INFORMATION TO USERS

While the most advanced technology has been used to photograph and reproduce this manuscript, the quality of the reproduction is heavily dependent upon the quality of the material submitted. For example:

- Manuscript pages may have indistinct print. In such cases, the best available copy has been filmed.

- Manuscripts may not always be complete. In such cases, a note will indicate that it is not possible to obtain missing pages.

- Copyrighted material may have been removed from the manuscript. In such cases, a note will indicate the deletion.

Oversize materials (e.g., maps, drawings, and charts) are photographed by sectioning the original, beginning at the upper left-hand corner and continuing from left to right in equal sections with small overlaps. Each oversize page is also filmed as one exposure and is available, for an additional charge, as a standard 35mm slide or as a 17"x 23" black and white photographic print.

Most photographs reproduce acceptably on positive microfilm or microfiche but lack the clarity on xerographic copies made from the microfilm. For an additional charge, 35mm slides of 6"x 9" black and white photographic prints are available for any photographs or illustrations that cannot be reproduced satisfactorily by xerography.
Broadband tunable excimer laser studies

Zhu, Yunping, Ph.D.

Rice University, 1987
**PLEASE NOTE:**

In all cases this material has been filmed in the best possible way from the available copy. Problems encountered with this document have been identified here with a check mark √.

1. Glossy photographs or pages ______
2. Colored illustrations, paper or print ______
3. Photographs with dark background ______
4. Illustrations are poor copy ______
5. Pages with black marks, not original copy ______
6. Print shows through as there is text on both sides of page ______
7. Indistinct, broken or small print on several pages √
8. Print exceeds margin requirements ______
9. Tightly bound copy with print lost in spine ______
10. Computer printout pages with indistinct print ______
11. Page(s) ___________ lacking when material received, and not available from school or author.
12. Page(s) ___________ seem to be missing in numbering only as text follows.
13. Two pages numbered ______. Text follows.
14. Curling and wrinkled pages ______
15. Dissertation contains pages with print at a slant, filmed as received _______
16. Other__________________________________________________________

__________________________________________________________

__________________________________________________________

University Microfilms International
RICE UNIVERSITY

BROADBAND TUNABLE

EXCIMER LASER STUDIES

by

YUNPING ZHU

A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE

DOCTOR OF PHILOSOPHY

APPROVED, THESIS COMMITTEE:

F. K. Tittel
F.K. Tittel, Prof. Elec. and Comp. Eng.
Chairman

R. Sauerbrey
R. Sauerbrey, Asst. Prof.
Elec. and Comp. Eng.

G.P. Glass
G.P. Glass, Prof. Chemistry

Houston, Texas

April, 1987
BROADBAND TUNABLE EXCIMER LASER STUDIES

by

Yunping Zhu

ABSTRACT

The principal purpose of this work has been the studies of the broadband tunable excimer laser namely XeF(C-A). Wideband tunability of this excimer laser was first demonstrated with a simple compact dispersive stable cavity in a transverse e-beam pumped geometry. More recently, a series of experiments has been performed to control the XeF(C-A) by dye laser injection. In any low-gain, short-pulse laser systems, rapid build-up of the optical field within the resonator is critical to good laser performance and extraction efficiency. Simultaneous injection of a "seed" signal into the cavity with laser pumping excitation can create a much faster increase in the intensity of the optical field. This rapid build-up of the optical field will aid the competition against radiative emission from the high-gain XeF(B-X) transition and non-radiative quenching of the XeF excimers by halogen donors and other constituents of the laser gas mixture.

Efficient, ultranarrow (0.001 nm to 0.005 nm) spectral output from an electron-beam excited XeF(C-A) laser medium (Ar/Kr/Xe/F₂/NF₃ mixture) has been observed by injection tuning. Amplification of an injected tunable dye laser pulse was achieved throughout the entire blue-green spectral region from 435 nm to 535 nm. Several different confocal unstable resonator geometries with magnification of 1.05 to 1.23 were investigated. A maximum output of about 85 mJ was measured
at 482.5 nm for a cavity with magnification $M = 1.1$, which corresponds to an energy density and intrinsic efficiency of 4.7 J/liter and $\sim 5\%$, respectively. These values are comparable to those of UV rare gas-halide lasers.

In order to gain a better understanding of the injection control process in XeF(C-A) laser, a semiempirical model using a pulsed regenerative amplifier approach has been established with good accuracy. A set of coupled rate equations are used for this purpose. Some key factors in cavity and pumping source design are addressed.
ACKNOWLEDGEMENTS

I would like to thank the many people whose assistance made this work possible. Dr. Tittel advised me throughout my work, and provided many ideas and helpful suggestions during the progress of the experiment. Dr. Sauerbrey was always available to provide assistance whenever I had difficulties. Much of the analysis in chapter 4 is under his guidance. Dr. Wilson spent many hours in the lab providing assistance with both hardware and software, troubleshooting, and doing maintenance. Without his time this work could not have been accomplished. Bill Nighan provided many interesting ideas and Dr. Marowsky paid us a few visits to highlight the experiment, especially the work shown in section 3.1. Jim Hooten did lots of work in keeping the lab in great shape. N. Nishida spent many hours during the early stage of the experiment. All of the members of my thesis committee spent considerable time reviewing the draft and suggesting improvements.

Finally, I must thank my wife Liqun for typing of this thesis. Without her love and encouragement, this work, which I dedicate to her, would not have been possible.

This work is supported in part by the Robert A. Welch Foundation, the Office of Naval Research, and the National Science Foundation.
| Chapter 1 | Introduction | ........................................... | 1 |
| Chapter 2 | Experimental Apparatus | ......................................... | 8 |
| | Electron-Beam Accelerator | ......................................... | 8 |
| | Reaction Cell | ......................................... | 11 |
| | Laser Resonator Optics and Dye Laser System | ................................ | 14 |
| | Diagnostics | ......................................... | 16 |
| Chapter 3 | Experimental Observations | ................................... | 21 |
| | Wideband Tuning by Prism | ........................................ | 21 |
| | Injection Control of the XeF(C+Δ) Laser Output | ................................ | 28 |
| | Injection Control of Xe₂Cl Laser | ...................................... | 54 |
| | Laser Efficiency | ......................................... | 58 |
| Chapter 4 | Model Investigations | ........................................ | 64 |
| | Analytical Model | ........................................ | 64 |
| | Results and Discussion | ....................................... | 69 |
| Chapter 5 | Future Directions and Conclusions | .................................. | 79 |
| | Future Directions | ........................................ | 79 |
| | Conclusions | ......................................... | 84 |
| Bibliography | ................................................ | 86 |

LIST OF FIGURES

1. XeF potential energy curves.................................2

2. Time integrated fluorescence spectra from
   XeF(C→A) and XeF(B→X) transitions........................4

3. Electron-beam generation system............................9

4. Reaction cell and intracell resonator....................12

5. Experimental apparatus.....................................15

6. Timing controls and monitors for injection experiment...19

7. XeF(C→A) fluorescence (a) and laser spectra (b)
   with NF$_3$ as the only fluorine donor and the laser
   spectra (c) of the optimized gas mixture..................23

8. The narrowband XeF(C→A) laser output spectra obtained
   with 2.2 mm iris (center) and 5 mm one (sideband).........25

9. Geometry of Brewster-angled quartz prism tuner..........27

10. The experimental set up for the measurement of bandwidth....33

11. Fabry-Perot etalon interferogram of a typical amplified
    output as observed using the OMA vidicon..................34

12. Temporal net gain of XeF(C→A) at 488 nm..................36

13. Wavelength dependence of the peak values of gain and
    initial absorption for XeF(C→A) mixture with (b)
    and without (a) Kr...........................................38
14. Temporal relationships of the dye laser, the e-beam pulse, the amplified XeF(C-A), and the free-running output..............40

15. Dependence of the amplified XeF(C-A) output energy on the delay time between a 482.5 nm dye laser pulse and e-beam firing.............................................42

16. The free running XeF(C-A) laser spectrum with temporal uncorrelated dye laser input pulse.........................44

17. The variation of the ratio of the injection controlled XeF(C-A) laser pulse vs. emission of the free running system for different timing........................................45

18. The XeF(C-A) temporal absorption/gain behavior at 435 nm, 445 nm, and 455 nm........................................46

19. Dependence of the 482.5 nm amplified output pulse on cavity magnification for optimum timing and 1 mJ injection........48

20. Qualitative comparison of superimposed spectra of XeF(C-A) fluorescence, the injection-controlled outputs at five wavelengths and free-running output.................................51

21. Amplified XeF(C-A) output as a function of wavelength with free-running spectrum as comparison..................52

22. Xe₂Cl laser power density as a function of wavelength......57

23. The amplifier gain of the injected dye laser throughput as a function of the throughput........................................59

24. The e-beam energy received by calorimeter as a function of Ar pressure........................................62

25. The positive branch confocal unstable cavity and the e-beam pumping region..................................67
26. XeF(C+D) output pulse energy dependence on the injected dye laser energy. The squares are the experimental results for cavity magnification of $M = 1.23$.

27. The amplified spectra of Ar$^+$ laser with XeF(C+D) spectra used for Fig.28.

28. The ratio of peak amplified Ar$^+$ line to XeF(C+D) laser spectra as a function of Ar$^+$ laser power.

29. Dependence of the laser output on pumping length from analytical model with a total e-beam current of 24 KA.

30. Dependence of the laser output on cavity magnification from the analytical model with pumping length of 20 cm.

31. Schematic of the scaled up laser cell.
CHAPTER 1

INTRODUCTION

This thesis describes broadband tunable XeF(C→A) excimer laser studies with special emphasis on efficient energy output and wavelength tunability. The motivation for developing such a broadband tunable excimer laser and other tunable sources is their potential applications in various field of science and technology such as optical communications, LIDAR, photochemistry, and ultra-short laser pulse amplification.

The potential energy curves for XeF system are shown in Fig. 1. Even though the B and C states were known to be very close in energy, early studies ignored the possible role of the XeF(C) state [1]. The XeF(C) state has acquired importance in its own right by the demonstration of lasing on the XeF(C→A) system in later 1970's. The broadband laser emission from the bound-free transition at 485±30 nm was first observed by Bischel et al. [2] with photodissociative excitation of XeF₂, by Ernst and Tittel [3] in an e-beam pumped Ar/Xe/NF₃ mixtures and by Fisher et al. [4] and Burnham [5] in a discharge pumped He/Xe/NF₃ mixture. It is accepted that the XeF(C) state is below the XeF(B) state by ~750 cm⁻¹[6,7]. Therefore, in thermal equilibrium at room temperature more than 95% of the combined population of the B and C states resides in latter. This makes XeF(C→A) a good candidate for such application where an efficient broadband tunable laser in the blue-green region is required. A typical time integrated fluorescence spectrum for e-beam pumped mixture comprised of Ar/Xe/NF₃/F₂ shows both XeF(B→X) and (C→A)
Figure 1. XeF potential energy curves

\[ \tau_{B-X} \approx 10 \text{ ns} \]
\[ \tau_{C-A} \approx 100 \text{ ns} \]
transitions (Fig. 2). Charge and excitation transfer to Xe from ionized and excited Ar and ion recombination and excited state interaction by $F^-$ and by $F_2$ and NF$_3$, respectively, result in very efficient formation of XeF(B,C) as described in detail in chapter 4 of reference 8.

Multipass amplification and tuning of the XeF(C$\rightarrow$A) laser was realized by injecting a dye laser into a stable resonator [9]. Wavelength tuning was also accomplished by rotating a prism between two 99% reflecting mirrors [10]. At first the Rice group demonstrated broadband tunability of the C$\rightarrow$A transition using a wideband maximum reflector and output couplers centered at different wavelength between 450 nm to 515 nm. Subsequently, an intracell Littrow prism or a high efficiency grating was also used to tune the laser output, but only in a longitudinally pumped cell with increased gain length [11] due to non-optimized gas mixture conditions at that time.

By the end of 1982, improved output power and spectral characteristics were achieved at Rice University for an electron beam pumped XeF(C$\rightarrow$A) laser by selective tailoring of the relevant kinetic processes [12]. Using a mixture of Xe in combination with two halogen donors NF$_3$ and F$_2$, along with a high pressure Ar buffer, a laser pulse energy density of 0.1 J/l was obtained.

Significantly improved XeF(C$\rightarrow$A) laser performance has been achieved using optimized output coupling and tightly focused resonator which allows a higher level of initial laser flux arising from spontaneous emission [13] and using multi-component gas mixture/buffer (NF$_3$/F$_2$/Xe/Kr/Ar) that has permitted synthesis of a medium with near
optimum properties for which XeF(C) is produced efficiently while transient atomic and molecular absorptions are minimized. This approach has yielded a laser pulse energy density and intrinsic efficiency (laser output/e-beam deposition) of 3J/litre and 3.3%, respectively, values that are comparable to those of wavelength shifted B→X rare gas halide excimer lasers [14 and also in Appendix A].

More recently, wideband tunability of the XeF(C→A) excimer laser in the spectral range of 447 to 524 nm was demonstrated with a simple compact dispersive cavity in a transverse e-beam pumped geometry. By using the above mentioned multicomponent mixture, a laser output energy density of 0.1 J/l at the center of the tuning range at 485 nm and a spectral bandwidth as low as 1 nm had been obtained [15]. But the intracavity tuning technique has some limitations. First, alignment and tuning of the intracell prism can be complex. The second disadvantage arises from the very limited number of cavity round trips which are available for amplification of the desired radiation mode from spontaneous emission during the relatively short period of XeF(C→A) gain duration (~ 30 ns). The dispersive element inside the cavity will also cause loss to the laser output which is especially undesirable for low gain laser like XeF(C→A).

In an effort to overcome these limitations that are observed for a XeF(C→A) excimer laser using a stable resonator and to achieve optimized output performance, a series of experiments have been performed that make use of injection control by means of an external seed laser [16,17]. In any low-gain, short-pulse laser system, rapid build-up of
the optical field within the resonator is critical to good laser performance and extraction efficiency. Simultaneous injection of a "seed" signal into the cavity with laser pumping excitation can create a much faster increase in the intensity of the optical field, compared to one that results from a build-up of spontaneous emission. This rapid build-up of the optical field will aid the competition against radiative emission from the high-gain XeF(B-X) transition and non-radiative quenching of the XeF excimers by halogen donors and other constituents of the laser gas mixture.

A stable resonator generally offers small active mode volume for energy extraction. On the other hand, injection control with an unstable resonator can fully utilize the resonator volume to achieve better energy extraction efficiency (laser output/energy stored in XeF(C)). For a homogeneously broadened transition, the injection of a short pulse laser within the bandwidth of the optical gain allows the maximum extraction of the stored energy into the bandwidth of the injected signal therefore to achieve tuning with much more simplicity. The far-field beam quality will also be improved due to the reduced beam divergence by means of a confocal unstable resonator.

Efficient, ultranarrow spectral output from an electron-beam excited XeF (C-A) laser medium has been observed by injection controlled tuning [17]. Amplification of an injected tunable dye laser pulse was achieved throughout the entire blue-green spectral region from 435 nm to 535 nm. Several different confocal unstable resonator geometries were investigated. A maximum XeF(C-A) laser output of ~ 85 mJ was measured at
482.5 nm for a cavity with magnification M = 1.1, which corresponds to an energy density and intrinsic efficiency of \( \sim 4.7 \text{ J/liter} \) and 5%, respectively. These values are comparable to those of the wavelength fixed XeF(B-X) excimer lasers. Such broadband transition can be used in ultra-short laser pulse amplification down to \( \sim 10 \text{ femto-second} \) range [18,19].

Details of the experimental set-up including electron-beam accelerator, reaction cell, laser resonator optics, and related diagnostic apparatus used in this investigation are summarized in Chapter 2. In Chapter 3, the experimental observations are analyzed which includes wideband tuning of XeF(C+X) laser by both internal prism and dye laser injection control. In order to gain a better understanding of the injection control process, an analytical model has been developed using a pulsed regenerative (or folded) amplifier approach. A set of coupled rate equations were used for this purpose. The results of numerical integration of the coupled rate equations is presented in Chapter 4 and which can be compared to experiments. The possibility of scaling this excimer laser is proposed in chapter 5, as well as potential future directions and conclusions.
CHAPTER 2

EXPERIMENTAL APPARATUS

2.1. ELECTRON-BEAM ACCELERATOR

The high energy e-beam used to excite the rare gas halide mixture was provided by a Physics International Pulserad 110 electron accelerator. Basically, the Pulserad system consists of five subassemblies: a dc power supply and controls; a trigger source; a Marx bank; a coaxial Blumlein pulse generator and a field emission diode.

