 m, while the 98.4 % mirror is plane. For runs with the 99.9-99.9 % mirror combination, laser output powers measured on both sides of the resonator were approximately equal. For the 99-99.9 % and 98.4-99.9 % mirror combinations, laser output power was coupled almost entirely on the side of the lower reflectivity mirror.

<table>
<thead>
<tr>
<th>Laser mirrors</th>
<th>Gain before lasing, %/cm</th>
<th>Laser power, W</th>
<th>Gain downstream without laser action, %/cm</th>
<th>Gain downstream with laser action, %/cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>99.9%-99.9%</td>
<td>0.173</td>
<td>2.5 W</td>
<td>0.202</td>
<td>0.156</td>
</tr>
<tr>
<td>99%-99.9%</td>
<td>0.174</td>
<td>7.8 W</td>
<td>0.198</td>
<td>0.155</td>
</tr>
<tr>
<td>98.4%-99.9%</td>
<td>0.181</td>
<td>6.7 W</td>
<td>0.209</td>
<td>0.150</td>
</tr>
</tbody>
</table>
From Table 3.6, it can be seen that laser output power achieved with the 99.9-99.9 % mirror pair was $P_L=2.5$ W, the same as measured without adding chilled helium flow (see Section 3.7), in spite of significantly higher gain with chilled helium added, $\gamma=0.173 \text{ %/cm}$ compared to $\gamma=0.112 \text{ %/cm}$. The highest laser output power achieved with adding cold buffer is $P_L=7.8$ W using the 99-99.9 % mirror combination, which is more than a factor of 2 greater than measured without the chilled He injection (see Section 3.7). For all three cases in Table 3.6, gain measured downstream of the resonator during lasing remained relatively high, above $\gamma=0.15 \text{ %/cm}$, which is only about 25 % lower than gain without lasing. This shows potential for coupling out additional power, using a second resonator located 12.7 cm downstream of the nozzle exit, as has been done in Section 3.7. However, an alternative approach to access a larger laser cavity volume using a folded resonator (see Section 2.1) was chosen instead (see Section 3.15 below).

A quasi-one-dimensional kinetic model developed in Ref. (78) has been used to predict laser power extracted from a transverse resonator at the present conditions. In the model, $O_2(^3\Sigma)$ and $O_2(^1\Delta)$ populations, $n_{O_2(^3\Sigma)}$ and $n_{O_2(^1\Delta)}$, ground state and excited iodine atom populations, $n_i$ and $n_{i^*}$, and the photon flux, $I [\text{cm}^{-2}\cdot\text{s}^{-1}]$, in the resonator cavity are governed by the following equations,

\begin{align}
    u \frac{dn_{O_2(^1\Delta)}}{dx} &= -k_f n_i n_{O_2(^1\Delta)} + k_r n_{i^*} n_{O_2(^3\Sigma)}, \quad (75)
\end{align}
where $x$ is the axial distance, $u=1400$ m/s is the flow velocity in the cavity, $k_f$ and $k_r$ are the forward and the reverse rates of reaction of Eq. (16), $\sigma$ is the absorption/ stimulated emission cross section, $L=10$ cm is the gain path, and $R_1=0.99$ and $R_2=0.999$ are the mirror reflectivities. The laser output power per unit cross sectional area of the laser beam is

$$P = \varepsilon[(1 - R_1) + (1 - R_2)]I$$  \hspace{1cm} (78)
corresponding to steady-state resonator conditions). The laser power predicted by
the model is significantly higher than measured in the present work, \( P_L = 7.8 \text{ W} \) (see Table 3.6), likely due to significant resonator losses or poor mirror alignment.

3.14 Precooling the Flow Through the Discharge

Successful experiments on gain increase by injecting a precooled flow into
the laser mixture suggested that precooling the main flow through the discharge
section may produce a similar effect. For this, the main flow chiller was installed
upstream of the discharge section, as is shown in Figure 3.40. First, gain
measurements were made using a liquid nitrogen cold bath at the following
conditions: 7% \( \text{O}_2\)-\( \text{He} \); 34 mmole/sec \( \text{O}_2 \), 446 mmole/sec \( \text{He} \), 375 \( \mu \)mole/sec \( \text{NO} \)
flow through the discharge section; \( p_0 = 80 \text{ torr} \) after injection of 100 \( \mu \)mole/sec \( \text{I}_2 \) in
90 mmole/sec \( \text{He} \); \( P_p = 2.75 \text{ kW} \). Gain measurements using a liquid nitrogen bath
showed absorption in the entire range of conditions tested. Since boiling points of
\( \text{NO} \) and \( \text{O}_2 \) are \( T = 122 \text{ K} \) and \( T = 90 \text{ K} \), respectively, i.e. significantly higher than
temperature of the liquid nitrogen bath, \( T = 77 \text{ K} \), it is likely that a substantial fraction
of one or both of these species may have condensed in the chiller.
Then the liquid nitrogen bath was replaced with a liquid nitrogen-ethanol bath with the temperature of $T=157$ K, higher than the boiling point of all species in the main flows such that condensation would not occur. In this case, gain did not increase as a result of main flow chilling and gain lineshapes showed no detectable reduction of temperature in the supersonic section compared to operation without the main flow chiller. It was concluded that liquid nitrogen-ethanol bath was not sufficiently cold to reduce the main flow temperature significantly. An alternative possibility of chilling the main He flow in a liquid nitrogen bath, and subsequently mixing with the $O_2$ and NO flow upstream of the discharge was discussed but not pursued. Proper mixing of the precooled helium and room temperature oxygen would require a sufficiently long delivery line, insulated to prevent heating of the mixture before it enters the discharge section.
3.15 Laser Output Power Measurements with a Z-pass Resonator

To access a larger volume of the laser cavity and to increase gain path, a folded, triple pass, “Z-shaped” laser resonator, shown in Figure 2.17, was designed and tested. This was motivated primarily by the results of gain recovery measurements (see Table 3.3 and Table 3.6), which show only modest gain reduction downstream of the laser resonator during lasing. The triple-pass folded resonator has a total gain pass of 30.8 cm, a factor of 3 greater than simpler transverse resonators used for laser power measurements in Sections 3.7 and 3.13.

