

Final Report the Physics of High Efficiency Argon Fluoride Lasers

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EXECUTIVE SUMMARY

The primary objective of the research conducted is to study the physics of an electron beam pumped argon fluoride (ArF) laser. Generating sufficient laser energy from excimer laser gas medium for potential inertial confinement fusion (ICF) and directed energy applications will require large laser cavities that will need to be pumped by electron beams, rather than by electrical discharges. ArF has a shorter wavelength (193 nm) and higher photon energy than KrF and could enable unique applications. NRL enhanced world leading technology in electron beam pumped high energy excimer lasers with the development of short ArF ultraviolet wavelength which continues to show prospective benefits to research in inertial fusion, mitigating laser plasma instabilities, and as a diagnostic tool. The high quality science discovered in this effort has enabled technology development for the most energetic argon fluoride laser at 200 J with near prospects of generating significant more yield. The physics of greatest consequence include measurements of the small signal gain and saturation intensity with accompanying ArF laser kinetics over a significant pressure and composition range. These and other findings from this program have been documented work in 27 various publications/communications of record including five refereed journal articles, one patent publication/application and two memorandum reports.

This report presents research conducted by Dr. Matthew F. Wolford (Code 6733), Mr. Matthew C. Myers (Code 6733) and Dr. Tzvetelina B. Petrova (Code 6721).



Fig. E1 — The World's most energetic Argon Fluoride Laser at 200 J created due to this Base Research Program. Pictured in the photo are Dr. Wolford and Mr. Myers with the Electra ArF laser facility.

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FINAL REPORT THE PHYSICS OF HIGH EFFICIENCY ARGON FLUORIDE LASERS

1. INTRODUCTION

The argon fluoride (ArF) research conducted took advantage of a strategic opportunity for NRL, and specifically the Plasma Physics Division. The resultant realization of the ArF research has led to national interest and excitement of ArF (193 nm) laser for potential science advances in inertial confinement fusion (ICF) due to potential very high target gain in addition to the significant reduction of laser plasma instabilities compared to conventional laser drivers [1]. In addition, the research has shown a pathway to high efficiency ArF laser drivers for the eventual realization of inertial fusion energy (IFE) as well. We note the potential benefit of ArF (193 nm) laser driver with inertial confinement (ICF) targets and materials represents ‘truly new ground’ for low cost smaller inertial fusion power plant.

Generating sufficient laser energy from excimer laser gas medium for potential inertial confinement fusion (ICF) and directed energy applications require large laser cavities that will need to be pumped by electron beams, rather than by electrical discharges. The Plasma Physics Division at NRL has been the world leader technology in electron beam pumped high energy excimer lasers for over more than the last two decades [2-4]. The substantial improvement in excimer wavelength optics due to the development of laser technologies in the lithography industry decades ago has led to an opportunity to develop high energy 193 nm lasers, which previously were thought to be unattainable.

The major intent of this research effort is to make precise detailed measurements of gain, gain to loss ratio, saturation intensity and intrinsic efficiency over a broad parameter regime using an oscillator, reduced aperture amplifier and oscillator- amplifier configuration. Development of an experimental database and accompanying ArF kinetics simulation capability enables the design of high energy ArF systems with confidence. To attain these goals the first step was to conduct basic research in regard to the laser physics for the ArF laser system to attain the important reactions and subsequent laser dynamics that take place. One aspect of basic research is development modeling tools to represent present measurements over dynamic range. Utilizing synergistic parts of experimentation and theoretical analysis predictive capability can be initiated and eventually developed. The ongoing basic research for argon fluoride (ArF) lasers is to enhance and improve has technological developments are realized.