The Pulserad uses a Marx bank to store energy and to develop high voltages. This bank is composed of capacitors, spark gaps, and resistors electrically arranged as shown in Fig.3. The Marx bank has 10 stages with a 0.01875 μF capacitor in each stage. These capacitors are charged in parallel by a ±50 KV dc power supply with total stored energy of near 1000 Joules. Although the e-beam machine is capable of producing beams delivering up to 200 Joules, only ~20 Joules was normally coupled into the laser cell. ~1.62 Joule was deposited into a typical laser mixture in a cavity volume of 18 cm³ with optimized laser output of ~85 mJ. Each capacitor is shunted by isolation resistors. Breakdown of the spark gaps initiated by a trigger pulse connects the capacitors in series and results in voltage multiplication. The triggering spark gaps can handle up to 10,000 shots without cleaning. These spark gaps are filled with 30 to 34 psi of SF₆ gas. In the normal procedure, the spark gaps should be purged with 40 psi SF₆ for 20 minutes in the beginning of a working day.
Figure 3. Electron-beam generation system
Both $\text{SF}_6$ pressure and charging technique for the Marx bank capacitors will influence the e-beam firing delay time after triggering (therefore the jitter level). With higher $\text{SF}_6$ pressure, the e-beam delay time will be longer until it cannot be fired any more. The e-beam jitter level will be higher. On the other hand, lower $\text{SF}_6$ pressure will advance the e-beam firing time until it breaks down by itself. Similarly, lower charging voltage will delay the e-beam firing and vice versa. The charging voltage is determined by the final voltage observed just before triggering, the charging rate, as well as how long to wait for firing the e-beam after charging up. In order to reduce the e-beam jittering to a minimum level the $\text{SF}_6$ pressure should be minimized. The charging rate should be steady by keeping the charging current constant and the final charging voltage of nominally 100 KV should be reached evenly.

The field emission diode consists of a graphite cathode, which is connected to the Blumlein inner cylinder, and a grounded steel mesh serves as the anode. During e-beam operation, electrons are accelerated from the cathode of 7.5 cm length towards the highly-transparent steel mesh. They must further pass through a 2 mil titanium pressure foil in order to enter the laser cell. The Ti foil must be supported by a Hibiachi plate since it separates the diode vacuum ($<10^{-5}\text{torr}$) and the laser cell pressure ($>6\text{ atm}$). The foil should be thin enough to permit high electron transmission through it and thick enough to withstand high pressures. The distance between the graphite cathode and mesh is also important since it affects the field profile and the impedance matching between the Blumlein circuit and the diode. A cathode to mesh
spacing of 0.5" to 0.6" has been typical in these experiments.

Several monitor outputs which characterize the performance of the e-beam accelerator are usually recorded as depicted in Ref.8. These monitors include Marx bank voltage monitor, Rogowski coil and Faraday cup. The current density at the optical axis of the laser cell is about 200 - 300 A/cm², with duration of about 10 ns. The e-beam pulse appears approximately 1 μsec after the initial trigger pulse is applied to the first spark gap with a jitter time of 20-40 ns depending on the SF₆ pressure in the spark gaps.

Recently, improved laser media pumping using electron beam concentration by means of a backscattering mirror has been realized. This backscattering mirror increases the electron-beam pumping density by as much as a factor of three at low gas pressure conditions [20].

2.2. REACTION CELL

The electron beam is injected through the Ti foil into a high pressure reaction cell. The cell is shown schematically in Fig.4. The design of the cell is described in detail in reference 21. The cell is capable of handling pressures up to 15 atm. But because of foil breaking problems, we usually limit the pressure to 10 atm and use typically a total pressure of 6.5 atm. Because of the all-stainless steel construction, corrosive gases such as F₂ can be handled safely. Two antireflecting coated sapphire windows have been mounted at the two ends of the cell. The laser mirrors are mounted inside the cell with mirror holders which are connected to bellows for easy external alignment of optical
Figure 4. Reaction cell and intracell resonator.
cavity. An extra port is available to allow insertion of such devices as Faraday cup or calorimeter.

The cell is connected to a versatile gas/vacuum system from which it can be isolated by means of valves located close to the cell. A Baratron gauge is used together with a mechanical gauge to provide precise measurements in the pressure range used for most halogen donors and rare gases. The manifold allows convenient selection of gases, and other gases may be easily connected to the system (up to 6 different gases). By using both a mechanical vacuum pump and a diffusion pump, the system can be evacuated to 1 mtorr and the cell to <10^{-4} torr before each gas fill.

The stainless steel reaction cell was carefully passivated by prolonged exposure to F_{2} prior to any experiment. Since the quenching and absorption by even a small amount of impurities can be severe, the quality of the gases used and the filling technique are important factors in obtaining consistent experimental results and good laser performance. The rare gases used (Kr and Xe) were of research grade ( > 99.995% pure as specified by the vendor), the buffer gas (Ar) used was of ultra high purity ( > 99.999% pure). Halogen donors such as NF_{3} were of technical grade (with a purity of 97.5%). For safety reason, F_{2} was used as 10% mixture with the other 90% being He. Knowing the relative volume between manifold and cell, NF_{3}, F_{2}, Xe, Kr, etc. were allowed to flow into the manifold slowly to a certain pressure with a precision of 0.1 torr. The buffer gases were blown into the cell through high flow regulators to allow turbulent mixing of the various components. For a new Kr contain-
ing mixture which has only small amount of F_2, the gas mixture can be used for more than ten shots without much deterioration.

2.3. LASER RESONATOR OPTICS AND DYE LASER SYSTEM

The arrangement shown in Fig.4 has several features which are important for achieving optimized laser output from low gain media. The laser resonator optics were placed inside the cell, thereby eliminate reflection losses which would occur at window surface in the case of external optics. Internal optics also allow use of a short cavity length to reduce absorption by unpumped gas and to achieve faster laser build-up.

The overall experimental apparatus used in this work is shown in Fig.5. For XeF(C≡A) laser injection control experiments a positive-branch confocal unstable, intracell optical resonator was used, typically consisting of a concave end mirror, with an injection-hole of 1.5 mm in diameter and a broadband high-reflectivity coating in the blue-green region of interest and a convex output coupler, with focal lengths f_2 and -f_1, respectively. These mirrors were separated by a distance L = f_2 - f_1 \approx 12.5\text{cm}. The output coupler is a meniscus lens of zero power, on the center of the convex side of the meniscus is a highly reflective coated spot of 14 mm in diameter. In this way, the output coupler can be placed in a normal mirror holder and aligned externally. The active region was a cylindrical volume defined by the clear aperture 14 \times M \text{ mm} (M = \frac{f_2}{f_1} is the cavity magnification) and the e-beam pumping length (10 cm).
Figure 5. Experimental apparatus
Because the e-beam accelerator produces strong vibration, the resonator must be aligned after every laser shot. The bellow mounts shown in Fig. 4 can also be used to hold other different cavity optics such as mirrors, Littrow prisms, gratings, apertures, etc. The length of the cavity could be varied by tightening or loosening the bellow, or by adding suitable spacers to the mirror holders.

Since the optics are exposed to the laser medium, the high reflectivity dielectric coatings must be protected from attack by corrosive gases such as fluorine. Both SiO₂ and Al₂O₃ protected optics have been successfully used in studies involving a fluorine environment. Also, the e-beam should not be fired in vacuum with mirrors in the cell since the energetic electrons will cause arcing across the mirror surface which can damage the mirror.

A excimer laser pumped dye laser system (Lambda Physik model EMG 101E/FL2002) was used in the injection control investigation. The bandwidth of this dye laser was measured to be 0.005 nm (0.001 nm with a FL83 etalon) as shown in section 2 of chapter 3. This seed oscillator delivered an output of up to 8 mJ in a 10 nsec pulse, tunable from 430 nm to 550 nm using Coumarin 2, 102 and 307. A telescope was used to reduce the injection beam diameter so that most of the available dye laser pulse energy could be injected into the cell.

2.4. DIAGNOSTICS

The most important data to be measured in this experiment include the temporal and spectral behavior of injected dye laser, XeF(C-A) free
running output, and injection controlled laser output using the arrangement shown in Fig.5.

The temporal evolution of above signals were monitored by a fast vacuum photodiode detector (ITT-F4000 S5). Neutral density filters were used to avoid the saturation of the photodiode. Color glasses were used to define the spectral region of interest. Temporal signals were recorded by a Tektronix R7912 transient digitizer with a time resolution of better than 2 nsec. The temporally integrated, spectrally resolved laser signal was recorded by an optical-multichannel-analyzer (OMA III), using a Jarrell-Ash 0.25 meter spectrometer model 82-422. The spectral resolution was up to 0.5 nm. The actual injection controlled XeF(C+\text{A}) laser bandwidth was measured directly using a Fabry-Perot etalon (FSR = 0.5 cm$^{-1}$) in order to get an interferogram of the laser signal which can then be focused on the OMA vidicon and processed by computer to get the bandwidth information. Furthermore, the temporal relationship of the dye laser and e-beam pulse was monitored by a storage oscilloscope. The precise timing relation of the dye laser and e-beam was adjusted by the timing circuits so that the volume filling pass of the injected dye laser beam overlapped the XeF(C+\text{A}) gain. In the following, each of these measurement paths is described in more detail.

The OMA is an instrument designed to capture a complete spectrum from single-shot events such as the fluorescence and laser output from the reaction cell. Neutral density filters are required to reduce the light intensity at the detector head. A quartz lens is used to focus the light on the input slit of the monochromator. The low resolution
grating of the monochromator yields approximately 2 nm resolution over a 300-400 nm bandwidth, the high resolution one yields about 0.5 nm over 140 nm. The wavelength separated output from the monochromator is detected by an array of photodiodes. The output of this array, representing light intensity at about 1000 different wavelengths, is sent to the OMA electronics for conditioning, digitizing and processing. The processed information is subsequently transferred serially to an IBM PC-XT computer. The spectrum is combined with calibration data (which is used to draw a wavelength scale) and stored on a diskette for display and reduction.

The vacuum photodiodes are normally biased at -2000 V dc. The output of the diode is carried on low noise Heliax cable through copper conduit to the display instrumentation located inside a bronze-screened Faraday cage. The cage provides isolation from the strong electromagnetic interference generated by the electron beam accelerator. The Tektronix R7912 transient digitizer is used to record the spectrally integrated temporal information from cell output. The digitizer is connected to the PDP 11/23 computer through a fast parallel data bus.

Absolute energy density measurements were made using calibrated vacuum photodiode detector. The calibration was verified by means of a Gentec ED-500/200 Joulemeter and/or Scientech volume absorbing disc calorimeter Model 36-0203. These methods of energy density determination were found to agree to within the estimated experimental error.

Details of the timing controls and monitors for the injection control experiment are shown in Fig.6. The function generator which acts
FUNCTION GENERATOR
~ 1 Hz

HP 214B
PULSE GENERATOR TRIGGER OUTPUT

DYE LASER

TI 6613
PULSE GENERATOR

ONE SHOT

DATAPULSE IOI REFERENCE

OMA AND DETECTOR GATE

INSTRUMENT IN FARADAY CAGE

PULSEPAK IO A SYNC OUT

PULSERAD IIO

TEKTRONIX 7834

TO TO DYE ROGOWSKI LASER COIL

Figure 6. Timing controls and monitors for injection experiment
like a master clock puts out a triggering signal for a HP 214B pulse generator. The output of this generator with variable delay time and duration can be used to trigger the excimer pumped dye laser system. A sample pulse from the excimer laser received by a photodiode can then be used as one of the timing signal observed on the CRT of Tektronix 7834 storage oscilloscope. Another trigger output from the HP pulse generator is fed to a timing delay unit to trigger a Datapulse 101 pulse generator which is enabled by a one shot trigger signal with duration of about 0.3 second. When this one shot trigger pulse overlaid with one of the pulses from the T.I. 6613 pulse generator, a trigger pulse will be generated to trigger both OMA III and Pulsepak 10A to fire the e-beam machine where the output from the Rogowski coil will serve as another timing signal on the CRT of the storage oscilloscope.
CHAPTER 3

EXPERIMENTAL OBSERVATIONS

Broadband wavelength tuning of XeF(C+A) laser has been realized by several groups [9-11] as discussed in chapter 1. This chapter describes the tuning performance and output powers obtained from optimized multicomponent rare-gas halide mixture. The experimental observations which are analyzed include wideband tuning of XeF(C+A) laser by both internal prism and dye laser injection control.

3.1. WIDEBAND TUNING BY PRISM

The experimental apparatus used in this work has been described in Chapter 2. A gas mixture comprised of 8 torr NF₃, 2 torr F₂, 10 torr Xe, 300 torr Kr, and 6.5 atm Ar was excited transversely by an intense electron-beam (1 Mev, 250A/cm², 10 ns FWHM). These were optimized mixture conditions for realizing efficient XeF(C+A) laser operation [14 and Appendix A, B]. Fig.4(b) shows the details of the resonator modified for efficient tuning of the XeF(C+A) laser. A Brewster-angled Littrow prism made of high quality quartz was used as the intracell wavelength selective element and as the flat end reflector with more than 99.9% reflectively between 440 and 530 nm. The output coupler is a concave mirror with large radius of curvature. This reflector utilized a broadband dielectric coating with 4% transmittivity around 485 nm. By placing an iris of suitable diameter in front of the output coupler, the laser
emissions can then be restricted to a few transverse modes. The different Ar ion laser lines can be used to align the cavity conveniently.

The spectrum in Fig. 7 for an optimized mixture shows the potential broadband tunability of the XeF(C-A) laser emission. In the case of untuned two-mirror resonator \( R_1 = \infty, R_2 = 6.7 \text{m}, \) as shown in Fig. 4a, the spectral bandwidth narrows from 80 nm (Fig. 7a) for the fluorescence to 30 nm (Fig. 7c). This figure also shows the comparison of the laser output spectra obtained with NF\(_3\) alone as the fluorine donor (Fig. 7b) and with an optimized multicomponent gas mixture specifically tailored to minimize medium transient absorption arising from various atomic and molecular species (Fig. 7c) as discussed in detail in reference 14 and Appendix A. Such an optimized mixture together with a tightly focused cavity has resulted in an earlier onset of laser oscillations and in more than two orders of magnitude increase in laser energy output. Due to the partial saturation of the absorbers, the structure in the laser spectrum is greatly reduced. With the optimized stable cavity condition \( R_1 = 0.5 \text{ m}, R_2 = \infty \) for a cavity length of 12.5 cm and output coupling of \( T = 5\% \) and an Ar buffer pressure of up to 10 atm, the laser energy density of 3 J/l has been realized [14 and Appendix A]. However, in order to achieve efficient broadband tunability as well as narrow bandwidth, a less tight cavity focusing condition is required than that for optimized energy extraction. Therefore, the cavity configuration was appropriately modified, as shown in Fig. 4a. A variable-size iris in front of output coupler is needed to further reduce the laser bandwidth as shown later. These factors among others (like buffer pressure) reduced the laser out-
Figure 7. XeF(C→A) fluorescence (a) and laser spectra (b) with NF$_3$ as the only fluorine donor and the laser spectra (c) of the optimized gas mixture.
put energy density by a factor of 10 to 0.3 J/l. Replacement of the flat maximum reflector by the Brewster-angled quartz prism (Fig.4b) further reduced the output energy density to 0.1 J/l at the center wavelength of the XeF(C→A) transition mainly due to the additional loss introduced by the prism. However, it increases the spectral brightness by one order of magnitude.