The resonator was tested at the following optimum gain conditions: 7 % O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 μmole/sec NO flow through the discharge section, p₀=93-95 torr after injecting 90-110 μmole/sec I₂ in 45 mmole/sec He carrier and 255 mmole/sec chilled He injection; Pₚ=3.25 kW. To measure gain at the resonator location before lasing, acrylic plastic inserts with wedged and AR-coated gain windows were installed instead of the mirror mounts at the mirror positions. This was done because the new sidewalls accommodating the mirror mount flanges are significantly thicker compared to the side walls used in all previous gain and laser power measurements, 4 cm thick vs. 2 cm thick. This created significant near stagnant flow regions at each mirror location, which could adversely affect the supersonic flow field and thus reduce gain. However, gain measured at these conditions was similar to the previous measurements with the thinner side walls (see Section 3.12), γ=0.17-0.20 %/cm in the entire supersonic flow region.
Figure 3.41 and Figure 3.42 show gain measured along the resonator hypotenuse (i.e. from port 3 to port 2, see Figure 2.17 and Figure 3.43). In Figure 3.41, the gain line is due to iodine atom population inversion achieved in the cold supersonic core flow at the temperature of T=105 K, inferred from the best fit Doppler lineshape, also shown in Figure 3.41. The gain path length through the supersonic core flow is about 10.8 cm, assuming negligible boundary layer thickness. The weaker absorption line is due to near-stagnant regions in the mirror cavities and boundary layers (see Figure 3.43), with a ‘best fit’ temperature of T=350 K, which is consistent with the temperature measured in the boundary layers during gain mapping measurements (see Figure 3.26). Line of sight integrated gain and absorption are shown in Figure 3.41 and Figure 3.42 since gain path length for both of these regions is somewhat uncertain. The gain line in in Figure 3.41 is shifted by $\Delta \nu = -418$ MHz relative to the absorption line, due to Doppler effect. The gain lineshape shown in Figure 3.41 also displays a Doppler shift of $\Delta \nu = -418$ MHz compared to gain lineshapes measured perpendicular to the flow direction, using resonator ports 1,2 and 3,4 due to the $22^\circ$ angle of the gain probe beam (see discussion of Figure 3.44).
Figure 3.41: Gain measured along a triple pass resonator hypotenuse (see Figure 2.17 and Figure 3.43 for the resonator schematic). 7% O$_2$-He; 22 mmole/sec O$_2$, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; $p_0=95$ torr after injection of 110 µmole/sec I$_2$ in 45 mmole/sec He and 255 mmole/sec chilled He; $P_p=3.25$ kW.

Figure 3.42: Absorption measured on a triple pass resonator hypotenuse (magnified compared to Figure 3.41). 7% O$_2$-He; 22 mmole/sec O$_2$, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; $p_0=95$ torr after injection of 110 µmole/sec I$_2$ in 45 mmole/sec He and 255 mmole/sec chilled He; $P_p=3.25$ kW. Doppler shift of the gain line relative to the absorption line is $\Delta v=418$ MHz.
To determine gain along resonator passes perpendicular to the flow direction (see Figure 3.43), flat, 99.9% reflectivity mirrors were installed at ports 2 and 3, and single pass gain was measured with gain probe laser beam propagating as shown in Figure 3.43, following ports 1→2→3→4. The results are shown in Figure 3.44. For this measurement, the gain probe beam was reflected by mirrors 2 and 3. The higher gain line is created by the superposition of gain from two transverse gain probe passes, 1→2 and 3→4 (both are unaffected by the Doppler shift). The lower gain line is created by gain on the resonator hypotenuse, pass 2→3, and shifted by $\Delta \nu = 402$ MHz. A single pass gain measurement taken for the opposite gain probe laser beam direction (i.e. 4→3→2→1) would show the gain on the hypotenuse pass shifted in the opposite direction, $\Delta \nu = -402$ MHz. Summarizing, the triple-pass resonator actually generates gain on three different wavelengths, since, during lasing, intraresonator photons are traveling simultaneously in both directions. These wavelengths are Doppler shifted relative to each other, such that the second pass (2↔3) does not contribute to gain generated on two other passes (1↔2 and 3↔4), and lasing occurs on three different lines. The flow temperatures inferred from the gain line shapes shown in Figure 3.44 are close to each other, as expected, $T=95$ K for the two perpendicular resonator passes, and $T=90$ K for the angled resonator pass.
Figure 3.43: Z-pass resonator with gain probe beam path

Figure 3.44: Gain measured on individual passes of the triple-pass resonator. 7% O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; p₀=95 torr after injection of 110 µmole/sec I₂ in 45 mmole/sec He and 255 mmole/sec chilled He injection; P₀=3.25 kW.