1.1 Benefit of Short Wavelength (193 Nanometer) Light

Fundamentally shorter wavelength has higher photon energy as well as higher frequency due the shorter photon period. The attributes of shorter wavelengths decrease the angles of diffraction compared to longer wavelength light. Therefore smaller focal spots are attained with shorter wavelength relative to longer wavelengths with equivalent diffraction laser beams due to the relative spot size scales with wavelength with a minor correction due to the refractive index of materials, which is constant for most materials. For inertial confinement fusion considerations the short wavelength with equivalent intensity has a higher ablation pressure on the target. In addition, the short wavelength component has a higher threshold for some laser plasma instabilities. Recently, an evaluation of the benefits of short wavelength laser target implosions has been published with more detail on target physics [5,6] then given here.

1.2 Previous Electron Beam Excimer Laser Development

One of the essential components for a viable inertial fusion power plant is the efficiency of the driver at conditions where substantial target gain can be achieved. For a laser driver there is a clear benefit of having a shorter wavelength, due to the increased coupling to the target and reduction of plasma instabilities. In addition a technique is required to minimize hydrodynamic instabilities and allow scalability to fusion power plant systems. Research on the krypton fluoride (KrF) laser has progressed due to the fact the potential exists of meeting all the requirements for laser fusion [7-10]. A pathway exists to an economically viable inertial fusion driver for KrF [11]. The fundamental wavelength of KrF is 248 nm a short ultraviolet wavelength and can be zoomed. As the wavelength becomes shorter increased absorption and increased rocket efficiency occurs resulting in larger kinetic energy to the target [12]. The United States Naval Research Laboratory, Plasma Physics Division, has demonstrated the use of induced spatial incoherence [13,14] with KrF lasers to minimize hydrodynamic instabilities [15]. To achieve the necessary energy for a KrF laser a few electron beam pumped amplifier stages would be used in series where each amplifier stage yields more controllable (pulse shape, spatial focus ability) laser energy than the laser input energy. Electron beam pumped KrF lasers has been examined by a number of institutions [15-22].

Argon fluoride (ArF) is another excimer laser system which can be pumped by electron beams [23-33]. One of the first examples of electron beam pumped ArF laser was from Sandia National Labs (shown in Figure 1), which produced 92 J [32-33]. This was soon after the first report of noble gas halide excimer laser (May 1975) by S.K. Searles and G.A. Hart at the Naval Research Laboratory (NRL) [34]. Argon fluoride lasers have a shorter wavelength than KrF. The short wavelength of 193 nm and may impact the target dynamics in a positive way. The benefits in hydrodynamic efficiency, laser plasma instabilities resulting in larger target gain could be realized as wavelength becomes shorter [35]. Full evaluation of the target physics will be left to an expert in this field [5,6]. One aspect of argon fluoride advancement is the evaluation of operating regimes where the ArF intrinsic efficiency is higher than observed with KrF. If the regime is accessible for large electron beam pumped systems it would 1) reduce the size and cost of the needed pulse power system and 2) increase the attractiveness of the ArF for laser fusion energy. The existing literature from the 1970's and 1980's is not precise or consistent enough to determine where or even if this high efficiency regime indeed exists. Nevertheless there is reason enough to suspect that ArF can have higher intrinsic efficiency than KrF over a broad range. The results that follow demonstrate a regime where the intrinsic efficiency is higher on a small scale and in process of being evaluated for larger scale systems.

2. APPROACH

Our approach was to find the maximum efficiency possible for an ArF electron beam pumped laser. This entailed an integrated approach with theoretical calculations and experiments to characterize the performance of the ArF laser. The technical approach of this proposal were: design, construct, and test an electron beam pumped ArF laser for very high intrinsic efficiency and evaluation for laser fusion driver capabilities; model validation for ArF electron beam pumped lasers to gain a predictive capability on building fusion class laser facilities; measurements of laser parameters including yield, time dependent emission (intensity), small signal gain (g_0), nonsaturable absorption (α), saturation intensity (I_{SAT}), and amplified spontaneous emission (ASE); time-dependent model and experimental comparisons for key species including ArF, Ar₂F, F₂ and electron density.