A bandwidth of ~2 nm with an intracavity iris of 5 mm diameter was obtained throughout the entire tuning range as shown in Fig.8 (two side-band spectrum). The bandwidth can be further decreased to 1 nm by inserting an iris with 2.2 mm diameter without loss of output energy density. The bandwidth of 1 nm was determined from the spectrum shown in Fig.8 (the center one) after correction for the finite resolution of the spectrometer and OMA combined system. These two sideband spectra shown in Fig.8 show excellent signal-to-noise ratios. They are far above the amplified spontaneous emission (ASE) background. The tuning range from 446 to 524 nm was limited by an increase of the output coupler and prism tuner coating transmission at these wavelength. Therefore, an overall tuning range of 90 nm from 440 to 530 nm should be possible with optimized wideband dielectrical coatings. The laser output energy in the spectral range from 460 to 520 nm followed the broad gain (Fig.7) with a decrease in output energy of ~20% in the spectral wings compared with that at the center wavelength.

The narrow spectral linewidth of 1-2 nm mentioned above can be explained in terms of the dispersive properties of the low-loss prism tuner and gain narrowing [22]. Furthermore, the channeling of most of
Figure 8. The narrowband XeF(C-A) laser output spectra obtained with 2.2 mm iris (center) and 5 mm one (sideband)
the available energy into such a narrow linewidth is a clear indication that XeF(C-A) transition is homogeneously broadened in the presence of high Ar buffer pressures. Fused quartz was chosen as the prism material instead of a more highly dispersive glass because of its resistance to optical damage by fluorine.

By referring to Fig. 9, the passive bandwidth \( \Delta \lambda_p \) can be derived as a function of a small angle \( \Delta \alpha \) and dispersion \( \frac{dn}{d\lambda} \) as below:

We have \( \beta - \gamma = 90^\circ - \alpha_B \) and \( \tan \alpha_B = n \) as shown in Fig. 9, where \( n \) is the refractive index of fused quartz, \( \alpha_B \) is the Brewster angle for the quartz tuner, \( \beta - \gamma \) is the angle of refraction with angle of incidence being the Brewster angle (\( \alpha_B = 55.646^\circ \) for \( n = 1.463 \)) at 486.1 nm.

\[
\frac{dn}{d\lambda} = \frac{dn}{d\alpha} \frac{d\alpha}{d\lambda} \quad \text{or} \quad \frac{\Delta \lambda_p}{\Delta \alpha} = \frac{dn}{\frac{dn}{d\alpha}}
\]

where \( \frac{dn}{d\alpha} = \frac{1}{\cos^2 \alpha_B} = 3.14 \)

Therefore, we have

\[
\frac{\Delta \lambda_p}{\Delta \alpha} = \frac{3.14}{\frac{dn}{d\lambda}}
\]

with \( \left| \frac{dn}{d\alpha} \right| = 6 \times 10^{-5} \text{ nm}^{-1} \) at \( \lambda = 486.1 \text{ nm} \) [23], an angular dispersion \( \frac{\Delta \lambda_p}{\Delta \alpha} \) of 52 nm/mr is obtained. Since the minimum divergence of a well-aligned resonator with an intracavity iris was diffraction limited (\( \approx 0.1 \text{ mr} \)), we attribute the low final active bandwidth \( \Delta \lambda_a \) to a considerable amount of gain narrowing.

Consideration of the processes for unsaturated amplification within an active medium in terms of a regenerative amplifier yields [22]:
\[ \beta = \gamma = 90^\circ - \alpha_B \]
\[ \tan \alpha_B = n \]

Figure 9. Geometry of Brewster-angled quartz prism tuner
\[ \frac{\Delta \lambda_a}{\Delta \lambda_p} \approx \frac{1}{\sqrt{g(0) L_{\text{eff}}}}, \quad L_{\text{eff}} = C \tau_p \]

where \( g(0) \) is the small signal gain and \( L_{\text{eff}} \) is the effective optical length of the amplifier. With a typical pulse duration \( \tau_p \) of 25 ns and 
\[ g(0) \approx 2.5 \text{ %/cm} \quad [14], \quad \sqrt{g(0) L_{\text{eff}}} \approx 4.3. \] Therefore, a observed bandwidth \( \Delta \lambda_a = 1-2 \text{ nm} \) resulting from the gain narrowing is reasonable.

3.2. INJECTION CONTROL OF THE XeF(C→A) LASER OUTPUT

Even narrower spectral linewidths and larger output energies can be realized with dye laser pulse injection and an unstable resonator configuration. Injection control of excimer lasers has been reported by several groups recent years with narrow bandwidth (< 0.01 nm) for KrF [16], XeF [16, 24] and HgBr [25]. Some UV excimer lasers like XeCl has been Raman shifted to a few discrete wavelengths in the visible region also with narrow bandwidth [26]. These lasers normally have quite high efficiency (≥ 1%), but their tuning ability is limited to about 1 nm (10 nm for HgBr).

Recent developments indicate that the electrically excited XeF(C→A) medium has considerable potential for development as an efficient optical source that is tunable throughout the entire blue-green region of the spectrum based on following factors. 1). Short-pulse, high-energy e-beam excitation of multicomponent gas mixtures (which contains ~ 500 torr of Kr) tailored specifically to minimize transient absorption has resulted in a broadband, free-running laser oscillator output having energy density and efficiency values comparable to those of the UV
XeF(3-X) transition [14]. 2). Using a nonoptimized gas medium [27] as an amplifier, a 482 nm output pulse having bandwidth of only 0.01 nm was obtained using dye laser injection control. Moreover, the narrowed spectral output was obtained at energy density and intrinsic efficiency level of ~ 3 J/l and 3.3 %, respectively. Therefore, with optimized gas mixture as well as optimized cavity magnification [27], a higher energy density can be realized. 3). Recent measurements have shown that the e-beam excited optimized XeF(C-A) medium exhibits relatively high peak gain ( > 2%cm$^{-1}$) throughout the entire 450-515 nm wavelength region (as shown in detail later in this section) indicating that efficient tuning over a broad spectral range should be possible. In this section, efforts to capitalize on these characteristics by injection control of an XeF(C-A) amplifier is described. The technique has resulted in amplified output pulses with energy densities exceeding 0.2 J/l throughout the entire 459-505 nm region with a peak of 4.7 J/l at the center wavelength of 482.5 nm.

I. EXPERIMENT

The transverse e-beam pumping conditions are the same as described in section 3.1. The gas mixture was composed of 6.5 atm Ar, 8 torr Xe, 300 torr Kr, 1 torr F$_2$, and 8 torr NF$_3$. Good mixing of the component gases and thorough fluorine passivation of the stainless steel reaction cell were found to be absolutely essential in order to ensure reliable laser performance. The passivation was carried out by keeping 0.5-2 atm of 10% F$_2$ mixture inside the cell for a few hours or overnight after the cell was opened for a long time during the foil change or cell
maintenance. Even so, it normally took up to one week to optimize the passivation condition with e-beam firing. Recently, a new passivation method was tested that resulted in a substantially shorter time required for passivation. A normal XeF(C→A) mixture was filled into the cell without cavity optics and the e-beam was fired every two minutes instead of every ten minutes for more than ten shots for each gas fill while monitoring the fluorescence behavior with a photo-diode. The laser cell can be passivated in one day or two using this method.

In order to obtain a uniform e-beam distribution and to increase the lifetime of the foil separating the low and high-pressure region of the cell, a stainless steel wire mesh was mounted between the e-beam cathode and the foil.

A. Injection control System

The excimer-pumped dye laser system is the same as shown in chapter 2 (Fig.5). Use of an intracavity etalon in dye laser resulted in significant bandwidth reduction to $9 \times 10^{-4}$ nm compared with $5 \times 10^{-3}$ nm without etalon. Due to the loss of dye laser energy caused by the beam-forming optics as shown in Fig.5, the maximum injected energy was limited to 1.5 mJ in these experiments. The timing relationship between the dye laser and e-beam firing was adjusted by timing circuits so that the volume-filling pass of the injected dye laser pulse overlapped the rise of the XeF(C→A) temporal gain profile.

B. Cavity Optics
The optical cavity used in these studies was a positive-branch confocal unstable, intracell resonator consisting of a concave end mirror with an injection hole of 1.5 mm diameter and a coating having a high reflectivity in the blue-green region, and a convex output coupler (Fig. 5). The mirrors, having focal lengths of \( f_2 \) and \(-f_1\), respectively, were separated by a distance \( L = f_2 - f_1 \), typically about 12.5 cm for our conditions, with the magnification given by the relation \( M = \frac{f_2}{f_1} \). The output coupler was a meniscus lens of zero refraction power having a highly reflective coated spot on the convex side with a diameter \( D \) of 1.4 cm. With this arrangement, the output coupler could be placed in a conventional mirror holder and aligned externally. The active region was a cylindrical volume defined by the e-beam pumping length (10 cm) and the clear aperture having a diameter \( D \times M \) cm.

Because the output coupling of the unstable cavity used in this experiment was larger than the value found to be optimum for a stable resonator (\(-5\%\)), and since oscillations for this type of unstable resonator builds up most readily in the lossless paraxial region [16], and (for these small magnifications) the injection hole constitutes a large loss to the paraxial volume, self-oscillation (free running) is not possible. Measurements showed that the output energy obtained by operating these resonators without dye laser injection was three to four orders of magnitude lower than that obtained using an optimized tightly focused stable resonator under otherwise similar conditions [14]. Thus, the primary role of cavity is to serve as the beam-expanding telescope of a regenerative amplifier. Therefore, the term "injection controlled"
is used rather than "injection locked".

In order to optimize the cavity magnification, various cavities were examined having magnifications of 1.05, 1.08, 1.14, and 1.23, respectively. Although such small values of magnification result in a considerable sensitivity to alignment [28], they provide the long amplification path (several hundred cm) required for efficient use of the XeF(C+O) gain medium.

C. Bandwidth Measurement

Fig.10 shows details of the experimental set up. Part of the output of the unstable, confocal cavity was focused on a diffusing screen (DS) which served to illuminate homogeneously an air-gap etalon of adjustable spacing. By means of a long-focal-length lens ($L_1 = 80$cm) a section of the emerging fringe pattern was imaged onto the entrance-window of the OMA vidicon. Fine alignment of the setup was performed with the 488 nm line of collinear Ar$^+$-laser and (after removal of the attenuators) with the throughput of the tunable dye laser radiation itself. Using an air-gap etalon with free-spectral-range (FSR) of 0.5 cm$^{-1}$, the ultimate resolving power of the whole setup for shot-wise linewidth measurements was 0.01 cm$^{-1}$ or 0.00024 nm at 485 nm. This resolution conveniently allowed a determination of the linewidth of the injected dye laser radiation: 0.2 cm$^{-1}$ (grating tuned) or 0.04 cm$^{-1}$ (grating tuned + intra-cavity etalon of 1 cm$^{-1}$ FSR). Synchronous firing of the e-beam pumped multipass amplifier resulted in essentially the same linewidth. Fig.11 shows as example a typical plot of an OMA III spectrum. The linewidth of 0.001 nm is narrower than the linewidth of homogeneous broadening of the
Figure 10. The experimental set up for the measurement of bandwidth
Figure 11. Fabry-Perot etalon interferogram of a typical amplified output as observed using the OMA vidicon
XeF(C→A) transition (≥0.01 nm) which is mainly contributed by the very fast decay of the repulsive XeF(A) state. With a ultra-narrow injected laser pulse, injection control of XeF(C→A) with a near transform-limited-bandwidth (≈4×10⁻⁵ nm for FWHM = 20 ns) might be realized [16].

II. TEMPORAL GAIN

Short-pulse, high-energy e-beam excitation of the XeF(C→A) medium is characterized by a period of strong transient absorption during the e-beam pulse, followed by the development of net gain in the afterglow regime [13,14]. At first, a relatively short gain duration (< 50 ns) and a relatively low value of the peak gain (< 1% cm⁻¹) limited the energy and efficiency of the electrically excited XeF(C→A) laser to an unacceptably low levels. However, in recent years, multicomponent mixtures have been developed that resulted in significant reduction in the transient absorption effects, thereby permitting much higher peak gain values (~2-3 % cm⁻¹), with a significant improvement in laser performance [13,14,29, see also Appendix B].

A. Role of Kr

The addition of Kr to XeF(C→A) laser mixture has been found to result in a considerable improved gain profile with e-beam excitation [14,29, also Appendix B]. Fig.12 shows the measured temporal evolution of the net gain at 488 nm (solid line) using an Ar-ion probe laser for mixtures comprised of 6.5 atm Ar, 16 torr Xe, 8 torr F₂, 8 torr NF₃, and 6.5 atm Ar, 10 torr Xe, 8 torr NF₃, 2 torr F₂, and 150 torr Kr. The dotted line is an analytical fitting for the Kr containing mixture for
Figure 12. Temporal net gain of XeF(C-A) at 488 nm
purpose of a model investigation as shown in chapter 4. The fractional concentrations of each constituent of the gas mixture are optimized and result in broadband extraction energy density values typically in the 1-3 J/l range with a free-running stable resonator [13,14]. However, Fig.12 illustrates the significant reduction in the initial absorption and the increase in peak gain when Kr is added to the mixture and all constituent fractional concentrations are reoptimized. The improved gain profile typical of Kr-containing mixtures has been found to increase the broadband output energy and intrinsic efficiency of a stable, free-running oscillator to levels comparable to those of the UV XeF(B-X) transition [14]. Fig.13 shows the wavelength dependence of the peak value of gain and initial absorption for mixtures comprised of 6.5 atm Ar, 16 torr Xe, 8 torr NF₃, and 8 torr F₂ (a), and 6.5 atm Ar, 10 torr Xe, 8 torr NF₃, 2 torr F₂, and 300 torr Kr (b). The result for Kr-containing mixture is typical of Kr partial pressures in the 150-600 torr range, and shows that the peak gain is increased significantly and the initial absorption is decreased throughout the entire wavelength range of interest.

In case of dye laser injection control, since the initial absorption by Kr containing mixture is greatly reduced (Fig.12), the energy available for amplification is increased. For this reason, the presence of Kr in the mixture has an even greater beneficial effect when the XeF(C-A) laser medium is used as an amplifier rather than when used as a free-running oscillator [14]. Additionally, for the conditions of Fig.12 and 13 with Kr, the concentration of Kr₂F is very large [29 and Appendix
Figure 13. Wavelength dependence of the peak values of gain and initial absorption for XeF(C-A) mixture with (b) and without (a) Kr
B]. Since the Kr$_2$F excimer is an absorber of UV radiation, its presence significantly reduces the possibility of competitive oscillation on the XeF(B→X) transition [29]. Also shown in Fig.13 is the possibility of a wider injection tuning range for a Kr-containing mixture, especially at shorter wavelengths due to the contributions from Kr$_2$F [29].

III. RESULTS AND DISCUSSION

A. Timing Considerations

Fig.14(a) shows the temporal relationship between the injected dye laser pulse, the e-beam excitation pulse, the injection controlled XeF(C→A) output at 482.5 nm, and the broadband output with the system operating as a free-running oscillator. This figure shows that the injection controlled output appears much earlier and rises much faster than that of the free running laser output due to the powerful initial intracavity photon flux provided by dye laser injection, while the free running laser has to build up from spontaneous emission. Also the large difference in terms of half-width (FWHM) between injection controlled pulse and the slow rise-time pulse of the free running laser allows a differentiation between both events.

Fig.12 and 14 show that the duration of the dye laser pulse and gain rise-time are both on the order of 10-20 ns, a time nearly the same as that required for the injected pulse to fill the cavity. For this reason, control of the timing of the dye pulse with respect to the firing of the e-beam was found to be a difficult and critical aspect of the present experiment. This problem was further complicated by the fact
Figure 14. Temporal relationships of the dye laser, the e-beam pulse, the amplified XeF(C-A) output, and the free-running output.
that a short period of initial absorption accompanies the e-beam firing (Fig.12), although at a much reduced level when using mixtures containing Kr as described previously.

Presented in Fig.15 is the dependence of the amplified 482.5 nm pulse energy on the delay time between the injection of a 1 mJ dye laser pulse and the firing of the e-beam. This figure shows that for magnification M of 1.08, for which approximately 86 percent of the dye laser energy is reflected back out through the injection hole, the temporal window corresponding to optimum timing is only 10-15 ns. For an M value of 1.05, the window was slightly larger, reflecting the fact that the transit time of the dye laser pulse through the cavity increases as M decreases, thereby relaxing the timing constraint somewhat. For the same reason, the optimum temporal window was reduced to about 5 ns for an M value of 1.23.