The frequency shift between the two gain lines in Figure 3.44 is too small to be resolved by the OMA used for emission spectroscopy measurements, and with no wavemeter available the laser output has not been spectrally analyzed. It is likely
that the majority of laser power is coupled out on the unshifted frequency (left line in Figure 3.44) because it has longer double-pass gain path, 40 cm compared to 10.8 cm for each of the shifted gain lines on the angle resonator pass. However, laser action at the shifted frequencies may well be possible. There is a mid-resonator flow region, between the upstream and downstream pairs of mirrors, with an axial flow length of 3.8 cm where the intraresonator field at the unshifted frequency will not interact with the flow. Gain may recover above laser threshold in this region, leading to laser action on one (or both) of the shifted frequencies, in addition to the unshifted frequency. Also note that in all our laser power measurements so far (see Sections 3.7 and 3.13), gain measured a few cm downstream of the resonator during lasing was only about 25% lower than gain measured at the same location without lasing, indicating significant power left available for coupling out.

The flow velocity in the laser cavity can be determined from the frequency shift between the two gain lines in Figure 3.44 and angled resonator pass angle, \( \theta = 22^\circ \), as follows:

\[
v = \frac{\lambda_0}{\sin \theta} \Delta v = \frac{1315 \text{ [nm]}}{\sin 22^\circ} \times 402 \text{ [MHz]} = 1411 \text{ [m/s]}, \tag{79}\]

where \( \lambda_0 = 1315 \text{ nm} \) is the \( I^* \) emission wavelength. The Mach number in the supersonic cavity is
\[ M = \frac{v}{\sqrt{kRT}} = 2.74, \]  

(80)

where \( k \) is the specific heat ratio, \( R \) is the gas constant, \( R=R_0/\mu \), where \( \mu \) is the molecular weight of the mixture, and \( T=90 \text{ K} \) is the flow temperature inferred from gain lineshape. This agrees well with the Mach number \( M=2.87 \) inferred from the static pressure traces shown in Figure 3.21, using a quasi-one-dimensional isentropic flow theory. Inference of flow Mach number at the present conditions, based on stagnation pressure and static pressure measurements would not be accurate due to the cold flow injection, which significantly affects flow stagnation temperature.

Laser output power was measured with three high reflectivity mirrors (99.9 % reflectivity), placed at locations 1, 2, and 3 of the triple-pass resonator and a 98.6 % or 99.5 % output coupler placed at location 4, see Figure 3.43. The laser power coupled out on the output coupler side of the resonator was 7.0 W for the 98.6 % mirror and 5.6 W for the 99.5 % mirror. For each case, a combined \( \sim 0.5 \text{ W} \) of laser power was coupled out through three high reflectivity mirrors, leading to total laser output powers of 7.5 and 6.1 W for the 98.6 % and 99.5 % configurations, respectively. This is lower than 7.8 W of laser power coupled out using a smaller, single-pass transverse resonator discussed in Sections 3.7 and 3.13. Note that mirror alignment of a four-mirror resonator was significantly more difficult. Also, mirrors could not be adjusted during the run to optimize the laser power, which
may have contributed to the reduced laser output power. Efforts to increase gain using iodine vapor dissociation in an auxiliary electric discharge continued (see Section 3.16) while a new set of sidewalls was manufactured to accommodate a single-pass transverse resonator, to be operated using a 1.5 cm (height) x 3.8 cm (length in the streamwise direction) apertures with larger 5.08 cm diameter mirrors.

3.16 Effect of Iodine Dissociation in an Auxiliary Discharge on Gain and Laser Power

As discussed in Chapter 1, dissociation of I$_2$ in an auxiliary discharge by direct electron impact has previously been reported to increase gain in an electrically excited oxygen-iodine laser ($^{71; 61}$). Reducing the amount of O$_2$($^1\Delta$) used for I$_2$ dissociation by reaction of Eq. (11) increases the effective O$_2$($^1\Delta$) yield that can be used for energy transfer to I atoms in the reaction of Eq. (16), which may well increase gain in a wide range of conditions. Additionally, the reduced cost of I$_2$ dissociation in terms of singlet delta oxygen yield may result in further gain increase by operating at higher iodine vapor flow rate. Note that our previous study of the effect of dissociation in an auxiliary electric discharge ($^{71}$) produced inconclusive results. Although significant iodine dissociation fractions were produced (up to 50 %), this resulted in gain increase by only 10-15 %. In reference ($^{71}$) the effect of iodine dissociation on gain was fairly insignificant compared to the effect of energy loading by the discharge and the effect of adding NO to the flow. However, in reference ($^{71}$), iodine was dissociated by a repetitive nanosecond pulse discharge in
the I\textsubscript{2} vapor-He delivery line, before the I\textsubscript{2}-He flow reached the injector. This approach leaves a possibility for I atom recombination prior to their injection into the flow. In the present work, we have attempted to dissociate I\textsubscript{2} vapor in an auxiliary RF discharge sustained near the I\textsubscript{2}-He injection location, in a cavity in the main flow channel (see Figure 2.9).

### 3.16.1 Iodine Injector and Auxiliary Discharge Configuration 2a

For these experiments, iodine injector 1, used in all experiments up to this point, was replaced with iodine injector 2a, shown in Figure 2.9. In this injector, two cylindrical rod electrodes, placed inside hollow alumina ceramic tubes, were used to sustain an auxiliary RF discharge near the location of injector ports, as shown in Figure 2.9. Initially, the auxiliary discharge electrodes were connected to the same 5 kW Dressler RF plasma generator that powered the primary electric discharge in the nozzle plenum. In this case, two pairs of electrodes for the primary discharge and for the auxiliary discharge were connected in parallel. However, it was found that in this configuration, breakdown is produced only in the auxiliary discharge. After that, the auxiliary discharge electrodes were connected to a separate ENI 600 W RF power supply with a MFJ-3000 manual impedance matching network.

