Tunable Electron Beam Pumped Excimer Lasers

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Introduction

Recently there has been considerable interest in diatomic and triatomic excimer lasers capable of UV and visible wavelength tunability. In this paper the characteristics of two new blue-green electron beam pumped lasers, XeF (C → A) [1] and Xe_2Cl [2] will be described.

The experiments carried out so far were performed in a stainless steel cell attached to the field emission diode of a Pulserad Model 110 electron beam generator shown in Fig. 1 and described in detail in Ref. 3. A beam of 1 MeV electrons with a pulsewidth of 10 ns (FWHM) and a current density of 0.8 kA/cm^2 was injected transversely into the laser medium through a 50 μm thick titanium anode foil over an area of 10 cm^2. This resulted in an input beam energy of 80 j. The laser cavity was formed by two reflectors positioned in the cell as shown in Fig. 2. The sapphire or fused silica vacuum windows were placed external to the cavity, thereby avoiding reflection losses which are significant for low gain laser media. Typically, one reflector was a 1 m radius mirror (R > 99.8%) while the other was a flat output mirror (T = 2%). The two mirrors were separated by 10 cm and the optical axis was 1.5 cm from the anode foil. The bellows design of the mirror mounts permitted convenient external alignment of the intracell reso-

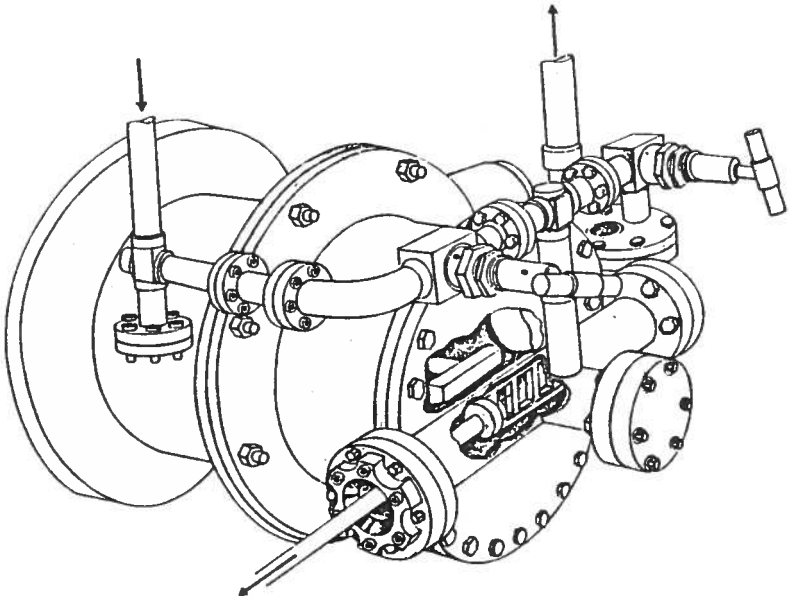


FIG.1

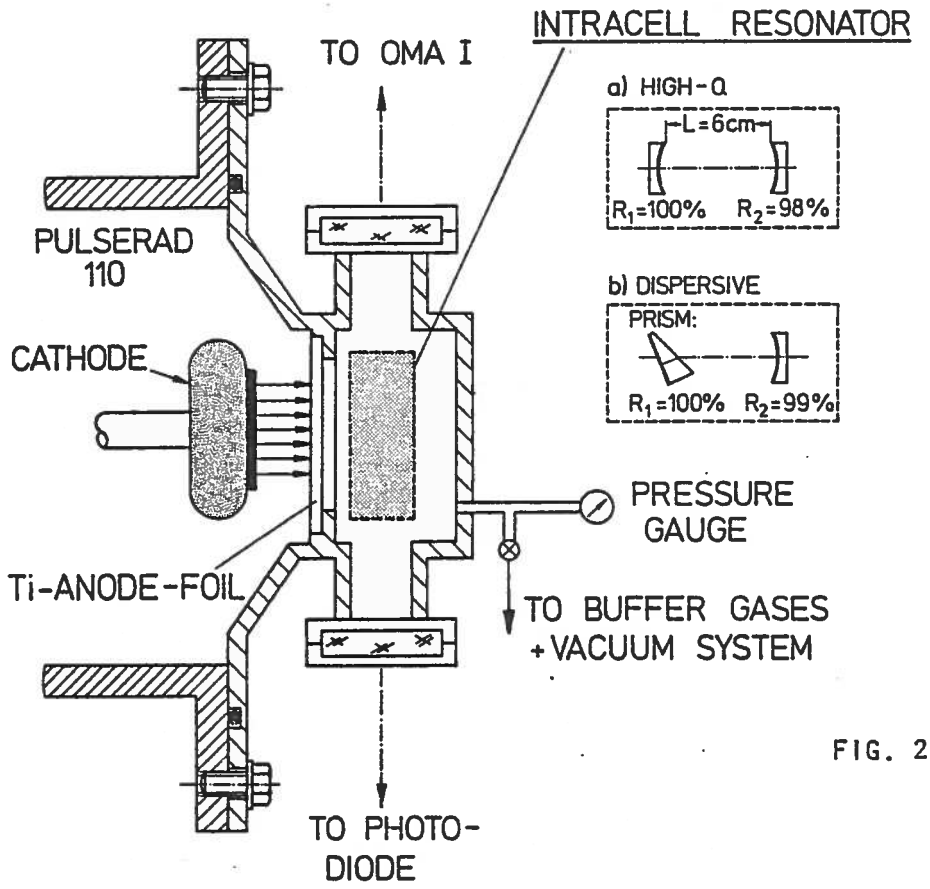

INTRACELL RESONATOR
TO OMA I
a) HIGH-Q
L=6cm
R1=100%
R2=98%
b) DISPERSIVE
PRISM:
R1=100%
R2=99%
PULSERAD
110
CATHODE
PRESSURE
GAUGE
Ti-ANODE-FOIL
TO BUFFER GASES
+VACUUM SYSTEM