2.1 Fabrication of ArF laser with utilization of existing NRL facilities

The vast majority of required equipment for construction of ArF electron beam pumped amplifier already existed within Code 6730 and were utilized. There were some key components which were purchased for

this program including optics. The optical materials at 193 nm are expensive for large meter class area. Therefore the focus of this program was on laser efficiency and laser parameters as opposed to total laser yield. The Electra laser facility [36] which has been used in previously in KrF laser development [37-40] as well as other applications [41-43] will be utilized. The Electra facility has detailed studied several components [44-47] to allow confidence in reproducible performance for extensive experimentation.

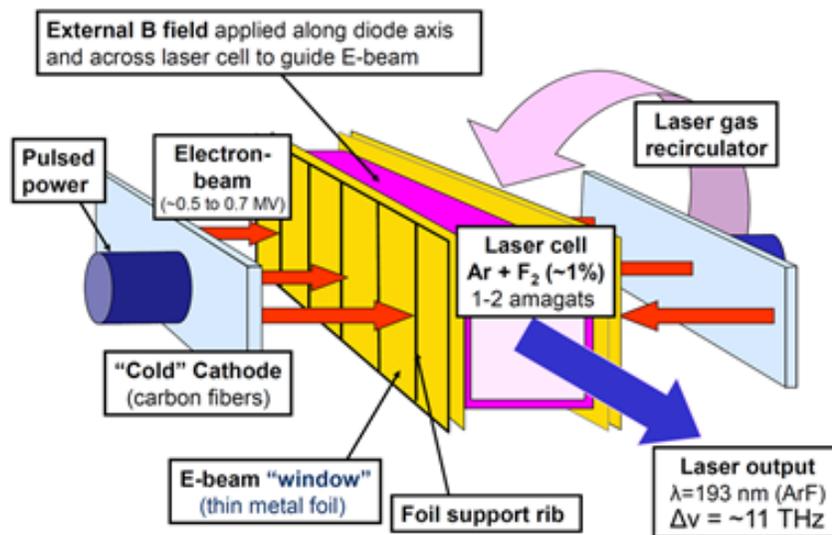


Fig. 1 — Schematic of argon fluoride (ArF) laser amplifier: Details of a modular, rep-rate, efficient ArF electron beam amplifier. Counter-propagating electron beams, from left and right, are used to create the laser gain medium, center of picture, with a gas recirculator for thermal management at high frequency operation. Picture of actual device in Fig. E1.

2.2 Modeling of electron beam amplifier

The Orestes code was able to correctly model the yield for a similar excimer laser (KrF) [48]. We modified and development a new ArF Orestes code to accurately model and predict the laser yield and other measurable diagnostic quantities for the ArF laser. The first step was to solve the electron Boltzmann equation for the Electron Energy Distribution Function (EEDF) of the ArF laser gas mixture[49], which also had been completed previously for KrF gas mixtures[50,51]. The EEDF determines the electron kinetics which includes electron and ion formation rate, excitation, ionization, attachment, and all other kinetic rates directly related to electrons. Another module of the code beyond the EEDF calculates the plasma chemistry. The plasma chemistry module examines the formation and destruction of molecules and will take into account of all species within the system. The combination of these modules resulted in a validated predictive capability for ArF electron beam pumped lasers.[52,53]

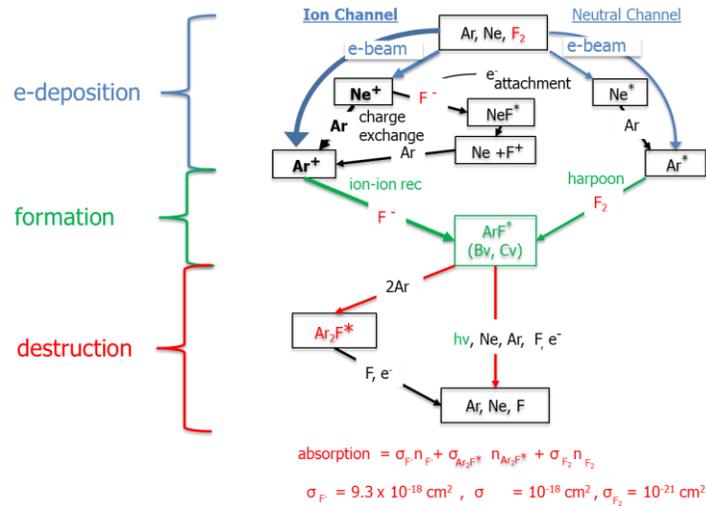


Fig. 2 — Schematic of argon fluoride (ArF) laser amplifier kinetics: Depicting the dominant reactions from both the ion channel and the neutral channel.