B. Amplifier Gain

Injection of a tunable dye laser in the unstable cavity leads to a considerable rejection of the dye laser radiation upon incidence on the convex cavity mirror. Depending on the magnification of the cavity, typically more than 80% of the incoming radiation is rejected. Therefore, amplifier-gain G is defined as the ratio of throughput with and without firing of the e-beam machine. In a series of experiments the dependence of G on timing of the dye laser pulse and its wavelength have been studied in detail. Especially for tuning experiments in the wings of the fluorescence band of the XeF(C-A) transition, where the free running system even with broadband mirrors does no longer oscillate, the
Figure 15. Dependence of the amplified XeF(C→A) output energy on the delay time between a 482.5 nm dye laser pulse and e-beam firing.
following procedure has been developed: in order to avoid the large shot to shot variations of the e-beam system, each signal has been normalized against the performance of the free running system (Fig.16). This normalization together with a refined determination of the precise temporal relation between e-beam firing and dye laser injection resulted in considerable improvement of the scatter of the experimental data. Fig.17 shows the variation of the ratio of the amplified dye laser pulse versus emission of the free running system for perfect timing (17c) and injection of the dye laser pulse prior ("too early") and after ("too late") the occurrence of optimized gain conditions. The absolute emission intensity for free running should be same unless under excimer medium saturation condition. Neutral density filters were used in case of Fig.17c in order to avoid the saturation of OMA by the amplified dye laser pulse under optimized timing condition.

During the course of this investigation, it was found that the injection controlled XeF(C-A) output as a function of delay time showed a behavior which can reveal the temporal development of the gain at various wavelengths. Fig.18 shows the large variation between absorption and gain for three selected wavelengths: 435 nm, 445 nm, and 455 nm. It is apparent from this compilation that, even in the short-wavelength wing of the C-A transition, there exists some amplifier-gain, provided the dye laser probing pulse is injected after the decay of the various absorbing species at the leading edge of the fluorescence. These data were taken with the dye laser pulse intensity very much below the saturation intensity of the medium. With increasing wavelength the
Figure 16. The free running XeF(C-A) laser spectrum with temporal uncorrelated dye laser input pulse
Figure 17. The variation of the ratio of the injection controlled XeF(C+0) laser pulse vs. emission of the free running system for different timing
Figure 18. The XeF(C-A) temporal absorption/gain behavior at 435 nm, 445 nm, and 455 nm
amount of absorption decreases and the gain increases.

C. Magnification Effects

In addition to a significant dependence on timing, the amplified output pulse energy also exhibited a strong dependence on cavity magnification for the specific conditions of our experiment. Fig.19 shows the dependence of the 482.5 nm output energy on M for near optimum timing (Fig.15) and a 1 mJ injection pulse energy. As M is reduced from 1.23, the output energy increases, a reflection of the increase in cavity feedback for the lower M values. However, as M approaches unity, nearly all the injected dye laser energy is reflected back out through the 1.5 mm diameter injection hole. Indeed, Fig.19 shows that the output energy decreases rapidly as the magnification is reduced to 1.05. Additionally, optical alignment was found to be very difficult using the M = 1.05 cavity [28].

The other explanation for the optimization of M values might be that the time taken by dye laser to fill the active volume of the cavity should be close to the time duration of XeF(2+2) threshold gain. In our case, the threshold gain duration is about 25 ns (Fig.12). While with injection hole size of 1.5 mm, it took dye laser about 40, 25, and 15 ns to fill the whole active volume for M = 1.05, 1.08, and 1.14, respectively. With an optimized temporal relation between dye injection and e-beam pumping for each cavity, in case of M = 1.08, the dye laser gets fully expanded to obtain the maximum energy extraction efficiency during the threshold gain period. In the case of M = 1.05, the dye laser fills only a small portion of the cavity volume in 25 ns. The extraction
Figure 19. Dependence of the 482.5 nm amplified output pulse on cavity magnification for optimum timing and 1 mJ injection.
efficiency will be low. While for $M = 1.14$, it requires only about 15 ns for the dye laser to fill the active volume. The dye laser could not fully utilize the longer gain duration ($\sim 25$ ns).

D. Output Energy and Efficiency

For the conditions of Fig.15 and 19, the e-beam energy deposition was found to be approximately 90 J/l, a value consistent with calculated values of medium properties as shown in section 4 of this chapter. Since the active volume defined by the mirror spacing and magnification values varied from 16.8 to 20.4 cm$^3$ for our conditions, the maximum of 85 mJ output obtained at 482.5 nm corresponds to an energy density of $\sim 4.7$ J/l and an intrinsic efficiency of approximately 5 percent. On a volumetric basis, these values are actually higher than those typical of room temperature XeF(B$\rightarrow$X) laser operation, and are comparable to XeF(B$\rightarrow$X) performance at the 450 K temperature found to be optimum for that laser [30, 31]. This is rather surprising in view of the fact that the C$\rightarrow$A quantum efficiency is $\sim 25$ percent less than that of the B$\rightarrow$X transition. However, because of the strongly repulsive nature of the XeF(A) state, the C$\rightarrow$A laser does not suffer from lower level population buildup as is the case with the B$\rightarrow$X laser. Additionally, the 6.5 atm Ar-Kr buffer mixture used in the present work results in B$\rightarrow$C state mixing and vibrational relaxation times of approximately 0.5 ns [6], which are very much less than those typical of optimum B$\rightarrow$X laser mixtures using Ne as the buffer at pressure of $\sim 3$ atm [30,31]. Since both lower level buildup and slow vibrational relaxation adversely affect the XeF(B$\rightarrow$X) laser energy and efficiency [30,32], the XeF(C$\rightarrow$A) medium has
advantages in this respect, provided the level of broadband transient absorption is controlled kinetically and the characteristically slow build up of optical flux that usually limits oscillator performance is overcome by using the C>A medium as an amplifier.

E. Wavelength Tuning

In order to determine the maximum possible injection control range, three different laser dyes (Coumarin 2, 102, and 307) were used separately to obtain an effective tuning range from 430 nm to 550 nm. Injection tuning of XeF(C>A) laser was realized from 435 nm to 535 nm. Fig.20 shows the qualitative comparison of superimposed time integrated spectra of the XeF(C>A) fluorescence, the injection-controlled output of five separate shots at several wavelengths, and a typical free-running oscillator spectrum, all for representative conditions. Because of the large cavity loss described previously, the maximum free-running output energy was always less than 0.1 mJ for our conditions. However, several mJ of amplified output were obtained for injection wavelengths as low as 459.4 nm and as high as 505 nm.

Presented in Fig.21 is the measured output energy as a function of wavelength for several different cavities, along with a typical free-running spectrum for comparison. The injected dye laser pulse is approximately 1 mJ. The cavity M values were 1.05 (solid triangle), 1.08 (solid square), and 1.14 (solid circle). The specific injection wavelengths were chosen to correspond to the peaks and absorption valleys that are always apparent in the free-running XeF(C>A) laser spectrum. This figure shows that the wavelength dependence of the minima observed in the
Figure 20. Qualitative comparison of superimposed spectra of XeF(C-A) fluorescence, the injection-controlled outputs at five wavelengths and free-running output.
Figure 21. Amplified XeF(C-A) output as a function of wavelength with free-running spectrum as comparison
amplifier output correlates reasonably well with the locations of the absorption valleys in the free-running spectrum. This is also in agreement with recent gain measurement using flash lamp pumped dye laser which shows the peak gain in the valleys are almost half of that at the peaks. The discrete absorption is due primarily to photo-transitions from $^{3}P_2$, $^{3}P_1$ states of Xe atoms to higher Rydberg levels [13,33].

While all the points shown in Fig.21 correspond to near optimum timing between the dye pulse injection and e-beam firing, only the 482.5 nm point was obtained using optimum cavity magnification (refer to Fig.19). Additionally, the measured dye laser throughput for a bare cavity was occasionally less than expected on the basis of the cavity magnification, a condition found to be caused by significant mirror surface deterioration under certain conditions. Therefore, with the exception of the 482.5 nm value, the measured amplified output energies of Fig.21 represent lower limits. Correcting the values of Fig.21 for differences in cavity magnification (Fig.19) and for the estimated effect of mirror surface deterioration suggests that, except for wavelengths coinciding with discrete absorption transitions, output energy density of more than 1 J/l should be attainable throughout the entire blue-green region of the spectrum.

Although relatively efficient (~0.3 percent) tuning has been demonstrated for wavelengths as low as 459.4 nm and as high as 505 nm, XeF(C-A) amplifier performance has been limited in the present investigation by the combination of a short active length (~10 cm), low values of cavity magnification (<1.3) required to compensate for the former,
and a critical sensitivity to timing. However, these factors do not represent fundamental limitations. Considering that the gain of the e-beam excited XeF(C→A) medium is relatively high (>2 percent cm⁻¹) over a 100 nm bandwidth centered at 480 nm, an improvement in performance should be forthcoming as a result of pumping geometries which are better suited to optimization of the laser cavity and more reliable timing techniques.

3.3. INJECTION CONTROL OF Xe₂Cl LASER

The spectroscopic and kinetic characteristics of electron-beam pumped triatomic excimers, Rg₂X, have been investigated because they are potentially capable of wide wavelength tunability due to a steeply repulsive potential curve in the ground state, and as a result the excited state fluorescence output has a bandwidth of several 10’s of nanometers [1] similar to the diatomic XeF(C→A) excimer system.

From the point of view of laser development, the broadband trimer emission is accompanied by long radiative lifetimes implying low optical gain for these excimers. Due to this limitation, only two triatomic excimers, Xe₂Cl centered at 500 nm and Kr₂F centered at 435 nm, have been demonstrated so far [34,35,36]. Electron-beam pump-induced transient atomic and molecular absorbers were found to severely limit the laser output power and efficiency of the blue-green triatomic excimers. Due to their low effective optical gain, tuning of these lasers using intracavity elements was unsuccessful [37].
In an effort to overcome some of the limitations observed for Xe$_2$Cl laser, a series of experiments has been performed to demonstrate tunability of this laser by means of injection control. Especially, the rapid build-up of the Xe$_2$Cl optical field by injection of a dye laser pulse will aid the competition against radiative emission from the high-gain diatomic transitions and non-radiative quenching of the Xe$_2$Cl molecule by halogen donors and other constituents of the laser gas mixture.

I. EXPERIMENTS

The experimental setup is the same as in the last section as well as the optical cavity and mirror coatings which have a maximum reflectivity range extended to 520 nm. CCl$_4$ has been found to be the optimum halogen donor for e-beam excited Xe$_2$Cl formation in terms of fluorescence yield and quenching behavior [1]. However, the high trimer quenching rate for CCl$_4$ limits the donor partial pressure to ~1.5 torr for optimum Xe$_2$Cl performance [38]. The optimized Xe$_2$Cl laser mixture contains 1.5 torr CCl$_4$, 300 torr Xe, 6.5 atm Ar, and 200 torr N$_2$. The presence of N$_2$ at this level will reduce both discrete and broadband absorption resulting from Xe$^*$ and Xe$_2^*$ as well as charged particles [39]. Good mixing of the different gases was achieved by turbulent flow of the high pressure gas components into the reaction cell. Each gas mixture could be used for up to 10 shots before a refill was needed.

II. RESULTS AND DISCUSSION
In this experiment, an injection controlled Xe$_2$Cl laser was tuned from 490 nm to 520 nm for the first time with high output and high spectral brightness (a term used to describe the laser power per unit wavelength within certain solid angle). Fig.22 shows the Xe$_2$Cl laser output power density as function of wavelength with the Xe$_2$Cl fluorescence spectrum for comparison. A peak power density of 160 KW/cm$^2$ was observed which corresponds to a more than three orders of magnitude increase in spectral brightness over that achieved previously for Xe$_2$Cl excimer laser [1]. From our experience with the homogeneously broadened XeF(C$^+$A) laser [17] as well as in Ref.40, the bandwidth of the Xe$_2$Cl laser which is also homogeneously broadened is expected to follow the injected dye laser, that is, $\sim$ 0.005 nm. The cavity magnification used is 1.08, corresponding to an output coupling of about 14 percent [28]. This output coupling is much larger than the optimized output coupling for the stable cavity of 2 % [1]. Further tuning outside 490-520 nm region was limited by either a smaller gain on the short wavelength side or the mirror coating reflectivity beyond 520 nm.

The injection controlled Xe$_2$Cl laser power density strongly depends on the delay of dye laser injection relative to e-beam pumping as is the case with XeF(C$^+$A) injection. The optimum time for the dye laser injection was found to be a few nanosecond after the e-beam pumping pulse. If dye laser is injected earlier, the initial absorption produced by e-beam excited species [1] would severely reduce the intensity of the injected laser pulse.
Figure 22. Xe₂Cl laser power density as a function of wavelength
Fig. 23 shows the amplifier gain of the injected dye laser throughput as a function of the throughput. This amplifier gain is a logarithmic function of the dye laser throughput energy. An amplification factor of up to 1/4 was demonstrated. Improvement of injection controlled Xe₂Cl performance by higher dye laser energy injection is possible because it may partially saturate the excimer absorption species in the cavity [41]. Even better results may be realized by increasing the pumping length and therefore larger M values.

A similar procedure may also be used for demonstrating injection control of Xe₂Cl laser in a discharge pumped excimer medium if higher pressure self-sustained stable discharge devices obtained with X-ray, UV or e-beam preionization are available [41]. This laser also has potential application in areas such as ultrashort pulse laser amplification [18, 19].

3.4. LASER EFFICIENCY

I. ENERGY DEPOSITION

Determination of the electrical-optical energy conversion efficiency corresponding to the measured laser pulse energy requires a knowledge of the e-beam energy deposition in the gas. Several threedimensional computer models [42, 43] for e-beam energy deposition and measurements [42] of gas pressure rise, all show that the energy deposited under conditions similar to those of this investigation is larger than that computed on the basis of simple stopping power calculations. The difference is due largely to backscattering from the cell walls and
Figure 23. The amplifier gain of the injected dye laser throughput as a function of the throughput
to multiple scattering in the foil separating the low and high pressure regions of the cell. To account for such effects, a factor of 2 correction to a stopping power calculation based on the Berger and Seltzer data [44] was used in order to obtain a reasonable measurement of the e-beam energy deposited in the active volume. Measured current density levels on the optical axis were found to vary from 200 to 300 Acm\(^{-2}\), depending on cathode-anode (stainless steel mesh) distance, e-beam control parameters, and unpredictable shot-to-shot variations. This range of current density, combined with Ar pressures in the 6-8 atm range, corresponds to energy deposition levels ranging from approximately 67 to 133 J/l. For typical condition of current density 250 Acm\(^{-2}\) and Ar pressure of 6.5 atm, the energy deposition is calculated to be about 90 J/l.