First, the auxiliary RF discharge was operated with the primary discharge turned off, to measure the iodine dissociation fraction achieved in the auxiliary discharge. For these measurements, the excited iodine atom population was assumed to be zero, such that the ground state I atom population could be inferred from absorption measurements on I→I* transition near \(\lambda=1315\) nm. Obviously, using
this assumption yields a lower bound estimate of the iodine dissociation fraction. In this case, the dissociation fraction is given as follows:

\[ \alpha = \frac{n_1}{2(n_{1z})_{\text{initial}}} = \frac{12 \gamma |\gamma| \dot{n}_{\text{tot}} k_B T}{7 \sigma \dot{n}_{1z} p} = 1.066 \cdot 10^{-3} \gamma \frac{\dot{n}_{\text{tot}}}{\dot{n}_{1z}} \frac{T^{3/2}}{p}, \]  

(81)

where \(\gamma\) is the absorption measured on the \((F'=3\rightarrow F''=4)\) line center, \(\sigma\) is the absorption cross section (see Eq. (21)), \(\dot{n}_{\text{tot}}\) is the total mole flow rate through the laser, \(\dot{n}_{1z}\) is the iodine vapor flow rate, and \(T\) and \(p\) are the flow temperature and pressure at the absorption measurement location (all express in SI units). The temperature has been inferred from the absorption lineshape using a Doppler lineshape fit. Note that absorption measurements with the main discharge turned off need to be made with caution. It was found that in this case, atomic iodine accumulation in optical window recesses, where the flow is nearly stagnant, may result in very high absorption on the iodine atom line, and overprediction of flow temperature by over 50%. Both of these effects would increase the apparent dissociation fraction, which at some conditions may exceed \(\alpha=1\). First series of iodine dissociation measurements was conducted with the auxiliary discharge powered by the 5 kW Dressler power supply, to explore the trend at high discharge powers. These measurements were taken at the same plenum pressure of \(p_0=90\) torr, with and without oxygen in the main He flow. Absorption and \(I_2\) dissociation fraction vs. auxiliary discharge power are shown in Figure 3.45 and Figure 3.46, respectively. It can been seen that both absorption and dissociation fraction are a
factor of two higher when oxygen is present in the main flow, compared to their values measured in helium flow without oxygen, at the same plenum pressure. Recombination of I atoms,

\[ I + I + M \rightarrow I_2 + M \]  \hspace{1cm} (82)

which occurs an order of magnitude faster with O\textsubscript{2} as a third partner \( (k_{74, O_2} = 3.7 \times 10^{-32} \text{ cm}^6/\text{sec}) \) than with He as a third partner \( (k_{74, He} = 3.6 \times 10^{-33} \text{ cm}^6/\text{sec}) \), cannot explain higher I atom concentrations in the presence of oxygen. It is likely that a significant fraction of electronically excited atoms, I\textasciitilde, is also generated in the auxiliary discharge. Excited iodine atoms persist in the pure He but are rapidly quenched by the ground electronic state O\textsubscript{2} molecules in the reverse reaction of Eq. (16). This effect could explain higher apparent iodine dissociation fractions at the conditions when oxygen is present in the flow but no O\textsubscript{2}(1\Delta) is generated in the main discharge, which has been turned off. Presence of O\textsubscript{2}(1\Delta) in the flow would turn on the forward reaction of Eq. (16), thus greatly reducing the net rate of I\textasciitilde quenching. Note that it would require only about 15 \% of the iodine atoms generated in the discharge to be excited to the I\textasciitilde state to reduce the absorption on the iodine line and the apparent dissociation fraction (i.e. when the I population is assumed to be zero) by a factor of two. However, emission spectroscopy measurements to measure the absolute concentration of I\textasciitilde have not been made.
Figure 3.45: Absorption vs. auxiliary discharge power. Helium or 7% O₂-He mixture; 0 or 22 mmole/sec O₂, 297 or 392 mmole/sec He, 375 µmole/sec NO flow through the discharge section; p₀=90 torr after injection of 120-160 µmole/sec I₂ in 50 mmole/sec He and 150 mmole/sec He.

Thus, the results of iodine dissociation measurements in the auxiliary discharge, shown in Figure 3.46, show that significant iodine dissociation fraction,
up to tens of percent, can be generated in the supersonic cavity by using an auxiliary discharge. In the next series of measurements, the auxiliary discharge (this time powered by the 600 W ENI power supply) was operated in conjunction with the primary discharge (powered by the 5 kW Dressler power supply) to determine the effect of the auxiliary discharge on gain in the supersonic cavity. During these measurements, the auxiliary discharge was powered in the middle of the run for 1-2 seconds, such that gain measured with and without the auxiliary discharge could be compared. During experiments using the downstream primary discharge section, gain with the auxiliary discharge off was $\gamma = 0.107 \% / \text{cm}$ and gain with the auxiliary discharge turned on was $\gamma = 0.166 \% / \text{cm}$. This result was obtained at the following conditions: 7 % $\text{O}_2$-$\text{He}$ mixture; 22 mmole/sec $\text{O}_2$, 297 mmole/sec $\text{He}$, 375 $\mu$mole/sec NO flow through the discharge section; $p_0 = 90$ torr after injecting 110 $\mu$mole/sec $\text{I}_2$ in 50 mmole/sec $\text{He}$ and 260 mmole/sec chilled $\text{He}$, $P_p = 2.75$ kW, $P_{aux} = 425$ W. During these experiments, interaction between the primary and auxiliary discharges was observed at some conditions, when the primary RF discharge would extend downstream to the auxiliary discharge electrodes. Even after the auxiliary RF power supply was turned off, visible emission from the auxiliary discharge electrodes (driven by the primary discharge), was still detected and the auxiliary discharge impedance matching network continued to detect power. To avoid damaging the auxiliary power supply, further experiments continued while powering the upstream primary discharge section, which was not observed to interact with the auxiliary discharge. Gain measured vs. auxiliary
discharge power (with the auxiliary discharge turned on and off) is shown in Figure 3.47 for the optimum gain conditions with chilled He flow used: 7% O2-He mixture; 22 mmole/sec O2, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; p0=95 torr after injecting 130 µmole/sec I2 in 48 mmole/sec He and 270 mmole/sec chilled He, Pp=2.75 kW. Note that previously, the highest gain achieved with cold He injection and the upstream primary discharge section powered (using iodine injector 1) was γ=0.088%/cm (see Table 3.5). The gain achieved during the present measurements, at similar flow and primary discharge conditions (using injector 2a) and with the auxiliary discharge turned off is somewhat lower, γ=0.04-0.05%/cm (see Figure 3.47). It can be seen that the auxiliary discharge (at Paux=0.4-0.6 kW) increased gain by up to a factor of 2 above the highest gain that had been achieved previously using the upstream discharge section, but still well below the highest gain that had been achieved using the downstream discharge section, γ=0.209%/cm (see Section 3.12).
Figure 3.47: Gain vs. auxiliary discharge power, with the upstream primary discharge section powered. 7 % O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; \( p_0 = 95 \) torr after injecting 130 µmole/sec I₂ in 48 mmole/sec He and 270 mmole/sec chilled He; \( P_p = 2.75 \) kW; \( x = 3.2 \) cm.