TO PHOTO-
DIODE

FIG. 2

nator. The fluorescence and laser signals were monitored in the axial direction time resolved by ITT F400-S-5 photodiodes and simultaneously spectrally resolved with a spectrograph or an optical multichannel analyzer (PAR OMAI). A Faraday cup probe mounted inside the reaction cell monitored the current density of the electron beam. In this manner it is possible to observe the time history of the electron beam excitation pulse and the subsequent temporal evolution of the fluorescence or laser pulses.

Spectral and temporal fluorescence, gain and laser measurements of high pressure Ar,Ne/Xe/NF_3 and Ar,Ne/Xe/CCl_4 mixtures for the B → X and C → A transitions of XeF centered at 351 and 486 nm and for XeCl and Xe_2Cl transitions at 308 and 518 nm respectively have been carried out. These observations are summarized below:

XeF (C → A) Laser Studies

Electron beam or discharge pumped XeF excimer systems usually operate at the 351 nm B → X transition (Fig. 3). Fluorescence measurements have shown the existence of a broad continuum band centered near 476 nm, which originates from the C state which lies below the B state by about 700 cm^{-1} and terminates on the repulsive A state. This has resulted in obtaining laser emission in the blue-green on the bound-free C → A transition at 486 nm [1, 4 - 8] in addition to the stimulated emission on the prominant bound-bound B → X laser transition at 351 nm first observed by Ault, et. al. [9], and Brau and Ewing [10]. Laser action on the C → A transition was demonstrated by Bischel [4] with photodissociative excitation of XeF_2, by Ernst and Tittel [1] of Rice University in e-beam pumped Ar/Xe/NF_3