II. MEASUREMENT

In order to measure the energy deposition density into the gas mixture, a calcimeter plate was inserted into the cell through the port for Faraday cup as shown in Fig.4 with an aluminium plate on the optical axis as an e-beam energy absorber. Under low pressure gas condition, the total number of electrons is (1 ev = 1.6\times10^{-19} Joul):

\[ Ne^- = \frac{E(J)}{Ee(ev)\times1.6\times10^{-19}} \]

therefore, \( Q = \frac{E(J)\times Qe^-}{Ee(ev)\times1.6\times10^{-19}} - Ix(FWHM) = J_{eb} \times AX(FWHM) \)

\[ J_{eb} = \frac{E(J)}{Ee(ev)\times AX(FWHM)} \]

where \( E(J) \) is the measured energy in Joules; \( Ee(ev) \) is the energy of
average electron in unit of ev; The total charge in electrons is \( Q \), while the charge in one electron is \( Qe^- = 1.6 \times 10^{-19} \) coulomb; \( J_{eb} \) is the current density; \( A \) is the area of calorimeter's aluminium plate; FWHM is the e-beam current duration (\( \sim 10 \) ns). We also have:

\[
E(\text{Cal}) = C M \Delta T = C M \Delta R K \quad \text{or} \quad E(J) = C M \Delta R K J_E
\]

\[
J_{eb} = \frac{C M \Delta R K J_E}{E(ev) \times A \times (FWHM)}
\]

where \( E(\text{Cal}) \) is the measured energy in calories; \( C \) is the specific heat; \( M \) is the mass of Al plate; \( \Delta T \) is the temperature change of Al plate; \( \Delta R \) is the change of the thermal resistor on back of the Al plate \( (\Delta R = 1.31 \times 10^2 V_{out} \) from bridge circuit with \( V_{out} \) being voltage read out); \( K \) is the thermal constant of the resistor; \( J_E \) is the conversion factor from calories to Joules. We have:

\[
C = 0.215 \text{ cal/g}^\circ\text{C};
\]

\[
J_E = 4.1855 \text{ J/cal};
\]

\[
M = 7.18 \text{ g};
\]

\[
E(ev) = 10^6 \text{ ev};
\]

\[
A = 7.3 \text{ cm}^2;
\]

\[
FWHM = 10 \text{ ns} = 10^{-8} \text{ sec};
\]

\[
K = 2.6^\circ\text{C/}\Omega
\]

\[
J_{eb} = 3.02 \times 10^4 V_{out} \text{ if } E(ev) = 1 \text{ Mev.}
\]

The energy deposition on the unit area of Al plate is:

\[
\frac{E(J)}{A} = 302 V_{out}
\]

The measurement result is shown in Fig.24. The distance from calorimeter plate to the foil was 2.5 cm. The e-beam energy deposited in
Figure 24. The e-beam energy received by calorimeter as a function of Ar pressure
8 atm of Ar is about 0.183 J/cm². Therefore, we obtain an energy deposition of 9.15 J/liter atm. In 6.5 atm of Ar, the total energy deposition was 59.5 J/liter. If we consider 10% reflection of e-beam by the Al plate, we have ~ 65.4 J/liter deposited energy. Therefore, if we consider additional contribution to the deposition due to backscattering from the cell wall, the calculated e-beam deposition of 90 J/l appears to be a reasonable number.

III. EFFICIENCY

For the optimum laser output achieved by injection control with a gas mixture of 8 torr NF₃, 1 torrF₂, 8 torr Xe, 300 torr Kr, and 6.5 atm Ar and an e-beam current density of Jₑb = 250 A/cm², the measured energy density is about 4.7 J/l using a Gentec energy meter calibrated photodiode which was also checked by a Scientec energy meter. This corresponds to an intrinsic efficiency of ~5% (4.7/90).

With 1 mJ dye laser injection pulse (FWHM = 10 ns) into a 1.5 mm coupling hole, the initial intracavity flux reaches the level of saturation intensity of XeF(C-A) medium. After multipass amplification, the intracavity flux approaches a level of more than 3 times the 6 MW cm⁻² saturation flux. For the condition of interest, we have about 9 J/l flowing through the XeF(C) state [8]. The measured output of 4.7 J/l corresponding to an extraction efficiency of ~50 %.
CHAPTER 4

MODEL INVESTIGATIONS

In the past decade a number of investigations have been made using numerical simulation models to predict the performance of injection controlled pulsed [45] and CW dye lasers [46], diode lasers [47], and excimer lasers [48,49]. In the case of excimer lasers, we must consider the presence of many kinetic and other reactions. An analysis in Reference 48 suggests that injection locking offers no advantage for shortening laser build-up time or improving the optical quality in KrF(B→X) system. However, for a low gain medium such as XeF(C→A), this is not the case as shown by experiment [17]. Therefore, in order to gain a better understanding of the injection control process in XeF(C→A) laser, a semiempirical model has been formulated using a pulsed regenerative amplifier approach. A set of coupled rate equations were used for this purpose as shown in section 4.1. The results of numerical integration of these equations are described in section 4.2. Some key factors in cavity and pumping source design will be addressed.

4.1. ANALYTICAL MODEL

For energy extraction calculations, it is adequate to use a simple set of coupled rate equations. A rate equation approximation of the photon flux, gain, and absorption neglects all wave effects, in particular diffraction. Thus, this semiempirical model of an injection con-
trolled laser describes the performance parameters based on experimentally determined properties of the gain medium, using a minimum of free parameters.

The set of coupled rate equations are as follows:

$$\frac{d q(x,t)}{dx} = G(t)q(x,t) - L(t)q(x,t) - \frac{\ln M}{d} q(x,t) + \frac{N_C(t)}{\tau_{CA}} \omega \int g(\nu) d\nu$$  \hspace{1cm} (1)$$

$$\frac{d G(t)}{dt} = P_C(t)\sigma_{SE} - \frac{G(t)}{\tau_C} - G(t)\sigma_{SE} q(x,t)$$  \hspace{1cm} (2)$$

$$\frac{d L(t)}{dt} = P_a(t)\sigma_a - \frac{L(t)}{\tau_a} - L(t)\sigma_a q(x,t)$$  \hspace{1cm} (3)$$

where \(q(x,t)\) is photon flux; \(\frac{dq}{dx}\) is the amplification of photon flux \((q)\) in propagating through each gain sheet of thickness \(dx\) along the optical axis; \(G(t)\) and \(L(t)\) are the measured gain and loss per unit length at 488 nm with optimized gas conditions; \(d\) is the cavity length; \(M\) stands for the cavity magnification; \(N_C(t)\) is the population density of XeF(C) states; \(\tau_{CA}, \tau_C, \tau_a\) are the radiative decay time of the C-A transition, effective decay times of the C states and absorbers, respectively; \(\omega\) is the solid angle into which the spontaneous radiation has to be emitted in order to contribute to the build-up of a free running laser signal; \(g(\nu)\) is the line shape function; \(P_C(t), P_a(t)\) denote the production rates for C state and absorbers derived from the measured small signal gain and loss; \(\sigma_{SE}, \sigma_a\) represent the stimulated emission and absorption cross-section. We have \(G(t) = N_C(t) \times \sigma_{SE}.\)
On the right side of Equation (1) the first and second terms describe the gain and loss, respectively, due to the active medium. The third term \(-\frac{1}{d} q\) is the loss of photon flux per unit length due to the expansion of the beam in the unstable resonator. The fourth term represents the spontaneous emission contribution where \(\int g(\nu) d\nu\) was integrated within the bandwidth of injected dye laser. We normally consider \(g(\nu)\) as a square function with a bandwidth corresponding to wavelength range 460nm-510nm. On the right side of both Equations (2) and (3), the first terms are the production terms. The second terms are the decay terms, due to radiation and nonradiative collision processes, and the third terms describe the losses due to stimulated transitions induced by the radiation field and the bleaching of absorbers by this field, both lead to saturation.

The injected dye laser flux was coupled into the rate equations by a boundary condition \(q(0,t)\) at the injection hole position \((x = 0)\). The deposition of e-beam energy into the gas mixture was assumed to be uniform throughout the cavity. In a positive branch confocal unstable cavity with a convex mirror used as an output coupler (as shown in Fig.25), the regenerative laser beams in the direction towards the output coupler are parallel with the optical axis in a geometrical approximation, while the reflected beams from the output coupler are divergent causing some loss in photon flux. If the loss of photon flux due to the expansion of the regenerative beam size was distributed along the beam path in a complete round trip, the third term on right side of Equation (1) is then obtained. Furthermore, the unstable cavity was treated like
Figure 25. The positive branch confocal unstable cavity and the e-beam pumping region
a folded amplifier. The influence of the unpumped region (\( \sim 2.5 \text{ cm} \)) is neglected compared to the pumped region (\( \sim 10 \text{ cm} \)).

In order to solve the rate equations, an analytical expression for the small signal gain and loss must be obtained. Then, the generally unknown production rates \( P_C(t) \) and \( P_A(t) \) can be derived from Equations (2) and (3) with the small signal approximation (q = 0) by fitting the solutions of (2) and (3) to experimentally determined gain and absorption signals shown in Fig.12. In most cases, the parameters were tailored to represent the measured data as closely as possible and to simulate precisely the temporal variation of both gain and absorption. Since we have very well measured temporal gain and output data at 488 nm (Ar\(^+\) laser line), most of our analysis were done near that wavelength. However, the techniques used are applicable to other wavelengths. The computer code developed for numerical integration of the coupled rate equations was tuned in this fashion by choosing the unknown or uncertain parameter values to provide agreement with former experiments. All subsequent runs were made without any further changes in these parameters. For example, \( \sigma_a \) was set to be \( 1 \times 10^{-17} \text{ cm}^2 \), while \( \tau_C \) and \( \tau_A \) were derived from earlier experiments and had values of \( \sim 10 \text{ ns} \) and \( \sim 8.3 \text{ ns} \), respectively.

The total length of the folded amplifier along the optical axis is determined by the injection hole and output coating size as well as cavity length and magnification. The duration of the signals depends on the initial injected dye laser duration and on how far into the tail of the laser pulse one wishes to investigate. The time step \( \Delta t \) is chosen
to fit up to 600 points under the initial pulse, while the space step \( \Delta x \) is chosen to limit the relative variation of the photon flux at each step. The actual integration of the equations is performed by a simple predictor-corrector procedure. The temporal advancement of gain and photon flux calculations is done sequentially. The small signal gain in the cavity is changed over a period \( \Delta t \), while keeping the photon flux constant. The gain is then held constant, while the photons are propagated. A "C" language program was used which was similar to the Fortran IV program used in Ref.50.

4.2. RESULTS AND DISCUSSION

The experimental (a) and analytical (b) results of temporal relationships of the dye laser, the e-beam excitation pulse, the amplified XeF(C+2A) output, and the broadband XeF(C+)A output with the system operating as a free running oscillator are shown in Fig.14. Generally good agreement exists between the calculated results and the experimental data. The calculated duration of injection controlled output is shorter than the measured output pulse due to the saturation of the gain medium in the center region passed by the laser beam which has been neglected. The regenerative beam actually only sees a doughnut shaped gain medium for any spatial sheet \( \Delta x \) due to the saturation of the center part passed by previous beams. Therefore, another space dimension transverse to the optical axis should be introduced.

Another observation is the injection gain and saturation behavior as shown in Fig.26. For cavity magnification \( M = 1.23 \), the injection controlled output was saturated for an injection energy of 1 mJ. The
same behavior is predicted by the analytical model (solid line). The slope of the predicted curve, which corresponds to the gain, is in good agreement with that of the experimental results (squares)[27]. Since the laser normally works near saturated regions, the change of solid angle \( \Omega \) from \( 10^{-3} \) to \( 10^{-5} \) in rate equation (1) will not change the output by much.

With a CW Ar\(^+\) laser injection, it was found in the experiment that the ratio of the amplified peak temporal integrated spectra of Ar\(^+\) laser to that of the XeF(C\(\rightarrow\)A) spectra (see Fig.27) was a linear function of the Ar\(^+\) laser power as shown in Fig.28. This relationship is not easily seen from the nonlinear coupled rate equations. The analytical results show a very good fit because, with very low input optical flux, the output flux should be a linear function of the input until the input flux is close to the medium saturation level (refer to Fig.26).

Another observation is that the e-beam pumped multipass amplifier output was found to have essentially the same spectral width as that of the injected dye laser pulse [17]. The linewidth of the injected dye laser radiation was either 0.005 nm (grating tuned) or 0.001 nm (grating tuned plus intracavity etalon of 1 cm\(^{-1}\) FSR). In the transient regime, the injected signal provides the laser medium with an initial field so that laser oscillation within the injected bandwidth does not have to build up from spontaneous emission. Consequently, the emission in this bandwidth increases in amplitude faster than the other wavelengths. Therefore, the spectral width of the amplifier should be essentially the same as that of the injected signal. This applies only when both pump
Figure 26. XeF(C+A) output pulse energy dependence on the injected dye laser energy. The squares are the experimental results for cavity magnification of $M = 1.23$
Figure 27. The amplified spectra of Ar$^+$ laser with XeF(C→A) spectra used for Fig.28
Figure 28. The ratio of peak amplified \( \text{Ar}^+ \) line to XeF(C-A) laser spectra as a function of \( \text{Ar}^+ \) laser power. The solid line is the analytical output.
pulse and injection pulse are very short like the conditions typical for this experiment [16,17,45].

The analytical model can be used in the design of cavity optics for future experiments. A calculation of a proposed e-beam geometry/cavity design has been made. The requirements and assumptions are as follows:

1. Pulse energy desired >0.5J.

2. Desired magnification $M \geq 2$. The various considerations include beam quality, injection efficiency (rate of injected energy coupled into the cavity to the total injected energy), and alignment sensitivity require a larger $M$ value [16,28].

3. Extractable energy density = 3J/liter as compared with best reported results of 4.7J/liter for peak of the gain spectrum [17].

4. The analytical gain is the same as previously measured, which means that same energy deposition per volume and other conditions are required.

5. No significant nonsaturable absorbers.

6. Saturation intensity is around 6 MW/cm$^2$.

7. Outer-diameter of output is set to be 2 cm, which means that the output coating spot size is $D \leq 1$cm.

8. The dye laser input power is 1MW (into an injection hole with a diameter of 1.5 mm) with duration of ~ 250 ns typical of flash lamp pumped dye laser. A long pulsed coaxial flash lamp pumped dye laser will eliminate the jitter problem of synchronizing an e-beam
generator to an excimer pumped dye laser.

On careful consideration of the listed requirements and assumptions, calculation shows that longer gain-medium lengths (> 50 cm) are generally desirable. But there is a trade-off between performance and the cost of a long cathode e-beam generator. Another factor is that with longer gain-medium lengths, ASE from the XeF(B→X) transition might become a serious problem for competing with the XeF(C→A) laser.

If we assume that the production terms on the right side of both Equations (2) and (3) will change linearly in the region around the present condition with respect to the e-beam current density, we can reduce or increase the production rate with respect to a longer or shorter cathode, therefore optimizing the cathode length for certain e-beam machine whose total current output and total pumping power are relatively constant. In case of the Pulserad 110 with a total current of ~ 24 KA, the optimized cathode length was found to be ~ 6" instead of 3", as shown in Fig.29. This corresponds to a pumped region of ~ 20 cm. The optimized cavity magnification was found to be M = 1.15, as shown in Fig.30.

In summary, an analytical model of the injection process using a regenerative amplifier approximation was developed. Such a model consisted of a set of coupled rate equations. They can be used to describe the injection controlled laser performance, as well as to assist in the optimization of the confocal unstable resonator design, such as the cavity magnification for certain cavity lengths and temporal gain profiles.
Figure 29. Dependence of the laser output on pumping length from the analytical model with a total e-beam current of 24 KA. The pumping density is normalized to 10 cm
Figure 30. Dependence of the laser output on cavity magnification from the analytical model with pumping length of 20 cm.
The simple photon flux equation presented in section 4.1 is adequate for energy extraction calculations, while calculations for investigating the optical quality of an injection controlled laser require far more elaborate techniques to determine the intensity and phase distributions transverse to the optical axis. Such calculations are performed routinely for CW infrared lasers without injection control, where the laser kinetics and optical mode evolve through interactive computations.
CHAPTER 5

FUTURE DIRECTIONS AND CONCLUSIONS

5.1. FUTURE DIRECTIONS

The main objective of the future research is to advance significantly state-of-the-art XeF excimer laser technology which should be based on our past experience and accomplishments in the area of broadband diatomic and triatomic excimer lasers. The specific goals and techniques to be used in advancing the development of tunable excimer lasers are summarized in the following subsections.

I. New Cell Design

One prediction made in chapter 4 is that longer gain-medium length and larger gain volume are generally desirable in the development of the XeF(C\to A) laser as an useful tunable laser source. To date, all our experiments have been performed using an active volume of less than 30 cm$^3$. In order to obtain useful energy from a XeF(C\to A) laser with a specific output energy of larger than 1 J/l, we need to evaluate relevant scaling parameters. However, this task is not immediately straightforward and will require the solution to several problems. Among these are competition from the XeF(B\to X) transition which has a very high gain and the round trips available within the gain duration for the laser medium to reach saturation.
As shown in Fig.1, XeF has two excited states, namely B and C. At room temperature, more than 95% of the excited state population in the XeF (B, C) manifold is in the C state under equilibrium condition with high pressure Ar buffer gas. However, the gain on the B→X transition is much higher than that on the C→A, mainly due to the fact that the spectral width of the B→X transition is much smaller (~1 nm) than it is for the C→A transition (100 nm); also the radiative lifetime of the B state (10 ns) is considerably shorter than that of the C state (100 ns). At the same time, high buffer gas pressures (6.5 atm) are necessary for effective energy transfer from the electron beam to the laser medium in order to ensure rapid mixing between these two states, with a relaxation time of well less than a nanosecond. Thus, in a region of excited XeF, amplified spontaneous emission of the B→X will develop much more rapidly than that of the C→A due to the high gain (~10%/cm for the B→X as compared to ~2%/cm for the C→A transition). However, it was demonstrated that the addition of krypton to the excimer laser gas mixture had the effect of active suppressing the oscillation of the XeF(B→X) laser transition by the absorption of Kr related species such as Kr₂F and Kr²⁺ near 351 nm [29]. Therefore, it appears that an extended length XeF(C→A) laser, with good suppression of spontaneous, superfluorescent XeF(B→X) competition should be possible.