Gain with and without the auxiliary discharge powered was also measured vs. NO flow rate, with and without chilled He injection. This was done to determine whether the optimum gain conditions changed with the auxiliary discharge turned on. The results are shown in Figure 3.48 and Figure 3.49, respectively. In these experiments, gain was measured 7.9 cm downstream of the supersonic nozzle exit. As can be seen from Figure 3.48, at low NO flow rates without chilled He injection gain with the auxiliary discharge turned on is actually lower than when the auxiliary discharge is off. However, the effect is reversed at high NO flow rates, thus suggesting that operation of the auxiliary discharge generates additional O atoms in the flow. With chilled He injection, gain is always higher with the auxiliary discharge turned on, even at low NO flow rates (see Figure 3.49). It is possible that operation
of the auxiliary discharge at a higher plenum pressure ($p_0=93$ torr with chilled He injection compared to $p_0=51$ torr without injection) results in less O atom generation, or that the effect of O atom generation is simply outweighed by the benefit of iodine dissociation at the reduced temperature achieved with chilled He injection. Similar to the results shown in Figure 3.47, gain using iodine injector 2a and with the auxiliary discharge turned off is lower than gain achieved at similar flow and primary discharge conditions using iodine injector 1 (see Table 3.5). We conclude that, while operation of the auxiliary discharge somewhat improves gain compared to what had been achieved with the upstream primary discharge section powered, using iodine injector 1, gain has not reached the values achieved with the downstream discharge section powered, using injector 1. The two differences between Injector 1 and Injector 2a are (A) Injector 1 is made of aluminum, while Injector 2a is made of acrylic plastic, and (B) presence of flow cavities accommodating the auxiliary discharge in Injector 2a. To determine which one of these two differences (injector material or flow geometry in the injector) resulted in superior performance of Injector 1, future work is planned using Injector 2b, which is made of acrylic plastic, similar to Injector 2a, but does not have flow cavities (see Figure 2.9).
Figure 3.48: Gain with and without auxiliary discharge powered vs. NO flow rate (using injector 2a without chilled He injection). 7% O₂-He; 22 mmole/sec O₂, 297 mmole/sec He flow through the discharge section; p₀=51 torr after injecting 140 µmole/sec I₂ in 48 mmole/sec He; Pₚ=2.75 kW; Pₚₐₓ=410 W; x=7.9 cm

Figure 3.49: Gain with and without auxiliary discharge powered vs. NO flow rate (using injector 2a with chilled He injection). 7% O₂-He; 22 mmole/sec O₂, 297 mmole/sec He flow through the discharge section; p₀=93 torr after injecting 140 µmole/sec I₂ in 48 mmole/sec He and 270 mmole/sec chilled He; Pₚ=2.75 kW; Pₚₐₓ=440 W; x=7.9 cm
To isolate the effect of plenum pressure increase and temperature reduction with chilled flow injection, gain was measured vs. He injection rate through the buffer flow injector, both for room temperature He injection and for chilled He injection, with the results shown in Figure 3.50 and Figure 3.51, respectively. One can see that room temperature helium injection, which increases plenum pressure but does not reduce flow temperature, decreases gain, as expected (see Figure 3.50). Turning on the auxiliary discharge at these conditions provides some benefit at moderate room temperature He injection rates, but this benefit decreases at higher He injection rates. As can be seen from Figure 3.51, chilled He injection (with the auxiliary discharge off) first increases gain somewhat, as expected, but gain eventually falls off as He injection flow rate is increased, which was not expected. For comparison, when using injector 1, gain increased with chilled helium injection flow rate for the entire range tested (see Figure 3.36). Finally, turning on the discharge, while using chilled He injection, produced a pronounced increase in gain at moderate chilled He injection rates, although it appears that gain also begins to fall off at high injection rates. As discussed above, gain measured using iodine injector 2a and with the auxiliary discharge off is lower compared to gain achieved at similar flow and primary discharge conditions with iodine injector 1 (see Table 3.5). While operation of the auxiliary discharge may improve gain compared to what had been achieved with the upstream primary discharge section powered, while using iodine injector 1, gain never reached the values achieved with the downstream discharge section powered while using injector 1.
Figure 3.50: Gain with and without auxiliary discharge powered vs. room temperature He injection rate (using injector 2a). 7 % O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; p₀=70-120 torr after injecting 130 µmole/sec I₂ in 50 mmole/sec He and additional warm He; Pₚ=2.75 kW; Pₐux=470 W; x=3.2 cm