2.3 Measurement of laser parameters including laser efficiency, small signal gain, saturation intensity

Pressure, and composition parameter studies were conducted measuring laser yield, laser pulse shape (time-dependent intensity), amplified spontaneous emission (ASE), electron beam deposition and consequently laser efficiency [54]. The measurement of the small signal gain, nonsaturable absorption and saturation intensity for the electron beam amplifier were conducted in amplification experiments using an ArF 193 nm discharge laser [54]. To minimize the effects of ASE, polarization was used to discriminate spontaneous emission from the stimulated emission. All of these measurements were directly compared to simulation output.

2.4 Measurements of time dependent species

Linear spectroscopy, emission and absorption were used to measure dynamics of pertinent species in the laser gas including ArF, and F₂. We intended to conduct more investigations including Ar₂F and other spectroscopic measurements that are observable in the ultraviolet, visible or infrared spectrum. Unfortunately, resources, time and the pandemic outbreak limited are ability to execute these particular goals.

2.5 Ionization Shock Front Experiment

The Ionization Shock Front Experiment was also a victim due to resources, time and the outbreak. Our initial intent of the Ionization Shock Front Experiment is described here. The ArF laser also offers the possibility of an unique laboratory astrophysics experiment to study the transition from a pure ionization wave to a photon driven shock front as forms around young, hot stars. The specific application is the generation of a single ionization shock front utilizing a low threshold ionization atomic gas (i.e., Li, Na, K, Ru, Cs). In addition, the same ArF laser will be used to investigate critical properties for fusion laser drivers, including propagation in various gases to limit transmission losses as well as optic performance evaluation. If the ArF laser efficiency is proven to be high enough it may have directed energy applications in space. There its short wavelength is an advantage towards minimizing beam diffraction and the short wavelength light makes defensive measures more challenging.

3. RESEARCH & TECHNICAL ACCOMPLISHMENTS

Significant progress was achieved through this research effort, which resolved the underlying physics of an electron beam pumped argon fluoride (ArF) laser. The ArF laser propagates in the deep ultraviolet at 193 nm. This program had 3 major technical areas of significant accomplishments, including the design and construction of the world's most energetic ArF laser at 200 J[55], modeling of the electron beam amplifier with the Orestes kinetics code[52,53], measurements and modeling of laser parameters including laser efficiency, small signal gain and saturation intensity [54,56]. Results of the project were transmitted in 26 publications using different types of media as well at various locations throughout the time of the project.