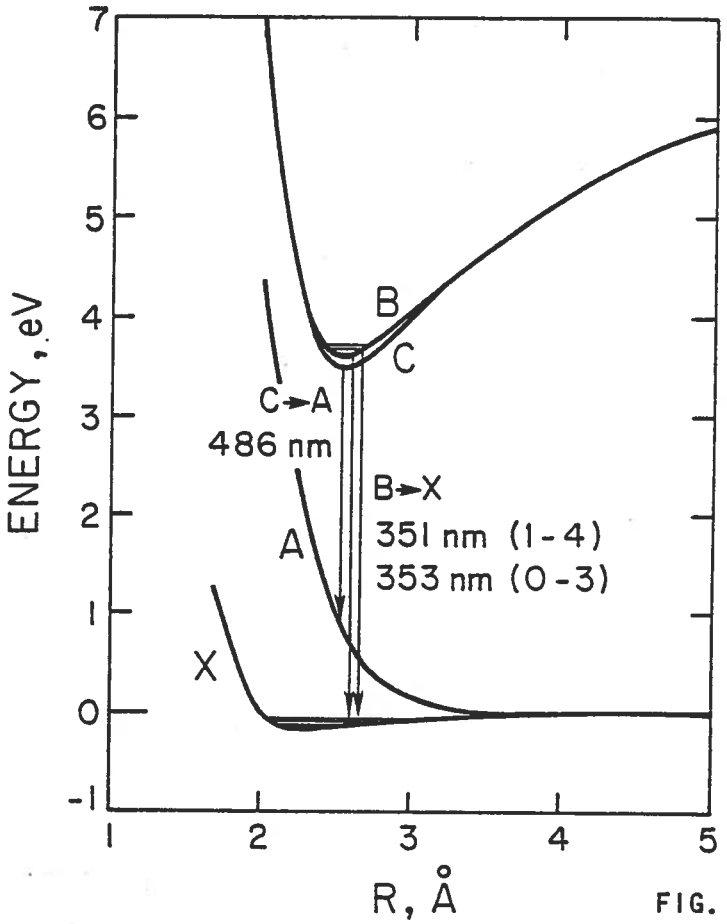

ENERGY, eV
R, Å
B
C
C→A
486 nm
B→X
351 nm (1-4)
353 nm (0-3)
A
X

FIG. 3

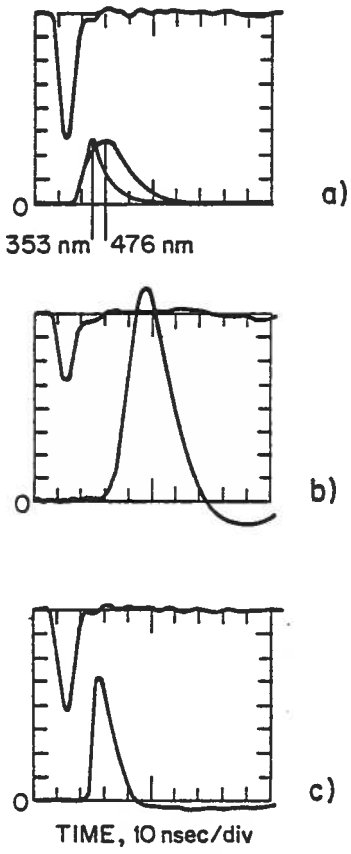
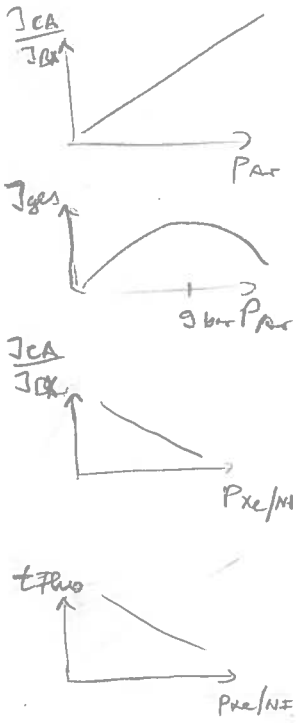

a)
353 nm
476 nm
b)
c)
TIME, 10 nsec/div

FIG. 4

mixtures, by Burnham [5] and by Fisher, et.al. [6] in discharge pumped $Ne/Xe/NF_3$ mixtures, and by Basov, et.al. using exploding wire pumped techniques [7].

Fluorescence studies established the optimum conditions for emission via the C → A transition as compared to the B → X transition. The fluorescence band is centered at 476 nm and has a FWHM of 55 nm. Typical temporal characteristics of the electron-beam pulse and fluorescence intensity at 353 and 476 nm for a mixture of 16 Torr Xe, 8 Torr NF_3, and 600 kPa Ar are shown in Fig. 4(a). For xenon pressures between 4 and 30 Torr and NF_3 pressures of 4 - 16 Torr the 476-nm/353-nm fluorescence intensity ratio increased with increasing Ar buffer gas pressure. The total fluorescence output increased with Ar pressures up to 900 kPa, and then decreased, possibly due to higher absorption by the Ar excimers [11]. Lower Xe and NF_3 pressures within the range mentioned above improved the 476-nm/353-nm fluorescence intensity ratio and lengthened the fluorescence pulses, but they considerably reduced the fluorescence peak power. The fluorescence pulse length was only determined by the Xe and NF_3 partial pressures, and not by the buffer-gas pressure. The 476-nm emission peak appeared ~20 ns later than the 353-nm output. When neon was used as a buffer gas instead of argon, the 476-nm/353-nm fluorescence intensity ratio always remained below 0.5, but the total output increased with pressure.