Figure 3.51: Gain with and without auxiliary discharge powered vs. chilled He injection rate (using injector 2a). 7 % O₂-He; 22 mmole/sec O₂, 297 mmole/sec He, 375 µmole/sec NO flow through the discharge section; p₀=68-96 torr after injecting 130 µmole/sec I₂ in 50 mmole/sec He and additional chilled He; Pₚ=2.75 kW; Pₐux=470 W; x=3.2 cm
Summarizing, the present measurements have demonstrated that the use of auxiliary discharge to dissociate iodine vapor injected into the main flow has potential to increase gain significantly. However, the use of Injector 2a has not produced gain higher than when using Injector 1. In particular, limited sets of data taken with the downstream primary discharge powered, while using Injector 2a, produced gain significantly lower compared to gain measured while using Injector 1 at the same flow and primary discharge conditions. Also, when using injector 2a, even without the auxiliary discharge powered, gain falls off with chilled He flow rate (see Figure 3.51), which had not been observed previously while using iodine injector 1. Finally, at high chilled He injection rates, gain was often unstable during the run and run-to-run reproducibility was poor. Thus, with the auxiliary discharge off, iodine injector 1 outperformed iodine injector 2a in the entire range of conditions tested.

3.17 Iodine Dissociation in the Primary Discharge

One of the primary goals of DOIL development is achieving closed cycle operation, where the gas mixture flow circulates in a continuous loop through the laser. Operating the laser in this mode would result in the presence of iodine in the primary discharge. Dissociation of I\(_2\) by direct electron impact in an auxiliary electric discharge has already been shown to increase gain for some operating conditions, which suggests that iodine dissociation in the primary discharge might also increase gain. To test this hypothesis, an additional iodine injector was installed
between the flow conditioning sections and the upstream discharge section, as shown in Figure 3.52. The design of this injector is discussed in Section 2.1.

![Figure 3.52: Schematic of additional iodine vapor injector installed upstream of the primary discharge section](image)

Gain measurements were taken at the baseline conditions: 7% O₂-He mixture; 34 mmol/sec O₂, 491 mmol/sec He, 375 µmol/sec NO, 120 µmol/sec I₂ flow through the discharge section; p₀=70 torr; no chilled flow injection; Pₚ=1-3.5 kW. Nearly the same absorption, between γ=-0.08 and γ=0.09%/cm, was recorded in the entire range of primary discharge power tested. Other flow and discharge parameters were varied to determine their effect on gain/absorption. In particular, gain/absorption was found to be nearly independent of O₂ fraction, NO flow rate, and discharge pressure (increased by injecting helium flow downstream of the primary discharge), and dependent only on I₂ flow rate.

Emission spectroscopy measurements were taken downstream of the discharge. Emission from the O₂(¹Δ→³Σ) transition, near λ=1268 nm, with and without iodine in the primary discharge is shown in Figure 3.53. One can see that singlet delta oxygen emission is undetectable when iodine is flown through the discharge. The absence of O₂(¹Δ) in the flow explains nearly the same absorption
observed during gain measurements. Since absorption did not vary with primary discharge power, it is likely that iodine was completely dissociated even at the lowest discharge power tested, \( P_p = 1 \text{ kW} \).

![Infrared emission spectra at \( \lambda = 1268 \) taken with iodine flown through the primary discharge to capture emission from the \( \text{O}_2(1\Delta \rightarrow 3\Sigma) \) transition. 7% \( \text{O}_2\)-He; 34 mmole/sec \( \text{O}_2 \), 491 mmole/sec He, 375 mmole/sec NO, 120 mmole/sec \( \text{I}_2 \) flow through the discharge section; \( p_0 = 69 \text{ torr} \); \( P_p = 2.0 \text{ kW} \); downstream primary discharge section powered.](image)

Emission from the \( \text{O}_2(1\Sigma \rightarrow 3\Sigma) \) transition, near \( \lambda = 760 \) nm, with iodine present in the primary discharge is shown in Figure 3.54 (emission without iodine in the primary discharge was not taken). The \( \text{O}_2(1\Sigma \rightarrow 3\Sigma) \) emission is comparable to the one at similar discharge and flow conditions but without iodine (see Section 3.2). The atomic emission lines have been identified as O and I atom emission. The presence of \( \text{O}_2(1\Sigma) \) indicates the discharge conditions are suitable for \( \text{O}_2(1\Delta) \) production, since the energy-dependent electron impact cross sections for the \( \text{O}_2(3\Sigma \rightarrow 1\Sigma) \) and \( \text{O}_2(3\Sigma \rightarrow 1\Delta) \) transitions behave in a similar way (see Figure 1.7). Thus, the absence of
$O_2(^1\Delta)$ emission suggests that it has been removed by a chemical process involving iodine molecules or atoms. Analysis of oxygen-iodine chemistry in the discharge, very likely involving vibrationally and electronically excited species, is beyond the scope of the present work. Operation of the primary discharge with a $O_2$-He-$I_2$ mixture resulted in an orange color solid deposit accumulated on the flow channel walls in the discharge section, similar to what is reported in reference (35). The deposit was not analyzed and was cleaned after experiments with iodine flowing through the primary discharge were finished.