3.1 World's Most Energetic ArF Laser

Design and construction of the world's largest ArF laser at the Electra facility took many iterations. A brief synopsis is provided. In order to construct an ArF large two large ($34.7 \times 32.7 \times 3.8 \text{ cm}^3$) fused silica windows were polished for transmission greater than 85% at 193 nm. The remaining losses are from Fresnel reflections from the uncoated surfaces of the fused silica windows and some losses through the thick (3.81 cm) standard grade fused silica material. Higher transmissivity fused silica windows are achievable with greater allocation of resources to fund anti-reflection coatings as well as higher quality optical materials. Funding for the program was basic research so material advancement of optics and optical coatings was not conducted within this program. The windows were installed on a 891 liter gas container which connected to the Electra facilities', two separate pulse power systems in this configuration providing counterpropagating electron beams each of 500 keV, 110 kA and a flat-top pulse width of 100 ns. Initial experimentation of the small signal gain showed the previous Electra cathode which spans over an area $30 \times 100 \text{ cm}^2$ did not create enough laser gain to attain substantial laser yield. Therefore a new cathode was developed for both counter propagating electron beams each having a reduced area to $10 \times 100 \text{ cm}^2$ [54]. New 25 micron stainless steel foils were installed to provide a barrier between the vacuum diode and the 891 liter laser gas mix containing argon and fluorine. Other materials were examined with less success for foil barrier between the laser gas and electron beam diode. The variables modified the most throughout the program for higher were optical configuration as well as gas constituent concentrations and laser gas pressure. Improvements were made in optical aperture to attain higher laser energy. Some of the results in chronological order include 100 J in double pass oscillator experiments with an optical aperture $10.5 \times 11 \text{ cm}^2$ with a total pressure of 0.82 atmospheres with 0.3% Fluorine and balance of argon. The next improvement was a dual parallel oscillator with two $11.9 \times 1.9 \text{ cm}^2$ apertures for a total yield of 137 J [56]. After this increase in laser yield we modified the laser optics to span the entire width of electron beam and made an oscillator with a piece of fused silica for a 8% Fresnel reflection. The $11.9 \times 30.0 \text{ cm}^2$ oscillator yield of 186 Joules. We further increased the yield by again modification of the optical design to close the distance down of the resonator to attain 200 J. Decreasing the resonator distance did two things, one increase the number of roundtrips while the electron beam was on and two reduced the slight absorption at 193 nm due to oxygen. In order to evaluate electron beam pumped ArF amplifier performance a new optical configuration was developed called an oscillator-amplifier configuration. The oscillator-amplifier configuration composed of an oscillator utilizing a portion of the generated ArF gain medium. Utilizing the same optics as described in the previous narrative update in detail, a $11.9 \times 11.9 \text{ cm}^2$ maximum clear aperture ArF oscillator was generated. The oscillator output was reflected from two 45 degree 193 nm high reflectors normal to each other generating a reflected laser beam offset and parallel to the initial oscillator. The parallel ArF laser beam passes back through the laser cell gain medium utilizing a new section of electron beam pumped volume to be amplified as an ArF laser amplifier. The sub aperture, $11.9 \times 11.9 \text{ cm}^2$, laser energy was measured in two locations after the oscillator then again after the amplifier stage. The results for the 8% 193 nm reflective output coupler was 57 J for the oscillator and 95 J for the amplifier. The results for the 16% 193 nm reflective output coupler was 54 J for the oscillator and 126 J for the amplifier. The results for the 23% 193 nm reflective output coupler was 51 J for the oscillator and 100 J for the amplifier. The conditions for all the oscillator-amplifier experiments mentioned above were 0.5% F2/99.5% Ar with a total pressure of 0.884 atmospheres total pressure.

Reduction in Cathode Area increases MW/cm^3

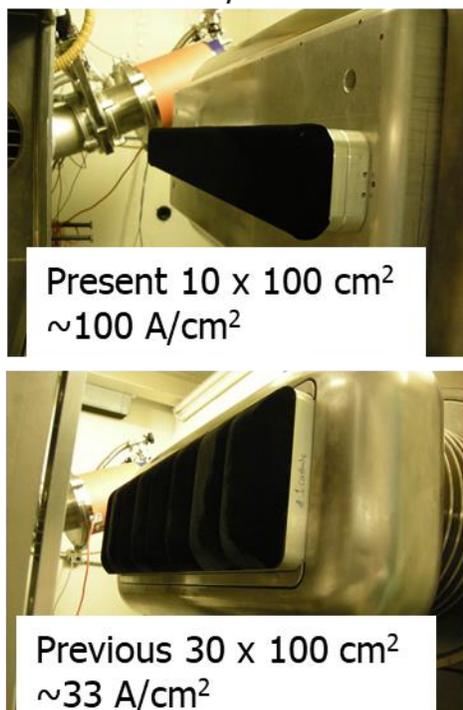


Figure 3. Modifications to the Electra electron beam laser system to allow higher current density and higher electron beam power deposition per unit volume with a cathode height reduction from 30 cm (below) to 10 cm (above).