Future experiments will be performed in a new reaction cell which is more than 50% larger than the present one as shown in Fig.31. By utilizing a series of different e-beam cathodes and optical cavity lengths, the effect of scaling the C→A laser to larger dimensions should
Figure 31. Schematic of the scaled up laser cell
be investigated. The other possibilities include a folded-cavity geometry to effectively double the gain length in the laser cell, which requires specially designed cavity optics.

II. SUPPORT SYSTEMS

The new cell will incorporate several features not found in our present experimental set-up. The cell and manifold will be pumped by a turbo-molecular pump, which will eliminate oil contamination of the system. A cryogenic gas purifier system and provisions for high pressure gas circulation will enable us to achieve much higher gas purity, longer gas life time, and better gas mixing conditions than has been possible up to now. Improved optical and electrical diagnostic features will provide more accurate characterization of the actual laser cell excitation conditions.

One of the consequences of making the optical cavity (and pumping source) longer will be that the pumping density will decrease. However, we recently found that an electron-beam back-scattering mirror constructed from a high Z material (like nickel coated lead), employed in the cell can increase the pumping density considerably [20]. This device employed in the new cell will at least partially compensate for the decreased current density due to the larger pumping region. The cell will be designed so that careful monitoring of the current density as well as pumping energy can be obtained. It may be that with a different pumping density, but increased cavity length, different gas kinetic conditions require a reoptimization of laser gas mixture. In particular, for a better system compatibility (with discharge pumping system where a
stable discharge is required) and reliable operation, it may be desirable to eliminate the use of NF\textsubscript{3} from the multicomponent rare gas halide mixture. This again will require reoptimization of the individual gas component partial pressure [14,29]. Furthermore, it will be necessary to determine the spatial gain profile for the new system due to the change in pump density distribution.

III. LASER CAVITY AND OPTICS

As discussed above, the laser cavity will be increased in size from the present 12.5 cm length to a maximum of 50 cm (in case of external cavity) with a effective diameter of ~ 3.5 cm. A schematic of the laser cell is shown in Fig.34 with convenient input/output gas flow, side fluorescence monitoring window (with can also be used to insert calorimeter), and four current density probe adapters for measuring the longitudinal current distribution.

In our last experiment, the moderate gain available in the C-A transition and short gain length in the reaction cell necessitated the use of unstable resonators of undesirable low magnification (M = 1.05 to 1.23) [17]. Several advantages can be obtained from configurations with high gain length product, allowing the use of the larger magnifications which are more appropriate to unstable resonators. These benefits include the following: (i) more efficient use of the available injected power, (ii) a better (larger) ratio in output-to-injected power, and (iii) considerable improvement of the output beam quality, in particular, lower beam divergence, therefore better far field intensity distribution, and (iv) greater ease in optical alignment.
In contrast to the previous small cell experiment [17], the new larger cell will allow for the use of external mirrors. The laser cell can have on both ends glass or quartz windows with broadband antireflection coatings for XeF(C→A) wavelength. The windows will be slightly titled with respect to the optical axis in order to avoid feedback of the competing XeF(B→X) transition to the amplifier. Furthermore, an external cavity design will considerably relax requirements for F₂ halogen compatibility of the resonator reflection coatings and improve upon the alignment stability achieved in the intracell geometry used before [17].

5.2. CONCLUSIONS

Efficient, ultraslow spectral output from an electron-beam excited XeF(C→A) laser medium has been observed by injection controlled tuning. Amplification of injected tunable dye laser pulse was achieved throughout the entire blue-green spectral region from 435 nm to 535 nm. These experiments were performed by exciting optimized Ar/Kr/Xe/F₂/NF₃ gas mixture with an intense electron beam. Several different confocal unstable resonator geometries were investigated. A maximum XeF(C→A) laser output of ~ 85mJ was measured at 482.5 nm for a cavity with magnification of about 1.1, which corresponds to an energy density and intrinsic efficiency of about 4.7 J/liter and 5 %, respectively. If comparable performance levels can be achieved using discharge excitation, the XeF(C→A) laser may become a competitive, tunable optical source for the blue/green region of the spectrum.
A spectral linewidth of 0.001 nm was realized which was essentially the same as injected dye laser linewidth. With a ultra-narrow injected laser pulse, the injection controlled output with a near transform-limited-bandwidth \( (4 \times 10^{-5} \text{ nm}) \) should be possible in a laser tuning range of 435 nm to 535 nm. The output energy density and intrinsic efficiency is limited by the energy deposition density and the production efficiency of XeF(C) population. Considering the energy density and efficiency demonstrated in this experiment, significant improvement in XeF(C+A) output performance should be forthcoming as a result of better pumping geometries, a longer active gain and cavity length, and therefore larger cavity magnification.
BIBLIOGRAPHY


33. H. Horiguchi, R.S.F. Chang, and D.W. Setser, "Radiative and two-body collisional deactivation rate constants in Ar for Xe(5p6P), Xe(5p6P) and Xe(5p7P) states," *J. Chem. Phys.*, vol. 75, pp. 1207-
1218, 1981.


1964).


APPENDIX A

Synthesis of Rare Gas-Halide Mixture
Resulting in Efficient XeF(C≡A) Laser Oscillation

by

W.L. Nighan, F.K. Tittel, W.L. Wilson,
N. Nishida, Y. Zhu, and R. Sauerbrey

Synthesis of rare gas-halide mixtures resulting in efficient XeF(C→A) laser oscillation

W. L. Nighan
United Technologies Research Center, East Hartford, Connecticut 06108

F. K. Tittel, W. L. Wilson, Jr., N. Nishida, Y. Zhu, and R. Sauerbrey
Electrical Engineering Department and Rice Quantum Institute, Rice University, Houston, Texas 77251

(Received 9 July 1984; accepted for publication 8 August 1984)

Significantly improved XeF(C→A) laser performance has been achieved using electron beam excitation of complexes, multicomponent gas mixtures specifically tailored so as to reduce medium transient absorption in the blue-green region. Use of Ar and Krypton together as the effective rare gas buffer-energy transfer species, along with a combination of NF3 and F2 to produce the desired F-donor molecule characteristics, has permitted synthesis of near optimum medium properties for which XeF(C) is produced efficiently while transient absorption is minimized. With this technique we have achieved laser pulse energy density and intrinsic efficiency of 2.2 ± 0.3 J/l and ~1.5%, respectively, values that are comparable to those of the B→X' rare gas-halide lasers.

Short pulse electron beam (e-beam) excitation of the blue-green XeF(C→A) laser transition is characterized by a period of strong transient absorption during the excitation pulse, followed by the development of net gain and subsequent laser-oscillation in the afterglow regime. Until recently, the combination of a relatively short duration of the net gain region (<50 ns) and a relatively low value of the peak gain (0.08 cm⁻¹) has limited the efficiency of this tunable laser to unacceptably low levels. However, by selectively tailoring kinetic processes through the use of a unique two-halogen mixture containing both NF3 and F2 so as to reduce transient absorption, a very significant improvement in XeF(C→A) laser performance has been achieved.1,2 Indeed, e-beam excitation of such two-halogen Ar-Xe mixtures has yielded laser pulse energy density in excess of 1.0 J/l, corresponding to intrinsic electrical-optical energy conversion efficiency estimated to be in the 0.5%-1.0% range. In this letter we report further improvement in XeF(C→A) laser performance resulting from the addition of Kr to an Ar-Xe-NF3-F2 mixture. This multicomponent mixture has permitted synthesis of near optimum medium properties resulting in XeF(C→A) pulse energy density and intrinsic efficiency of 2.2 ± 0.3 J/l and ~1.5%, respectively. This improvement was obtained without increasing the energy deposited in the gas by the e-beam. Thus, a level of XeF(C→A) laser performance has been achieved which, for the first time, is comparable to that typical of UV B→X' rare gas-halide laser transitions.

In this investigation laser excitation was provided by an electron beam having an energy of 1 MeV and a pulse duration of 10 ns (full width at half-maximum). The e-beam current density at the center of the optical axis was ~250-300 A/cm², as measured with a Faraday probe. A stable, intra-cell optical resonator was used consisting of a totally reflecting (R > 99.6%) mirror having a radius of curvature of either 0.5 or 1.0 m, separated by 12.5 cm from a flat output mirror having a reflectivity of 95%, a value found to be optimum for the present conditions. The active region was the ~28 cm³ volume defined by the clear aperture (1.9 cm diameter) and the pumped length (10 cm). Specific details of this experimental arrangement and related diagnostic apparatus are described in Ref. 1.

A cw Ar-ion laser was used¹ to measure the temporal evolution of the gain/absorption at several wavelengths throughout the blue-green region. Figure 1 shows a representative gain-absorption profile at 483 nm for an optimized Ar-Xe-NF3-F2 mixture under conditions for which the laser pulse energy density and intrinsic efficiency were typically 1.5 ± 0.3 J/l and ~1.0%, respectively. Comprehensive analysis¹ of medium kinetic processes indicates that for these conditions the initial period of strong absorption is primarily the result of photoionization of the 4s, 3d, and higher lying states of Ar, and of the Xe 6p, and 5d states, along with photodissociation of Ar3⁺(2s⁰) and Ar5⁺. These broadband absorption processes more than offset the positive contribution of XeF(C') excimer molecules during and immediately following the period of e-beam excitation. Although the Ar-related absorption processes decay rapidly, photoionization of Xe excited states is very significant even after the gain

![Graph](image-url)

FIG. 1. Temporal evolution of the net gain measured at 483 nm for mixtures comprised of 6.5 atm Ar, 8 Torr NF3, and partial pressures of Xe and F2 optimized with and without Kr as indicated. The measured e-beam current density on the optical axis for these conditions was typically 275 ± 25 A/cm², corresponding to a volumetric energy deposition estimated to be ~150 J/l (Ref. 1).
becomes positive, thereby substantially reducing the peak gain value from that possible due to XeF(C) alone.\textsuperscript{1}

During the course of this work it was found that addition of Kr dramatically reduced the initial absorption of the previously optimized Ar-Xe-NF\textsubscript{3}-F\textsubscript{2} mixture.\textsuperscript{8} Based on our analysis of the change measured absorption in pure Ar and in Ar-Xe mixtures, it appeared that the most likely explanation for this observation was that Kr reduced the concentration of both Ar\textsuperscript{4} 1\textsuperscript{2}S\textsubscript{1} and Ar\textsuperscript{1} during the e-beam excitation pulse, while providing additional decay channels for the Xe\textsuperscript{6p} and 5d states. Both gain and fluorescence data indicated that the additional quenching of XeF(C) by Kr was also significant, however, so that the magnitude and duration of the gain typical of our initial experimentation with Kr were not improved. Thus, in these early studies, no increase in laser pulse energy/efficiency was achieved with mixtures containing Kr.\textsuperscript{1}

Because the influence of Kr on the measured blue-green absorption in various mixtures was found to be so pronounced, recent efforts have focused on systematic measurement and evaluation of gain temporal profiles and laser pulse energy, while varying the fractional concentrations of Xe, F\textsubscript{2}, and NF\textsubscript{3} in mixtures of Ar and Kr at a total pressure of 6.5 atm. Although the Kr pressure was varied from a few Torr to several atm, particular emphasis was placed on Kr pressures in the 0.1–1.0-atm range for which the absorption peak during the e-beam excitation pulse was found to be minimized. Presented in Fig. 1 is the measured gain profile for a reoptimized mixture containing 0.2 atm Kr, excited under conditions essentially identical to those of the mixture without Kr, also shown in the figure. The significant reduction in absorption during the excitation pulse with Kr in the mixture is readily apparent, as is the higher value of peak gain and the increase in gain duration to about 30 ns. These features were found to be typical of Kr pressures throughout the entire 0.1–1.0-atm range.

Measurements show that the magnitude of the absorption minimum (Fig. 1) with Kr in the mixture is relatively insensitive to the specific values of Xe or F\textsubscript{2} pressure, but that the peak gain (and its rate of decay) is dependent on the concentrations of these species. Indeed, the optimum concentrations of both Xe and F\textsubscript{2} are found to be significantly lower than their values in the absence of Kr.

Although our measurements indicate that the primary role of Kr is reduction of the concentrations of Ar and Xe related species that absorb in the blue-green region,\textsuperscript{1} the measured rate of rise of the XeF(C–A) fluorescence and its peak value both are significantly higher with Kr in the mixture, particularly for large F\textsubscript{2} concentrations. Analysis shows that these effects are much more pronounced than would be expected on the basis of faster XeF(B/C) state mixing\textsuperscript{9} due to Kr, suggesting that XeF(B/C) formation is enhanced when Kr is present. One possible explanation for this observation is that Xe displacement reactions involving either KrF or KrF\textsubscript{2} are more effective than their Ar counterparts.\textsuperscript{3} However, the enhancement in XeF(C) formation with Kr in the mixture appears to be of less importance than the reduction in transient absorption.

Presented in Fig 2 is the dependence of laser pulse energy density on Kr pressure for the specific Ar-Xe-F\textsubscript{2}-NF\textsubscript{3} mixture found to be optimum for Kr pressures in the 0.1–1.0-atm range.\textsuperscript{3} The four-component mixture optimized in the absence of Kr consistently results in laser pulse energy density values of 1.3 ± 0.3 J/1 for the present conditions, a value not unduly sensitive to either the radius of curvature of the total reflector or of the transmission of the output coupler. Figure 2 shows that as the Kr pressure is increased above ~0.05 atm for the reoptimized mixture, the laser pulse energy increases above the ~1.5 J/1 base level of the reference no-Kr mixture. For Kr pressures in the 0.4–0.8-atm range, a broad maximum in laser energy density is achieved, corresponding to ~50% increase over that of the reference mixture, a result that is consistent with the trend exhibited by the measured gain/absorption profiles. The maximum 2.2 ± 0.3 J/1 laser energy density typical of these conditions corresponds to an intrinsic energy conversion efficiency estimated\textsuperscript{1} to be approximately 1.5%.

Measurements were also carried out for Kr pressures in the 3–4-atm range with no Ar in the mixture, a condition for which the energy deposited by the e-beam would be very nearly equivalent to Ar-buffered mixtures at a total pressure of 6.5 atm.\textsuperscript{3} In these tests, with the Xe and F\textsubscript{2} pressures again reoptimized, the maximum laser energy density was in the 0.5–0.7 J/1 range, a value much lower than that of either the no-Kr reference mixture or the optimized mixture with Kr present.\textsuperscript{4} With the Kr pressure in the 3–4-atm range, addition of varying amounts of either Ne or Ar up to pressures of several atm resulted in laser pulse energy values about the same as those using Kr alone, although the energy deposited in such cases was substantially increased. This is in contrast to the situation for either the Ar-buffered reference mixture or the optimized Ar-Kr mixture, for which it was found that increasing the energy deposition by increasing either the e-beam current density or the Ar pressure resulted in higher laser output. Indeed, for the optimum conditions of Fig. 2, but with the Ar pressure increased from ~6.5 to ~8.5 atm, the laser pulse energy was found to increase from 2.2 J/1 to ~3.0 J/1. Thus, all of our evidence indicates that the use of Ar and

\textbf{FIG. 2.} Laser pulse energy density dependence on Kr pressure as measured using a calibrated vacuum photodiode detector. The mixture was composed of Ar and Kr at a total pressure of 6.5 atm, 8 Torr NF\textsubscript{3}, 10 Torr Xe, and 2 Torr F\textsubscript{2}. The indicated base level refers to the mixture optimized in the absence of Kr; the symbol (O) indicates the measured laser energy with no Ar in the mixture.
Kr together as an effective buffer-energy transfer species results in a medium that, when electrically excited, is characterized by significantly lower concentrations of excited-ionized species that absorb in the blue-green region than is the case using either Ar or Kr alone.