![Emission spectrum](image)

Figure 3.54: Visible emission spectrum at $\lambda=760$ nm taken with iodine flown through the primary discharge to capture emission from the $O_2(^1\Sigma \rightarrow ^3\Sigma)$ transition. 7 % $O_2$-He; 34 mmole/sec $O_2$, 491 mmole/sec He, 375 µmole/sec NO, 120 µmole/sec $I_2$ flow through the discharge section; $p_0=70$ torr; $P_p=3.0$ kW; downstream primary discharge section powered; Atomic emission lines identified using NIST Atomic Spectra Database as I or O atom emission. Peak O atom emission at 777 nm is approximately $4.2 \times 10^4$ counts.
Chapter 4: Summary and Conclusions

In the present work, a scaled up electric discharge oxygen-iodine laser has been designed and tested, with a total discharge volume of 200 cm$^3$, a total gas flow rate up to 780 mmole/sec of a O$_2$-He-NO-I$_2$ mixture, and a gain path in the Mach 3 laser cavity of 10 cm. Singlet delta oxygen is generated in transverse capacitively coupled RF discharge, at discharge powers up to 5 kW, operated in the plenum of a supersonic nozzle at discharge pressures of $p_0=40$-$120$ torr, with O$_2$(^1\Delta) yields up to $Y=7.5\%$. Iodine vapor in helium carrier is injected into the main flow downstream of the discharge, and the mixture is subsequently expanded in a supersonic nozzle to $M=3$. Flow temperature reduction during rapid supersonic expansion shifts equilibrium in near-resonance energy transfer process between O$_2$(^1\Delta) and iodine atoms toward formation of excited I({^2P}_{1/2},F'\!=\!3) state, thus generating absolute population inversion in iodine atoms. Small signal gain up to $\gamma=0.209\%/\text{cm}$ on the $\text{I}({^2P}_{1/2},F'\!=\!3)\!\!\to\!\!\text{I}({^2P}_{3/2},F'\!=\!4)$ magnetic dipole allowed transition near $\lambda=1315$ nm is measured in the supersonic test section, at flow temperatures of approximately $T=100$ K.

Laser action is achieved in transverse resonator in the supersonic cavity, with total laser output power up to $P_L=7.8$ W. The most significant advances resulting in improved laser performance are (i) optimization of the primary electric
discharge for high $O_2(^{1}\Delta)$ yield and low flow temperature rise, (ii) the addition of NO to remove O atoms generated in the discharge, (iii) optimization of the $I_2$ vapor flow rate, and (iv) reduction of flow temperature by injection of chilled helium downstream of the primary discharge, prior to supersonic expansion. The laser operates at steady state flow conditions for up to 5-10 seconds, with the run time limited by the vacuum system volume. The relatively short run time makes it difficult to control the iodine vapor flow rate and optimize the laser resonator for power extraction (mirror alignment). Time between successive runs, 10-25 min is limited by the pumping rate.

The supersonic section originally included an angle step diffuser, designed to increase steady-state flow run time and reduce the effect of back pressure in the vacuum system on the flow in the laser cavity. However, the present experiments, discussed in Sections 3.1, 3.6, have demonstrated that the diffuser did not increase run time but considerably reduced gain in the downstream region of the supersonic cavity, likely due to boundary layer separation upstream of the diffuser. The diffuser was therefore removed, which improved both flow quality and gain uniformity in the supersonic cavity.

The primary electric discharge performance was characterized using infrared and visible emission spectroscopy, discussed in Section 3.2. Singlet delta oxygen yield was measured using absolute emission spectroscopy measurements of the $O_2(^1\Delta, v'=0\rightarrow^3\Sigma, v''=0)$ magnetic dipole allowed radiative transition near $\lambda=1268$ nm, calibrated with blackbody source radiation. Flow rotational temperature
downstream of the discharge was inferred from visible emission spectra of the $O_2(^1\Sigma, v'=0 \rightarrow ^3\Sigma, v''=0)$ magnetic dipole allowed radiative transition near $\lambda=762$ nm. At the present conditions, the rotational temperature is essentially the same as the translation temperature. At typical flow rates through the primary discharge, singlet delta oxygen yield increases nearly linearly at low discharge powers but eventually levels off at $Y=7.5\%$ near $P_p=3.0\text{-}3.5$ kW. Flow temperature increases linearly with the discharge power, at a rate of $\sim 25 \text{ K/kW}$, for the entire range of powers tested.

Initial gain measurements, discussed in Sections 3.3 and 3.4, demonstrated the hyperfine (nuclear spin orbit interaction) components of $I^*(2P_{1/2})$ and $I(2P_{3/2})$ states to be in equilibrium in the supersonic cavity, and the $I^*\rightarrow I$ gain spectrum to be in good agreement with theory. Gain was shown to be strongly dependent on NO flow rate, due to removal of O atoms generated in the primary discharge by reactions with NO. Without NO addition, oxygen atoms rapidly deactivate excited iodine atoms, $I^*$. $I_2$ vapor injection flow rate was optimized to achieve maximum gain. At low $I_2$ flow rates, gain is reduced due to low iodine atom density in the flow, and at high $I_2$ flow rates gain falls due to $O_2(^1\Delta)$ depletion in $I_2$ dissociation reaction (Eq. (11)). The maximum gain achieved during the initial gain measurements is $\gamma=0.116\% / \text{cm}$. From temperature in the supersonic cavity, $T=100\text{-}110$ K, was determined from a Doppler fit to the gain lineshape.