Table 1. NRL ArF Laser Energy Records over 100 J

Date	Energy (J)	Location
9/17/2019	137 J	NRL
11/15/2019	172 J	NRL
11/19/2019	173 J	NRL
2/14/2020	186 J	NRL
3/5/2020	200 J	NRL

3.2 ArF Orestes Code

The second significant technical achievement was modeling of the ArF electron beam amplifier with Orestes Code was a multi-step process. The first module of the code, which is to solve the electron Boltzmann equation for the Electron Energy Distribution Function (EEDF), was completed [49]. The EEDF determines the electron kinetics which includes electron and ion formation rates, excitation, ionization, attachment, and all other kinetic rates directly related to electrons. The electron beam energy deposition and collisional reaction rates with electrons were calculated from the electron energy distribution function

for a wide range of e-beam deposition powers ($P_{\text{beam}}=10 \text{ kW/cm}^3\text{--}3 \text{ MW/cm}^3$) and fluorine concentrations ($x_{\text{F}_2}=0.01\text{--}10\%$). This work resulted into a refereed journal article publication. The plasma chemistry module for the electron beam pumped ArF laser has been developed to examine the formation and destruction of molecules and accounts for all species within the laser gain medium. Estimates of the pertinent molecular, atomic, ion and electron reaction rates have been made with the best literature values known to date. The combination of the EEDF module and the plasma chemistry module give a predictive tool for an electron beam argon fluoride laser. The predictive capability has been developed for the range of conditions of the Electra electron beam pumped ArF laser. This predictive capability has been shown [52,53]. Numerical simulation tools are used to improve basic understanding of the ArF plasma kinetics, which is important for achieving high efficiency lasing. Numerical simulations were done by the Orestes code, which was originally developed for KrF lasers, and modified for ArF laser development on Electra e-beam facility. The lasing media has complex geometry with mirrors, windows and walls from different materials, that are taken into account for transmission and reflectivity in the laser propagation equation. These data are experimentally determined and used in Orestes as input parameters. Lasing is modeled by laser propagation equation, numerically solved together with the species balance equations. We performed detailed studies of the upper laser population and determined the most important gain and loss reactions, as well as absorption calculation analyses that are based on time-dependent 1D rate equations solutions. In order to perform these analyses, a series of codes have been developed to extract and plot data from Orestes. This enabled parametric studies of laser properties as a function of e-beam power, gas pressure and gas composition. Radiation transport module in Orestes accounts for spontaneous radiative decay and stimulated emission. A number of kinetics models have been explored in order to understand the sensitivity of plasma absorption and excited species generation. It was found, that the stepwise ionization of argon is one of the dominant processes affecting the gain calculations because the main absorber of laser radiation are the excited argon atoms, Ar(4p). A number of auxiliary codes have been developed to study different reaction rates as a function of the electron temperature. The non-equilibrium plasma kinetics also require accounting of the gas heating during the e-beam pulse.

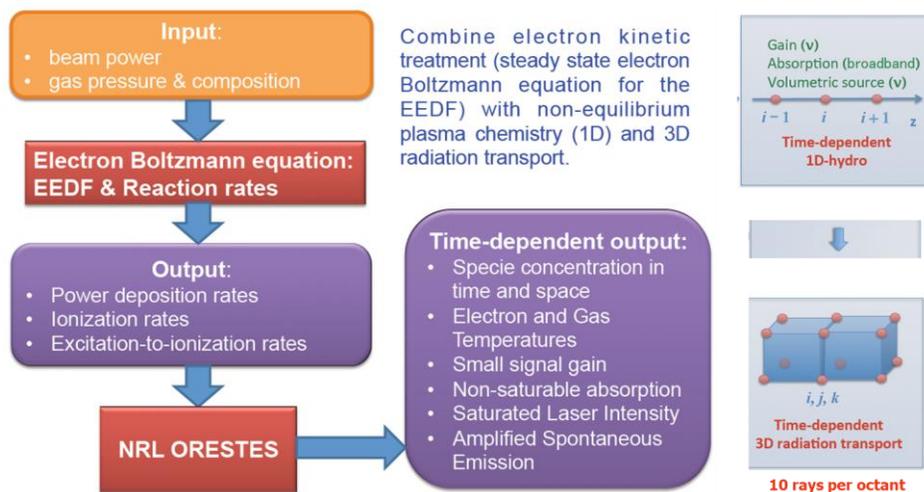


Figure 4. Pictorial representation of the ArF Orestes code development, which was a significant component of this project.