Presented in Fig. 3 are the time integrated laser spectra for optimum mixtures with and without Kr for conditions similar to those in Fig. 1. Although there are some differences, the two spectra are generally similar except for a somewhat deeper absorption valley centered near 480 nm in the Ar-Kr mixture. Also shown in this figure is the measured wavelength dependence of the peak value of the zero-field gain, normalized to its value at 485 nm. The relatively weak wavelength dependence of the gain suggests that efficient tuning of the electrically excited XeF(C→A) laser may be possible throughout a large portion of the 450-516-nm range.

This investigation has shown that a combination of rare gases (Ar + Kr) and fluorine molecules (NF3 + F2) permits synthesis of near optimum XeF(C→A) laser properties for which XeF(C) can be produced efficiently (5%-10%) while transient absorption is minimized. The resulting optical extraction efficiency of 20%-25% is unique for an electrically excited XeF(C→A) laser. Indeed, the values of laser energy density (2-3 J/l) and intrinsic efficiency (1%-2%) typical of the present electron beam excited XeF(C→A) laser medium are comparable to those of other blue-green lasers such as HgBr(B→X) and wavelength shifted XeCl(B→X) or XeF(B→X). If comparable performance levels can be achieved using discharge excitation, the XeF(C→A) laser may become a competitive, tunable optical source for the blue-green region of the spectrum. Additionally, it is likely that mixture synthesis of the type employed in this investigation will find application as a means to improve the performance of other laser systems.

It is a pleasure to acknowledge the contributions to this work of our colleagues: Dr. Y. Nisheton of Techtron, Dr. G. Marowsky of the Max-Planck Institut für Biophysikalische Chemie, and Mr. F. Eimerl of the University of Würzburg. This work was supported in part by the Office of Naval Research, the National Science Foundation, and the Robert A. Welch Foundation.

3. With an Ar pressure of 6.5 atm the addition of Kr up to pressures of ~0.3 atm results in an insignificant increase in e-beam energy deposition. In order to account for the effect of Kr at higher pressures, the Ar partial pressure was reduced so as to maintain a total pressure of 6.5 atm. This approximation to constant energy deposition becomes relatively poor for Kr pressures ~1 atm since the e-beam stopping power of Kr is about twice that of Ar. However, our experiments show that, for Kr pressures above 2 atm, the maximum attachable laser energy (~1.0 J/l) for an optimized mixture is relatively insensitive to either the partial pressures of various gas buffer species, the total pressure, or the energy deposited. See text.

5. The rate coefficients for quenching of KrF by Xe have been measured and is found to be approximately 2.5 times larger than the corresponding ArF reaction. These results will be published elsewhere.

6. D. Campbell, C. H. Fisher, and R. E. Coster, Appl. Phys. Lett. 37, 348 (1980). These investigations found that use of Kr instead of Ar as an XeF(C→A) laser buffer resulted in improved performance, but their experimental conditions were very different than those of the present work.

APPENDIX B

Kinetically Tailored Properties of Electron-Beam Excited XeF(C→A) and XeF(B→X) Laser Media Using an Ar-Kr Buffer Mixture

by

W.L. Nighan, R.A. Sauerbrey, Y. Zhu, F.K. Tittel, and W.L. Wilson, Jr.

Kinetically Tailored Properties of Electron-Beam Excited XeF(C → A) and XeF(B → X) Laser Media Using an Ar–Kr Buffer Mixture


Abstract—Use of a two-component buffer gas comprised of Ar and Kr results in electron-beam excited XeF(C → A) laser pulse energy and intrinsic efficiency values comparable to those of UV rare gas-halide lasers. Herein we report measurements of transient absorption confirming that the primary effect of a buffer comprised of Ar and Kr is a significantly lower level of ionized and excited species that absorb in the blue-green spectral region. Spectral analysis of a variety of mixtures shows that the Ar-Kr buffer also benefits XeF(C → A) laser performance due to an increase in gain in the 400–450 nm region caused by the presence of the KrF excimer. In addition, a large increase in absorption at ~351 nm, also due to KrF, suppresses oscillation on the competitive XeF(B → X) transition and, for certain conditions, makes efficient simultaneous oscillation of the XeF(B → X) and XeF(C → A) laser transitions possible.

1. INTRODUCTION

Electrically excited rare gas-halide laser mixtures typically are comprised of a high pressure (> 1 atm) rare gas background, or buffer, a second rare gas species at much lower pressure from which the rare gas-halide eximer molecule is comprised, and a halogen donor at a partial pressure of a few torr [1, 2]. Selection of a particular buffer species and its pressure is dictated by several requirements: 1) vibrational relaxation of the rare gas halide (RGH) eximer by the buffer must be fast compared to deactivation processes, 2) collisional quenching of the RGH by the buffer must be minimal, 3) transient absorption at the laser wavelength by buffer-related ionized and excited species must be held to an acceptable level, and 4) when electron-beam (e-beam) excitation is used the buffer gas must have a relatively high stopping power, while electric discharge excitation requires a buffer compatible with formation of a stable discharge. Additionally, each specific RGH laser often has one or more unique requirements affecting buffer gas selection, but they are generally of less importance than those listed above. Thus, determination of the rare gas buffer species that is best for a particular situation is clearly a compromise involving several conflicting factors.

Based on several years of experimentation supplemented by analysis, the optimum buffer for the e-beam excited XeF(351 nm) laser has been found to be neon [1, 3], while helium is usually used when discharge excitation is employed [4]. The 308 nm XeCl laser uses either neon or helium as the buffer [4, 5], while the KrF (248 nm) laser usually uses helium when excitation is provided by a discharge [6] and argon when e-beam excitation is employed [6, 7]. Additionally, the best performance from the broad-band e-beam excited XeF(C → A) laser (450–510 nm) has been obtained using argon as the single buffer [8]. Recently, investigations have been carried out focused on evaluation of the merits of using two rare gas components to form the buffer gas [9–12]. Notable success has been realized using an Ar–Kr combination to form the high pressure buffer for an e-beam excited XeF(C → A) laser [11, 12]. This approach has resulted in a dramatic improvement in the net gain of the broad-band XeF(C → A) transition centered at ~480 nm. Indeed, laser pulse energy density and intrinsic efficiency values have been demonstrated that compare very favorably with those of the more highly developed UV RGH B → X lasers, by using the XeF(C → A) medium either as a broad-band oscillator [11], or as a wavelength selectable amplifier [12]. In addition, use of Ar–Kr buffer gas mixtures permits relatively efficient, simultaneous oscillation of the XeF(B → X) and C → A laser transitions [13]. Interpretation of these results led to the conclusion that the primary effect of Ar and Kr in combination was a significantly lower level of transient absorption in the blue-green spectral region [11]. Herein we report on recent work supporting this conclusion, along with evidence of additional factors that benefit XeF(C → A) laser performance, including: 1) faster mixing of the XeF B and C states, 2) a contribution to the net gain in the 400–450 nm region due to the presence of a high concentration of the KrF excimer, and 3) a large increase in absorption at UV wavelengths, also due to KrF, which suppresses oscillation on the competitive XeF(B → X) transition and, for certain
conditions, makes possible relatively efficient, simultaneous oscillation of the XeF(B → X) and XeF(C → A) laser transitions.

The details of the experimental arrangement and related diagnostics used in this investigation are described in Section II. In Section III the effect of Kr as an additive to reduce transient blue-green absorption is treated. Spectroscopic data relevant to the role of Ar–Kr mixtures on mixing of XeF(B, C), and on the influence of Kr on XeF(C → A) laser performance are presented in Section IV, while a discussion of conditions for which simultaneous XeF B → X and C → A laser oscillation can be obtained is presented in Section V.

II. EXPERIMENT

A. Electron-Beam and Reaction Cell

The overall experimental arrangement used in this investigation is illustrated in Fig. 1. A Physics International Pulser and 110 electron-beam generator was used to transversely excite the high pressure gas mixtures [8], [12].

The electron beam energy was 1 MeV and the pump pulse duration was 10 ns (FWHM), producing a pump energy density of ~135 J/l, as measured by a calorimeter and a Faraday cup probe. The excited region was the cylindrical 28 cm² volume defined by the clear aperture (1.9 cm diameter) and the pumped length (10 cm).

The stainless steel reaction cell was well passivated by prolonged exposure to F₂ prior to any experiments. High purity gas mixtures of research grade Ar, Xe, Kr, NF₃, and F₂ were used, with the F₂ in a 10–90 F₂–He mixture. Good mixing of the gas components was achieved by turbulent flow of the high pressure gas components into the reaction cell. A fresh gas mixture was used for each shot, although up to ten shots could be made before any significant deterioration of the mixture was observable.

B. Diagnostics

The temporal evolution of the fluorescence and the laser output were monitored by a fast vacuum photodiode detector (ITT-F4000SS). Neutral density filters were used to avoid saturation of the photodiode, and interference and color glass filters were used to define the spectral regions of interest. Signals were recorded by a Tektronix R 7912 Transient Digitizer. The time resolution of the entire system was better than 2 ns. The temporally-integrated, spectrally-resolved fluorescence was recorded by an optical-multichannel-analyzer (OMA III), using a Jarrell-Ash 0.25 meter spectrometer having a spectral resolution of ~0.3 nm. Data from the Transient Digitizer were processed using a PDP 11/23 minicomputer. The OMA III signals were recorded by an IBM PC-XT computer.

The spectrometer and the OMA III have a wavelength dependent sensitivity. In order to account for this, the spectral sensitivity of the system was determined by three different methods and the measured spectra were appropriately corrected. The first procedure utilized N₂(C → B) fluorescence emission lines [14]; the second method used a Xe flashlamp with a known spectral intensity dis-

![Fig. 1. Schematic illustration of the experimental apparatus and arrangement. NDF = neutral density filter, CF = color glass filter, VPD = vacuum photo diode, and OMA = optical multichannel analyzer.](image)

tribution together with narrow frame interference filters to define the spectral sensitivity measurement regions; and the third utilized an Ar⁺ laser with known power at each transition line. Using these techniques the wavelength response of the OMA III system was quantified for the 330–520 nm spectral region, and all experimental data were corrected to account for the spectral response of the detection system.

C. Gain / Absorption Measurement

A CW Ar-ion laser was used to measure gain at several blue-green wavelengths, concentrating on the ~480 nm region for which the XeF(C → A) gain is a maximum. Three passes of the probe beam through the cell were used in order to maximize the signal-to-noise ratio. The laser probe signal was focused on a Lasermetrics 3117 PIN diode through a narrow-band interference filter, a color-glass filter, and an iris located 10 m from the laser cell. The detector was located inside a Faraday cage to minimize electrical noise pickup and stray fluorescence. A mechanical shutter was used to produce a 4 ms laser probe pulse to avoid saturation of the detector. The electron beam pulse was synchronized to appear in the middle of the laser probe pulse. In the CW mode, the detector diode is linear with input power for output currents up to 4 mA. However, the time response of the detector for currents above 2 mA began to deteriorate. Therefore, the detector current was always maintained below 2 mA, for which the time response was better than 2 ns.

III. ABSORPTION IN THE BLUE–GREEN SPECTRAL REGION

A. Net Gain in XeF(C → A) Laser Mixtures

In order to compensate for a stimulated emission cross section [15] having a peak value of only ~10⁻¹⁵ cm², intense pumping is required to produce adequate gain on the XeF(C → A) transition, a circumstance resulting in very high concentrations of excited and ionized species, many of which absorb at the laser wavelength [8]. Thus,
Figure 2. Temporal evolution of the e-beam excited XeF(C → A) net gain profile measured at 488 nm using an Ar-ion probe laser for mixtures comprised of 6.5 atm Ar, 16 torr Xe, 8 torr NF₃, and 8 torr F₂, (a), and 6.5 atm Ar, 10 torr Xe, 8 torr NF₃, 2 torr F₂ and 150 torr Kr (b).

Figure 3. Temporal evolution of the absorption measured at 488 nm in 6 atm Ar and in a mixture comprised of 6 atm Ar and 16 torr Xe (solid lines). The absorption measured for the same conditions but with 150 torr Kr added to each mixture is also shown (dashed lines).

Electrical excitation of the XeF(C → A) medium is usually characterized by a period of strong transient absorption during the excitation pulse, followed by development of net gain in the afterglow regime [8], [11]. Presented in Fig. 2 is the measured temporal evolution of the net gain for representative e-beam excited mixtures with and without Kr, vividly illustrating the significant reduction in the initial absorption occurring during the excitation pulse, and the increase in peak gain when an optimized Ar-Kr buffer mixture is used [11]. The results of Fig. 2 are typical of the wavelength region between 460–510 nm [12], and are found to be relatively insensitive to Kr pressure for values in the 0.1–1.0 atm range, although optimum gain is achieved for a Kr pressure of ~0.5 atm.

B. The Role of Kr

1) Transient Absorption in Laser Mixtures: Detailed modeling [8] of XeF(C → A) medium properties for the mixture of Fig. 2 that does not contain Kr provided evidence that the primary absorbing species during and after the e-beam excitation pulse are Ar₂⁺, Ar₃⁺, Ar⁺(4p, 3d) and Xe⁺(6p, 5d), with the latter being the dominant absorber and one having a particularly slow rate of decay. With a few tenths of an atmosphere of Kr present in the mixture, species such as Ar₂⁺ (and its precursor Ar₂⁺⁺) and Ar₃⁺ are converted to Kr⁺ and Kr⁺ on a nanosecond time scale, with the latter atomic species apparently replacing Ar-related molecular species as effective XeF(B, C) precursors. That is, we postulate that upon addition of Kr certain molecular XeF(B, C) precursors that exhibit broad-band absorption have been partially replaced by atomic precursors that do not. Moreover, Kr-related molecular species such as Kr₂⁺ or KrF⁺ that might also be expected to contribute to broad-band absorption at blue-green wavelengths are formed at a slow rate compared to corresponding Ar-related species since the Kr pressure is much less than that of Ar. Also, heteronuclear species such as ArKr⁺⁺ or ArK⁺⁺ are produced at a slow rate because their three-body reaction rate coefficients are generally much less than those of similar homonuclear species [16]. For these reasons, in Ar-Kr buffered mixtures it appears that the role of molecular absorbing species as XeF(B, C) precursors is reduced compared to the situation typical of Ar-buffered mixtures, and that the rate of decay of long lived absorbing species such as Xe⁺(6p) is increased.

2) Absorption in Ar and Ar-Xe Mixtures: The above interpretation is supported by measurements of absorption in Ar and Ar-Xe mixtures with and without addition of Kr. Presented in Fig. 3 is the time dependence of the measured transient absorption in such mixtures for conditions otherwise similar to those of Fig. 2. In this illustrative example, for Ar at 6 atm the peak absorption during the e-beam excitation pulse is reduced by about 50 percent with 0.2 atm Kr added, a change interpreted as due to reduced concentrations of Ar-related molecular absorbers as described above. The peak absorption occurring during the excitation pulse in the Ar-Xe mixture is affected only slightly with 0.2 atm Kr added, since in that case Xe plays a role similar to that of Kr as regards reduction in the concentration of Ar-related molecular species. However, Fig. 3 shows that the temporal decay of the absorption in the Ar-Xe mixture is exceptionally slow, extending far into the afterglow region. Modeling [8] has shown that the absorption in that case is due almost entirely to photoionization of Xe⁺(6p) states for which there is no effective loss channel with Ar as the buffer. However, with Kr added to the Ar-Xe mixture the rate of absorption decay in the afterglow is increased significantly, indicative of the presence of more effective Xe⁺(6p) exit channels [8] with Kr in the mixture.