Initial laser power measurements, discussed in Sections 3.5 and 3.7, were conducted at the optimum gain conditions determined by the initial gain measurements (Section 3.4). A stable transverse resonator was operated 3.2 cm
downstream of the supersonic nozzle exit, achieving maximum laser output power of $P_L=3.1$ W using a 99% - 99.9% combination of 2.54 cm diameter laser mirrors. Gain measurements downstream of the laser resonator indicated insignificant gain reduction (or rapid gain recovery) such that additional power could be extracted using a second resonator. Stable transverse laser resonators were operated simultaneously 3.2 cm and 12.7 cm downstream of the nozzle exit, extracting 2.9 W and 0.9 W, respectively, with total laser output power of $P_L=3.8$ W.

Significant reduction of power extracted from the downstream resonator, in spite of similar gain measured at both locations, suggested that laser performance may be affected by boundary layer growth in the laser cavity. Gain and flow temperature distributions were measured along the vertical coordinate in the supersonic cavity coordinate (through the boundary layers on the top and bottom walls). The results were compared to CFD modeling calculations using a 3-D compressible Navier-Stokes flow code, as discussed in Section 3.8. The results of gain distribution measurements showed a significantly smaller positive gain region at the downstream resonator location, partially explaining lower output power measured in the downstream resonator. The thermal boundary layer thickness predicted by the flow code is consistent with the measurement results, although the flow temperature was under predicted in the entire cavity. This discrepancy may be due to uncertainty in the temperature measurements, discussed in Section 3.9.

Operating two sequential RF discharge sections, with the total discharge volume increased from 200 cm$^3$ to 400 cm$^3$, discussed in Section 3.10, has not
resulted in gain increase. Similar $O_2(^1\Delta)$ yields were achieved with both discharge configurations (with one or two discharge sections powered). However, operating two discharge sections produced higher flow temperature, which resulted in somewhat lower gain. Replacing helium in the main laser mixture flow by argon drastically reduced gain and produced absorption on iodine line, as discussed in Section 3.11. This result contradicts kinetic modeling calculations, which predict that larger discharge energy fraction goes to $O_2(^1\Delta)$ excitation in oxygen-argon mixtures, compared to oxygen-helium mixtures.

A chilled flow injector was installed between the iodine vapor injector and the supersonic nozzle, as discussed in Section 3.12, to reduce the flow temperature prior to supersonic expansion. Injection of helium chilled in a liquid nitrogen bath reduced the supersonic flow temperature by about about 20%, from $T \approx 125$ K to $T \approx 100$ K. The discharge and flow parameters were optimized again, resulting in gain increase up to $\gamma = 0.209$ %/cm. Experiments were also conducted with injection of chilled nitrogen, with less pronounced effect. A main flow chiller was installed upstream of the main discharge section inlet, as discussed in Section 3.14, to further reduce the main flow temperature. Operation of the main flow chiller using a liquid nitrogen bath resulted in absorption measured in the supersonic cavity in the entire range of conditions tested, likely due to either or both of $O_2$ or NO condensation in the main flow chiller. The main flow chiller was also tested using an ethanol-liquid nitrogen bath to preclude condensation. At these conditions, no flow temperature reduction or gain increase were detected.
Iodine dissociation by electron impact in an auxiliary discharge operating near the iodine vapor injection port was studied to examine its potential for further gain increase, as discussed in Section 3.16. An alternative iodine vapor injector (Injector 2a) containing an auxiliary discharge sustained in cavity recesses was installed in place of the original iodine vapor injector (Injector 1). While the auxiliary discharge was turned off, significantly lower gain was recorded at all conditions tested with Injector 2a, compared to Injector 1. Operation of the auxiliary discharge in Injector 2a resulted in higher gain, but gain still did not reach the maximum previously achieved with the Injector 1. Although the use of iodine dissociation in auxiliary discharge did result in gain increase at some operating conditions, operation with Injector 2a did not produce overall improvement compared to operation with Injector 1. Further experiments are needed to determine the primary cause of lower gain measured using Injector 2a, which may be due to negative effect of discharge cavities on flow quality and mixing, or due to interaction between aluminum Injector 1 and the primary discharge.

The effect of iodine vapor flowing through the primary discharge has been studied, as discussed in Section 3.17. An additional upstream iodine injector was installed just upstream of the primary discharge section, such that iodine vapor could be added to the primary discharge gas mixture. These measurements resulted in absorption measured in the supersonic section at all conditions tested. At these conditions, emission spectroscopy measurements did not detect any O$_2$(^1Δ) downstream of the primary discharge.
Laser output power measurements have been conducted at the optimum gain conditions with chilled flow injection, determined in Section 3.12. The results of these measurements are discussed in Sections 3.13, 3.15, and 3.18. The stable transverse laser resonator discussed in Sections 3.5 and 3.7 was used to couple out laser power of $P_L=7.8$ W using a 99% - 99.9% combination of 2.54 cm diameter mirrors. Significant gain was again measured downstream of the resonator, indicating significant potential for additional power coupling using a laser resonator with a longer gain pass. For this, a Z-pass resonator was designed and operated using four 5.08 cm diameter mirrors. The Z-pass resonator accessed a larger flow volume compared to the smaller transverse resonator, but achieved lower output power of $P_L=7.5$ W.

The present work demonstrates operation of an electrically excited discharge oxygen-iodine laser with a scaled electric discharge and laser resonator (discharge up to 5 kW, discharge volume up to 400 cm$^3$, discharge pressure up to 100 torr, resonator gain path up to 30.8 cm). These results are critical for further development and scaling of oxygen-iodine lasers. Gain and laser output power measured in the present work represent a significant improvement compared to our previous results (71); however, significant additional progress is needed to demonstrate DOIL scalability to multi-kilowatt range, to be considered a viable alternative to COIL. Currently, the main effort in the field of directed energy delivery is in development of solid-state lasers.
References


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