3.3 Measurements and Modeling of Laser Parameters (g_0 , I_{SAT})

The third significant technical measurements and modeling of laser parameters including laser efficiency, small signal gain (g_0), and saturation intensity (I_{SAT}). Small signal gain measurements for the electron beam

pumped ArF laser have been attained over a range of pressures (12 psi – 20 psi) utilizing argon and fluorine gas. A 12 ns laser pulse from a commercial discharge laser was used as seed laser. By varying the input intensity of the seed laser using neutral density filters over a limited aperture of the electron beam pumped volume $30 \times 30 \times 100 \text{ cm}^3$ determination of the small signal gain could be determined. The measurements were made in both single pass mode, a single pass of the 1 meter long gain medium, as well as double pass mode, a passing the initial laser seed back and forth across the media to get larger output signals. The laser intensity was measured with calibrated photodiodes with known, and measured, 193 nm transmission interference and neutral density filters. The energy deposition into the gas from the electron beam was measured by a pressure transducer for all of the 100's of laser shots to determine the small signal gain. The energy deposition then was used to estimate the power deposition using the current and voltage pulse from the electron beam with a finite voltage cut-off due to the propagation of the electron beam through a 25 micron stainless steel foil. A modified Rigrod analysis was used to evaluate the small signal gain. The resultant small signal gain measurements are from single pass measurements 1.84%/cm at 12 psi with a power deposition of 0.395 MW/cc, 1.77%/cm at 16 psi with a power deposition of 0.497 kW/cc, and 1.29% at 20 psi with a power deposition of 0.583 MW/cc. All of these conditions had ~99.7% Ar and 0.3% F₂. An important note is that the ArF electron beam modeling code had a predictive result of ~1.8%/cm in this electron beam deposition range. In addition, we made double pass gain measurements to attempt to determine the saturation intensity. The results from the single pass gain measurements were not reaching the high enough intensities to determine the saturation intensity. At these low gains a double pass measurement were closer in creating enough intensity, but did not have sufficient gain to get to high enough intensities to accurately determine saturation intensity. We determine the small signal gain using the modified Rigrod analysis similar to single pass gain and discounting the transmission through the rear window twice, reflectivity of mirrors at 193 nm, and losses in short path of air. The small signal gain measurements for the double pass measurements are 1.37%/cm at 12 psi with a power deposition of 0.395 MW/cc and 1.10% at 20 psi with a power deposition of 0.583 MW/cc. The analysis gives a lower result due to assumptions that the optical transport was 100% from exiting the amplifier and being put back in the amplifier in the double pass, which we know is inaccurate. Further refinement of the modified Rigrod will be complete to analyze the raw data to determine the actual small signal gain measurements. The double pass configuration was also more challenging due to lower input intensity levels competed with amplified spontaneous emission (ASE) which lowered the measured gain. Therefore in order to evaluate the potential of electron beam pumped ArF lasers modification to increase the electron beam power deposition. The $30 \times 100 \text{ cm}^2$ cathodes were replaced with $10 \times 100 \text{ cm}^2$ cathodes. The modification included designing and fabricating new cathodes as well as modification of the anode and cathode spacing to maintain a nominal 500 keV with increased current density. After installation of the new cathodes with reduced height double pass signal gain measurements were made at 12 psi with ~99.7% Ar and 0.3% F₂. The small signal gain was significantly larger at 3.21%/cm (even with the lower performance double pass measurements as explained above) with an estimate power deposition of 1.09 MW/cc. The small signal gain measurements are consistent with the electron beam pumped ArF laser modeling predictions made earlier in the year. These experimental results were shown [54]. The saturated intensity from the double pass with higher gain could be estimated and suggest that the intrinsic efficiency is higher than KrF lasers. Further measurements show that ArF laser intrinsic efficiency (laser energy out/electron beam energy deposited for the flat-top region of the electron beam) is consistent with previous literature measurements of 17-18% [24,26,54]. This substantially higher intrinsic efficiency suggests the possible wall-plug efficiency for a full scale large electron beam pumped ArF laser could be 10% using the same evaluate techniques to determine the KrF wall plug efficiency is 7% [55].