3) Quenching and Absorption in Kr-Containing Mixtures: The fact that an Ar-Kr buffer mixture results in substantially less blue-green absorption than use of Ar alone suggests simple substitution of Kr for Ar rather than use of the two together. In order to explore this possibility, measurements [11] were carried out using Kr as the buffer for XeF(C → A) laser mixtures at pressures such that the e-beam energy deposition was essentially the same as for the mixtures of Fig. 2, i.e., Kr pressures of 3–4 atm. For these tests the partial pressure of each mixture constituent was reoptimized, a very important factor, yet
no conditions were found for which Kr-buffered laser mixtures performed as well as either Ar or Ar-Kr buffered mixtures similar to those of Fig. 2. Moreover, examination of the fluorescence and laser output as Kr pressure is increased above the optimum level for Kr-containing mixtures provides evidence of additional XeF quenching and/or blue-green absorption processes not observed in Ar buffered mixtures. Since the rate coefficients for two-body quenching of XeF(B, C) by either Ar or Kr are small, as is the three-body coefficient for XeF quenching by Ar [17], we feel that the most probable additional quenching process is three-body quenching of XeF(B, C) by Kr with Ar acting as the third body. A rate coefficient value \( \geq 10^{-12} \text{ s}^{-1} \text{ cm}^3 \) for the reaction XeF(B, C) + Kr + Ar \( \rightarrow \) products is consistent with our observations and with those of others as well [18]. Additionally, it is likely that Kr-related broad-band absorbing species are produced at high Kr pressures. Whether the observed effects are due to quenching, absorption, or some combination of the two, use of Kr as the sole buffer at pressures above approximately one atmosphere apparently introduces processes which, for the present conditions, offset the aforementioned advantages of reduced blue-green absorption in Ar-Kr buffered mixtures.\(^1\)

IV. Spectral Analysis

Although the primary effect of an XeF(C \( \rightarrow \) A) buffer gas comprised of both Ar and Kr appears to be a reduction of absorption in the blue-green region, spectral analysis of laser mixtures has revealed other beneficial effects. Presented in Fig. 4 are time-integrated fluorescence spectra for an e-beam excited XeF(C \( \rightarrow \) A) laser mixture for various values of Kr pressure. Aside from the expected appearance of the 248 nm KrF(B \( \rightarrow \) X) fluorescence, examination of Fig. 4(a)-(d) reveals other interesting features. Upon addition of 50 Torr Kr, the XeF(B \( \rightarrow \) X) fluorescence decreases significantly, but there is no measurable change in the value of the peak XeF(C \( \rightarrow \) A) fluorescence near 480 nm. Certainly, Kr addition at the level indicated changes the XeF(B, C) precursors as described previously, and introduces the possibility of additional loss processes as well. However, for an Ar pressure of 6 atm the XeF B and C states are collisionally coupled and therefore their populations should either increase or decrease together in response to changes in XeF formation or loss processes due to the presence of Kr. One explanation for the decrease and then leveling off of the time integrated XeF(B \( \rightarrow \) X) fluorescence as Kr is added (Fig. 4(a)-(d)) is enhanced B-C mixing. Since > 95 percent of the XeF(B, C) population is in the C state for the condi-

\(^1\)J. D. Campbell, C. H. Fischer, and R. E. Cramer, Appl. Phys. Lett., vol. 37, p. 348, 1980. The authors found that use of 1 atm Kr instead of 2 atm Ar as the buffer for an e-beam excited XeF(C \( \rightarrow \) A) laser resulted in increased laser energy, an effect attributed to a lower level of broad-band absorption in the Kr-buffered mixture. However, in that work the e-beam current density was more than an order of magnitude less than that of the present work, and the duration of the e-beam pulse was about two orders of magnitude longer, factors resulting in medium characteristics very different and output energy levels very much less than those of the present investigation.
tions of Fig. 4, faster B-C mixing favoring the C state would be observed as a reduction in the B → X fluorescence intensity, with the XeF(C) fluorescence (population) remaining essentially unchanged. Krypton is known to be much more effective than Ar in driving the coupled XeF(B, C) manifolds towards equilibrium [17], and comparison of Fig. 4(a) and (b) suggests that this effect of Kr may be significant even for an Ar pressure of 6 atm and an Ar-Kr ratio of ~100:1.

A. KrF Kinetics

Fig. 4 also shows a distinct change in the nature of the spectra between 375 and 425 nm as the Kr pressure is increased. This feature is not caused by a change in the XeF(C → A) spectra but, rather, results from the gradual development of broadband KrF fluorescence centered at ~400 nm [19]. Presented in Fig. 5 are spectra for conditions similar to those of Fig. 4(c), but with the Xe pressure varied. In the absence of Xe the KrF fluorescence is clearly apparent [Fig. 5(a)]. Indeed, the mixture of Fig. 5(a) is essentially the same as that found to be optimum for e-beam excitation of the KrF laser [20]. However, addition of as little as 1 torr Xe [Fig. 5(b)] significantly alters the spectrum with the appearance of both 351 nm XeF(B → X) and XeF(C → A) fluorescence, the latter overlapping the KrF fluorescence to produce a nearly constant spectral intensity extending from 375-500 nm. The significant decrease in the magnitude of the KrF fluorescence when only 1 torr of Xe is added reflects the large rate coefficient (>10^{-10} \text{s}^{-1}\text{cm}^3) for KrF quenching by Xe [21]. Nonetheless, the KrF number density is comparable to that of XeF(C) for optimum XeF(C → A) laser conditions [8], ~5 × 10^{15} \text{cm}^{-3}. Fig. 5 shows the dependence of the peak KrF population on Kr pressure for conditions similar to those of Fig. 4, estimated on the basis of fluorescence data and assuming a 200 ns lifetime [19] for KrF.

1) KrF Formation: Very recent measurements [22] have shown that the four-atomic rare-gas-halide exciplex ArF is stable and is readily formed from ArF in Ar–F2 and Ar–NF3 e-beam excited mixtures at pressures ≤1 atm. Since the binding energy of ArF is somewhat larger than that of ArF (0.22 eV), this new finding suggests that KrF formation may have an important bearing on the KrF population in the present experiment. Indeed, we believe that the leveling off of the KrF number density as the Kr pressure is increased above ~100 torr (Fig. 6) is the result of three-body quenching of KrF, probably resulting in the formation of KrF.

The trend exhibited by the data of Fig. 6 is consistent with a rate coefficient of ~10^{-10} \text{s}^{-1}\text{cm}^3 for the reaction, KrF + Kr + Ar → KrF + Ar. Based on this interpretation, it seems likely that R2X → R1X kinetics may play a significant role for conditions typical of many rare-gas halide lasers. Additionally, it appears that all prior measurements of R1X rate coefficients, lifetimes and other phenomena dependent on interpretation of R1X collision processes should be reexamined in light of this development.

Fig. 5. Time-integrated fluorescence spectra for a mixture comprised of 6 atm Ar, 200 torr Kr, 8 torr NF3, and 1 torr F2 (a), and the same mixture with 1 torr Xe added (b), 3 torr Xe (c), and 8 torr Xe (d). Note that the relative intensities of the B → X KrF and XeF fluorescence should be the same in Fig. 4(c) and Fig. 5(b) for which the experimental conditions are the same. However, the calibration correction applied to the OMA data is very large in the UV so that the absolute KrF(B → X) intensity levels of Figs. 4 and 5 are rather uncertain.
2) KF Effect on Blue-Green Gain: Presented in Fig. 7 is an enlarged version of Fig. 4(d) in which are indicated the KrF and XeF(C → A) contributions to the time-integrated fluorescence in the 300-600 nm region. The fact that the XeF(C) and KrF populations are comparable in XeF(C → A) mixtures containing Kr suggests that KrF may be making a contribution to the gain on the short wavelength side of the laser spectrum. However, the broader spectral width, longer natural lifetime, and shorter wavelength of KrF compared to XeF(C → A) results in a stimulated emission cross section that we estimate to be only ~2.5 x 10^-18 cm^2 at its peak at 400 nm, a value approximately one fourth that of the XeF(C → A) transition. Thus, any influence of KrF on the gain for the wavelengths ~480 nm must be quite small.

Other factors also have to be considered, however. Fig. 8 presents the temporal evolution of both the KrF and XeF(C → A) fluorescence spectra, showing that the peak in the KrF fluorescence precedes that of XeF(C → A) by 15-20 ns. Therefore, even though its stimulated emission cross section is small, a large KrF population favors an earlier buildup of gain for wavelengths to the short wavelength side of the XeF(C → A) gain maximum. For example, based on the data of Figs. 7 and 8 we estimate that the KrF contribution to the peak gain at ~450 nm could be as high as 10 percent. Such an effect could be significant when the C → A gain medium is used as a wavelength tuned amplifier [12, 23]. This line of reasoning suggests that any increase in the gain of the XeF(C → A) laser medium due to the presence of KrF should be observable only on the short wavelength side of the laser spectrum. In fact, comparison of free-running laser spectra for Ar and Ar-Kr buffered mixtures shows a ~50 percent greater increase in the XeF(C → A) laser intensity around 470 nm than at 490 nm when Kr is added [11].

V. SIMULTANEOUS UV/VISIBLE LASER OSCILLATION

Although more than 95 percent of the total XeF(B, C) population resides in the C state for the conditions of primary interest to us, since the stimulated emission cross section [1] for XeF(B → X) is 3-4 x 10^-18 cm^2, the intense pumping required to produce adequate XeF(C → A) net gain usually results in even larger transient gain at the XeF(B → X) 351 nm wavelength. Thus, even though steps can be taken in the design of the C → A cavity to minimize the possibility of B → X oscillation, the B → X gain is frequently so high that amplified stimulated emission of the B → X transition occurs thereby depleting the gain on the desired C → A transition.

In earlier work [24] we took advantage of this characteristic in order to demonstrate the feasibility of simultaneous UV/visible laser oscillation on both the B → X and C → A transitions of the XeF excimer. However, the dual wavelength cavity used was far from optimum so that very low levels of laser energy and efficiency were obtained. Recently [13], these dual wavelength laser experiments
were repeated with two important differences: 1) a cavity optimized for laser oscillation at 351 nm and at 480 nm was used, and 2) Kr was added to Ar to form a two component buffer mixture as in the work described herein. The cavity was comprised of a mirror having nearly total reflectivity in the UV and blue-green regions, and a multiple coating out-coupler having a transmission of 20 percent at -350 nm and 10 percent between 460 and 510 nm. Fig. 9 presents the measured laser pulse energy densities for the UV and blue-green transitions of XeF as a function of Kr pressure, obtained using this dual wavelength cavity for conditions otherwise similar to those of Figs. 3-8. With no Kr in the mixture and using a C -> A cavity alone, previously these conditions resulted in C -> A laser pulses having an energy density of approximately 1 J / J / 11. However, Fig. 9 shows that when the optimized dual-wavelength cavity is used, no C -> A laser oscillation is observed in the absence of Kr, but the B -> X transition oscillates with an output pulse energy density of ~ 1 J / J , a relatively high energy density level considering that the mixture used is very different from that found to be optimum for the XeF (B -> X) laser. [3], [25]. Addition of Kr results in an immediate decrease in B -> X output, followed by simultaneous oscillation of the UV and visible transitions, and eventually C -> A oscillation alone for Kr pressures above ~ 300 torr.

A. UV Absorption

The decrease in B -> X laser energy (Fig. 9) upon addition of Kr is the result of a reduction in the XeF(B) population relative to that of XeF(C) as discussed previously [Fig. 4(a) - (d)] and, perhaps more significantly, because of the likelihood of a strong increase in absorption at ~ 351 nm due to the presence of KrF. Calculations of the low-lying states of ArF and KrF have shown that the presence of F + has a negligible effect on the states of ArF and KrF, respectively [26]. For this reason, the strong near UV absorption of the rare gas dimer ions [27] should carry over directly to the triatomic rare gas halides. Since the dominant absorption of KrF has a cross section of ~ 4 x 10^-13 cm^2 at the 351 nm wavelength of the XeF(B -> X) transition [27], the KrF population estimates of Fig. 6 indicate that a Kr partial pressure as low as 50 torr is likely to result in a very significant KrF contribution to absorption at 351 nm. We feel that this is the primary cause of the decrease in XeF(B -> X) laser output as Kr is added (Fig. 9). This trend continues as Kr pressure is increased and for Kr pressures above about 150 torr, for which the presence of Kr results in a reduction in absorption in the blue-green region, significant laser output is obtained from the C -> A transition. For Kr pressures between about 150-250 torr combined UV/visible output in excess of 0.5 J / J is obtained using the cavity optimized at both the B -> X and C -> A wavelengths, corresponding to an intrinsic efficiency of ~ 0.4 percent. However, as the Kr pressure is increased above ~ 300 torr, for which optimum C -> A performance has been demonstrated [11], the B -> X output decreases to a very low level. For this condition the beneficial influence on C -> A laser performance is at its maximum, while the peak absorption at 351 nm due to KrF is estimated on the basis of the KrF absorption cross section to be on the order of 10 percent cm^-1. In view of the fact that the cavity used for this demonstration was specifically designed to support XeF(B -> X) oscillation (Fig. 9), we interpret these results as evidence that the two component Ar-Kr buffer used to optimize XeF(C -> A) laser performance alone, when used with C -> A optics, significantly reduces the possibility of competitive oscillation on the parasitic B -> X transition, a particularly important consideration for the design of efficient XeF(C -> A) lasers.

VI. Summary

The findings of this investigation show that use of Ar and Kr together to form synthesized rare gas buffer properties results in unique XeF(C -> A) laser medium characteristics not attainable using either species alone. All aspects of the medium are affected by the two-component buffer, including: XeF(B, C) formation and quenching: B, C state vibrational relaxation and mixing; and transient absorption in both the UV and blue-green spectral regions. For this reason use of two rare gas components to form the buffer introduces an extra element of flexibility and control as regards optimization of laser medium characteristics, with no penalty in the area of system complexity. Indeed, e-beam excited XeF(C -> A) media optimized on the basis of the two-component buffer described herein have resulted in levels of blue-green laser pulse energy density and intrinsic efficiency rivaling those of UV RGH lasers, with the added element of broad-band tunability [11], [12], [23].

Although the present work is focused on e-beam excited XeF(C -> A) laser media, there is no apparent reason why other RGH lasers using either e-beam or discharge excitation cannot be improved by increasing the number of mixture constituents beyond the usual three. Preliminary evidence supporting this conclusion is provided by our recent demonstration [28] of simultaneous laser oscillation on the KrF (248 nm) and XeF(351) RGH transitions and on the ArF(193 nm) and KrF (248 nm) transitions using discharge excitation of unoptimized mixtures comprised of He, Kr, Xe, and F2, and He, Ar, Kr, and F2, respectively. In that work XeF B -> X laser oscillation at 351 and KrF oscillation at 248 nm were demonstrated separately at pulse energy levels only slightly less than the highest obtainable using optimized three component mixtures in the same device. Thus, considering the complexity of the kinetics of these systems and the fact that no attempt was made to optimize the four component mixtures, there is reason to believe that the efficiency of UV RGH lasers may be further improved through use of four (or more) component gas mixtures once optimum mixtures and complementary pumping levels and cavity properties are identified.
ACKNOWLEDGMENT

The skillful experimental assistance of B. Zook, J. Kinross-Wright, and J. Hosten is gratefully acknowledged.

REFERENCES

Frank K. Tittel (S'72, M'78) was born in Berlin, Germany, in 1933. He received the M.A. and Ph.D. degrees from Oxford University, Oxford, England.

From 1959 to 1967 he was a Research Physicist at the General Electrical Research and Development Center, Schenectady, NY. Since 1967 he has been at Rice University, Houston, TX, where he is a Professor with the Department of Electrical and Computer Engineering. His research interests include laser devices, nonlinear optics, and laser spectroscopy.

Dr. Tittel is a member of the IEEE Laser and Electro-Optics Society, the Optical Society of America, and the American Physical Society.

William L. Wilson, Jr. (S'76-M'77) was born on February 6, 1943. He received the B.S. degree in 1965, the M.S. degree in 1966, and the Ph.D. degree in 1972, all in electrical engineering, from Cornell University, Ithaca, NY.

From 1971 to 1972 he was an Instructor-Research Associate with the Electrical Engineering School at Cornell. From 1972 to the present he has been associated with the Department of Electrical and Computer Engineering, Rice University, Houston, TX, where he now holds the position of Professor. His research interests include tunable excimer lasers and solid-state devices.

Dr. Wilson is a member of Tau Beta Pi, Eta Kappa Nu, Sigma Xi, the IEEE Magnetics Society, IEEE the Microwave Theory and Techniques Society, and the IEEE Electron Devices Society.