The temperature dependence of the measured fluorescence peak inten-

sity was investigated [12]. For 600 kPa argon buffer gas the B → X fluorescence increases with temperature up to 530K and then starts to decrease [Fig. 5]. At the same time the C → A fluorescence decreases over the whole temperature range. Between 300 and 530K the B → X/C → A fluorescence ratio changes by a factor of 3.5, which compares to a change of the B/C population ratio by a factor of 4 in case of a Boltzmann distribution. With 600 kPa neon buffer gas the B → X fluorescence intensity at room temperature was as high as that for argon buffer gas at the optimum temperature of 520K. For Ne the fluorescence intensity decreased for elevated temperatures. The observed C → A fluorescence was extremely small for Ne. Therefore all C → A measurements were only performed with Ar buffer gas.

Gain measurements were performed using an argon ion laser as a cw probe laser. After passing the cell the beam was directed through several apertures to a monochromator with a fast photomultiplier placed 6.5 m away in a Faraday cage. A brick wall and lead shielding helped to avoid X-ray noise generated by the e-beam accelerator (see Fig. 6). Influence of fluorescence light from the cell could not be detected and attenuation of the laser light with filters prevented the photomultiplier from saturating. Gain on the B → X and C → A transitions was measured using the argon ion laser lines at 351 and 488 nm respectively. The experiments were performed with mixtures of 16 Torr Xe, 8 Torr NF_3, and 400 and 600 kPa Ar or Ne. Gain data for both the B → X transition at 351 nm and the C → A transition at 488 nm are shown in Fig. 7.