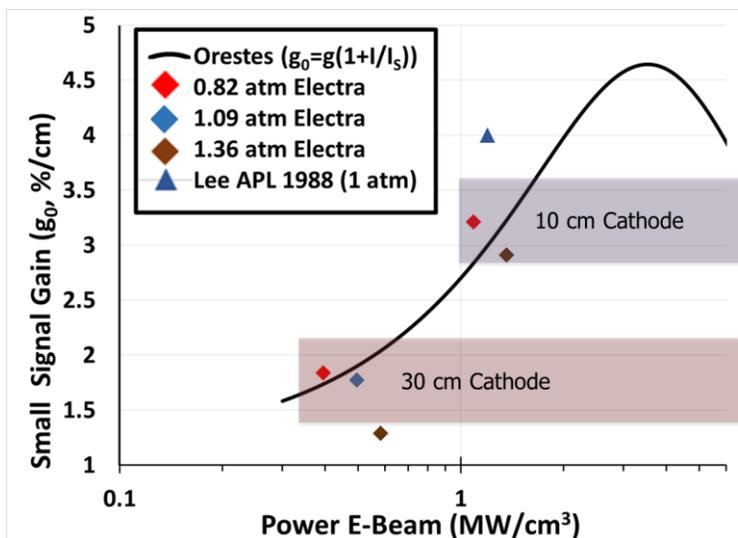


Figure 5. Measured small signal gain from the Electra facility using two different cathodes (10 cm, higher electron beam power and 30 cm, lower electron beam power). Comparison of the ArF Orestes code (solid line) with the Electra results as well as results from Lee *et al.* [26].

4. NEXT STEPS

The next steps are to develop a pulse power system optimized for an ArF laser as opposed to KrF laser. This system would have a lower voltage to better match the lower stopping power of the ArF laser gas mixture. Development of an electron beam deposition model with varying magnetic field would aid in designs of the laser cell. In addition, the laser cell be elongated in the electron beam propagation direction to efficiently use all the electron beam deposition power. The current density would be higher above 1 MW/cc to get substantial gain per centimeter for amplifier performance unless close to saturation intensity. Higher transmission windows as well as an antireflection coating would substantially improve performance. The volume around the amplifier would be able to remove oxygen to avoid absorption losses at 193 nm. For repetition-rate performance the ArF laser has similar technologies as KrF. The advancement of solid state switches for lifetime and the ability to use superconducting magnets for wall plug efficiency. One aspect is unclear whether lower density with higher velocity in gas flow improves or decreases effectiveness of convection foil cooling. Front end technologies for excimer laser need to be confirmed for ArF. The most important next step is getting sufficient funding for advancements of inertial fusion energy drivers.

5. CONCLUSIONS

The argon fluoride (ArF) laser has made substantial progress with modest investment. The resultant work has created the world's largest ArF laser at 200 Joules, and a modeling development which shows large fusion scale amplifiers would be substantially more efficient than KrF. The future is bright for ArF research taking off due to recent developments. The ArF laser has the greatest potential as a fusion driver for high gain from inertial confinement fusion (ICF) and inertial fusion energy (IFE). The excimer wall plug efficiency of ArF is 10%, which is substantially higher than KrF (7%). The qualities of ArF makes it not only the best laser driver, but most likely the best of known drivers for IFE applications.

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