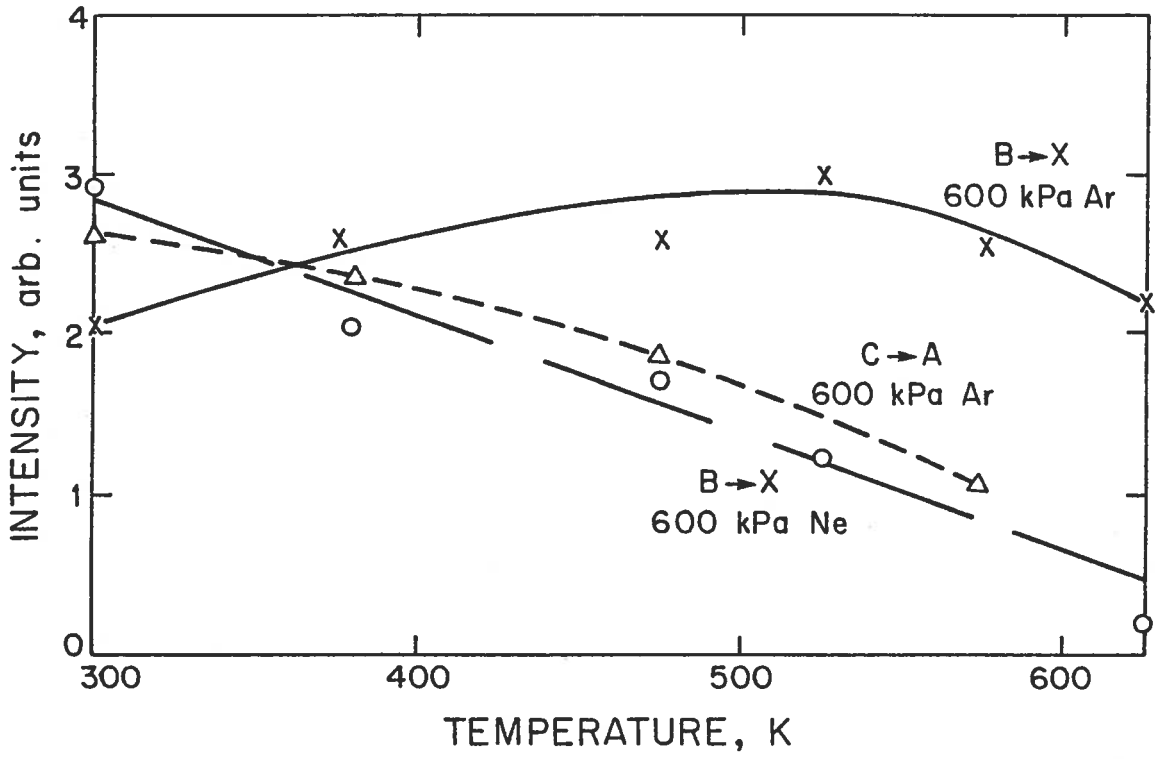

INTENSITY, arb. units
TEMPERATURE, K
B→X
600 kPa Ar
C→A
600 kPa Ar
B→X
600 kPa Ne
0
1
2
3
4
300
400
500
600

FIG. 5

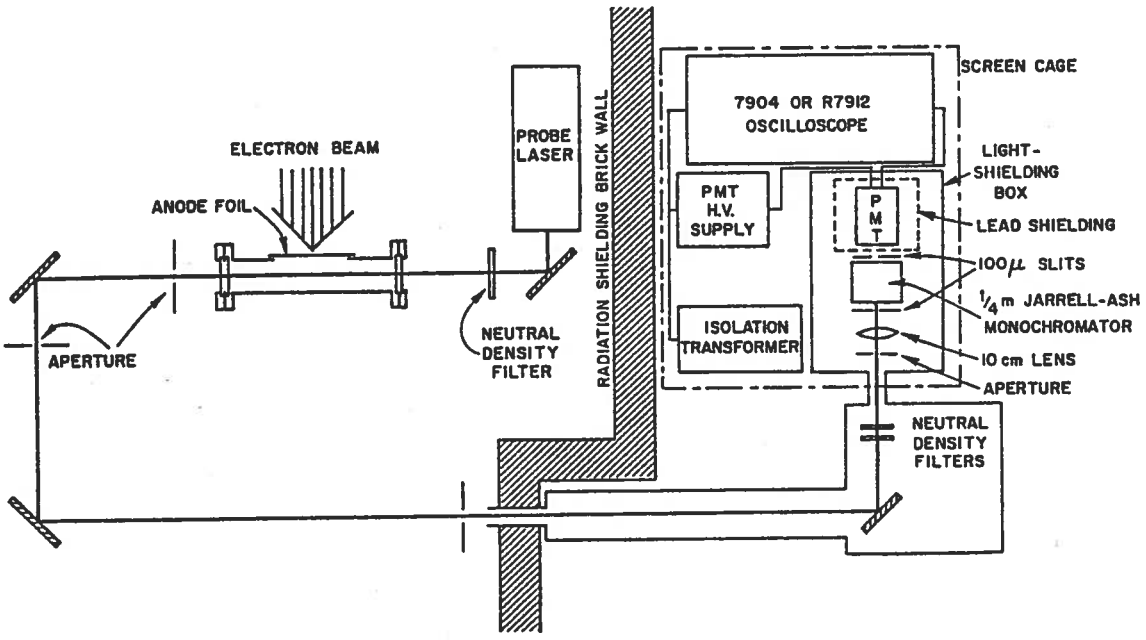

ELECTRON BEAM
ANODE FOIL
APERTURE
PROBE LASER
NEUTRAL DENSITY FILTER
RADIATION SHIELDING BRICK WALL
7904 OR R7912 OSCILLOSCOPE
SCREEN CAGE
PMT H.V. SUPPLY
ISOLATION TRANSFORMER
P M T
LIGHT-SHIELDING BOX
LEAD SHIELDING
100μ SLITS
¼m JARRELL-ASH MONOCHROMATOR
10 cm LENS
APERTURE
NEUTRAL DENSITY FILTERS

FIG. 6

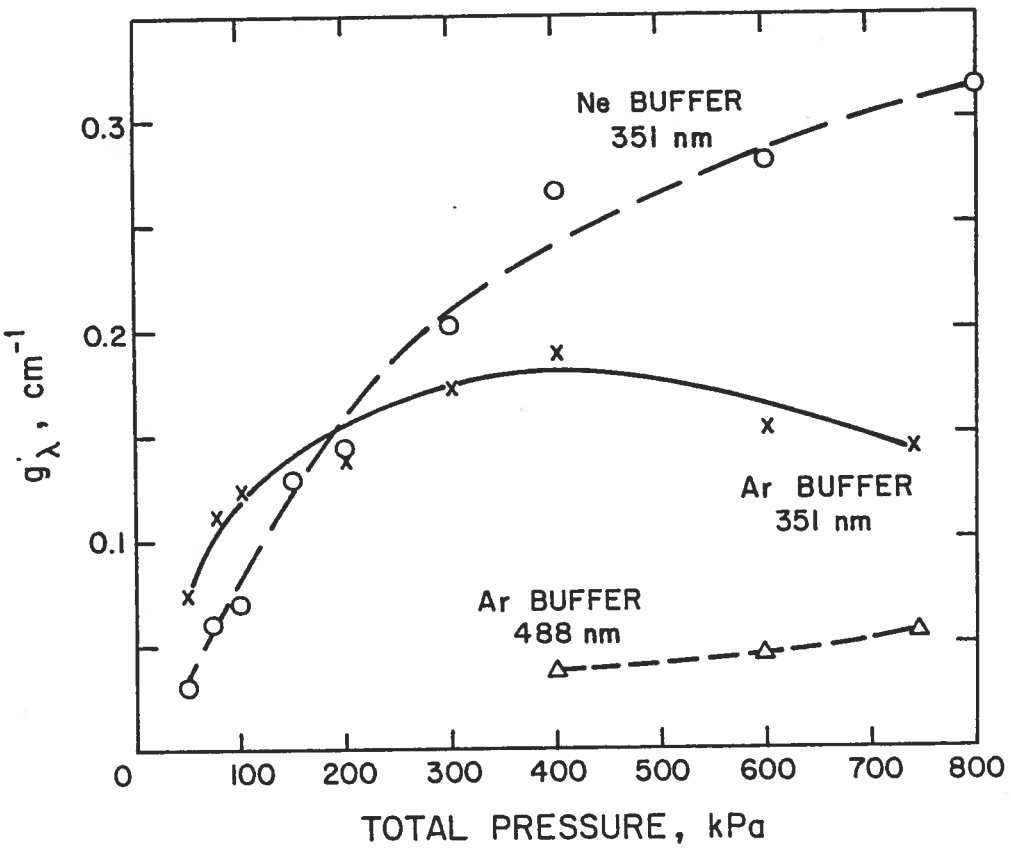

Ne BUFFER
351 nm
Ar BUFFER
351 nm
Ar BUFFER
488 nm
0.3
0.2
0.1
0
100
200
300
400
500
600
700
800
g'_λ, cm^{-1}
TOTAL PRESSURE, kPa

Fig. 7

Laser emission occurred for mixtures of Xe partial pressure between 4 and 25 Torr, NF_3 pressure between 4 and 13 Torr, and Ar buffer gas pressures of 350 - 800 kPa. A mixture of 16 Torr Xe, 8 Torr NF_3, and 600 kPa Ar proved to be optimum with a peak output power of the order of 5 kW for a broadband 98 reflectivity output coupler (460 - 510 nm). The laser pulse occurs in the afterglow of the e-beam excitation, as depicted in Fig. 4(b), which is in agreement with the results of the gain measurements reported in Ref. [13]. Lasing just above threshold occurred when an 80% reflectance coupler was used. From these observations a gain of approximately 25% per round trip can be deduced for an optimum Ar/Xe/NF_3 mixture, which is slightly higher than the gain of 8% per pass reported in Ref. [13]. A laser spectrum of the C → A transition at room temperature is shown in Fig. 8. Enhanced intracavity absorption bands, due to transient species, are responsible for the line structure of the laser output, which is centered at 486 nm. The envelope had a FWHM of 12 nm.

Xe_2Cl Laser Studies

Transitions of interest in emission from diatomic XeCl and triatomic Xe_2Cl excimers are shown in Fig. 9 which depicts the potential energy curves for those species. In the case of XeCl (Fig. 9a) both an intense, narrow linewidth B → X transition and a much weaker, broadband C → A transition are observed at 308 and 345 nm, respectively. For triatomic Xe_2Cl broadband emission centered at 500 nm occurs between strongly bound ionic upper states and repulsive ground states [14,15] as shown in Fig. 9

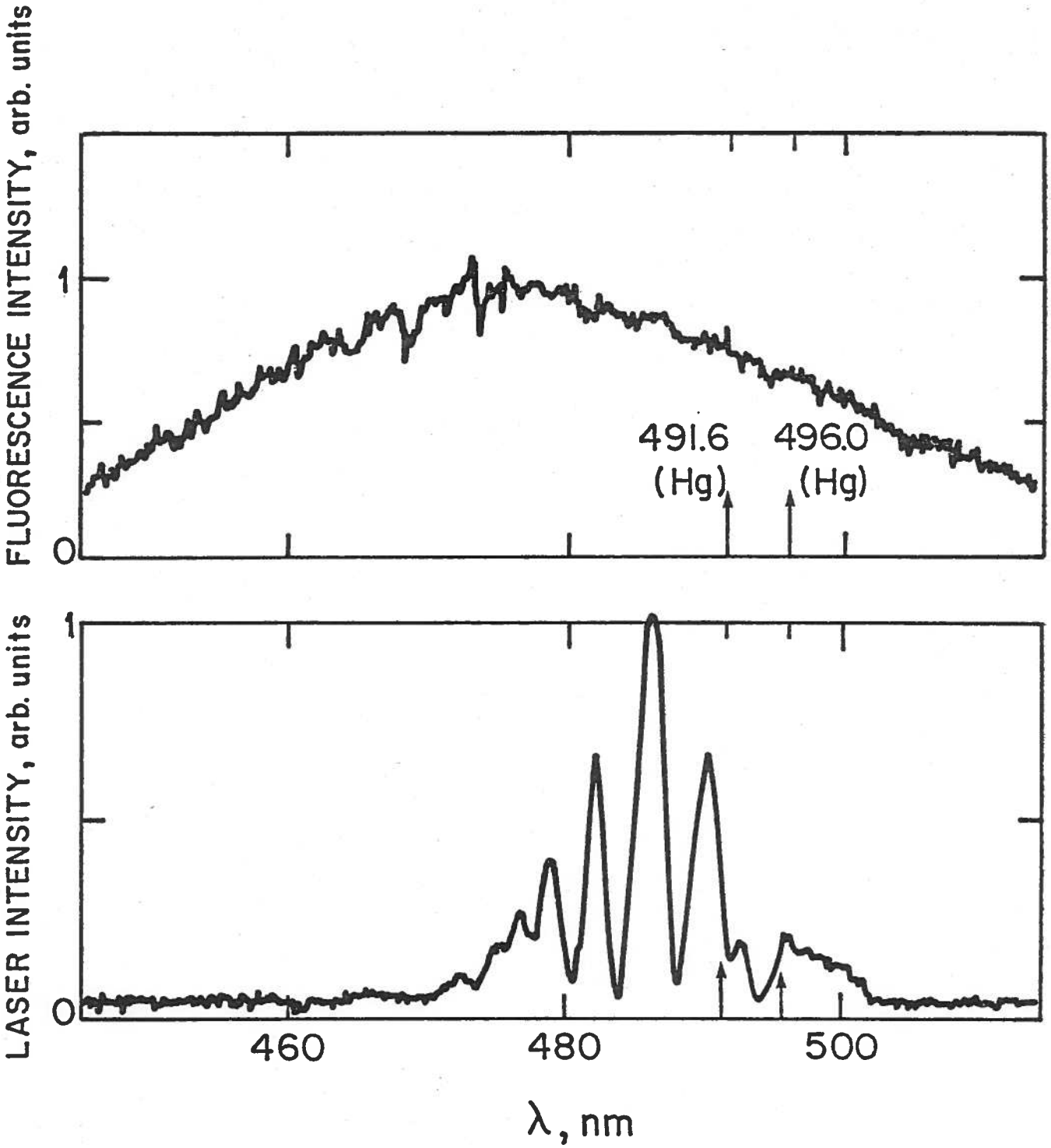

FLUORESCENCE INTENSITY, arb. units
1
0
491.6
(Hg)
496.0
(Hg)
LASER INTENSITY, arb. units
1
0
460
480
500
λ, nm

FIG. 8

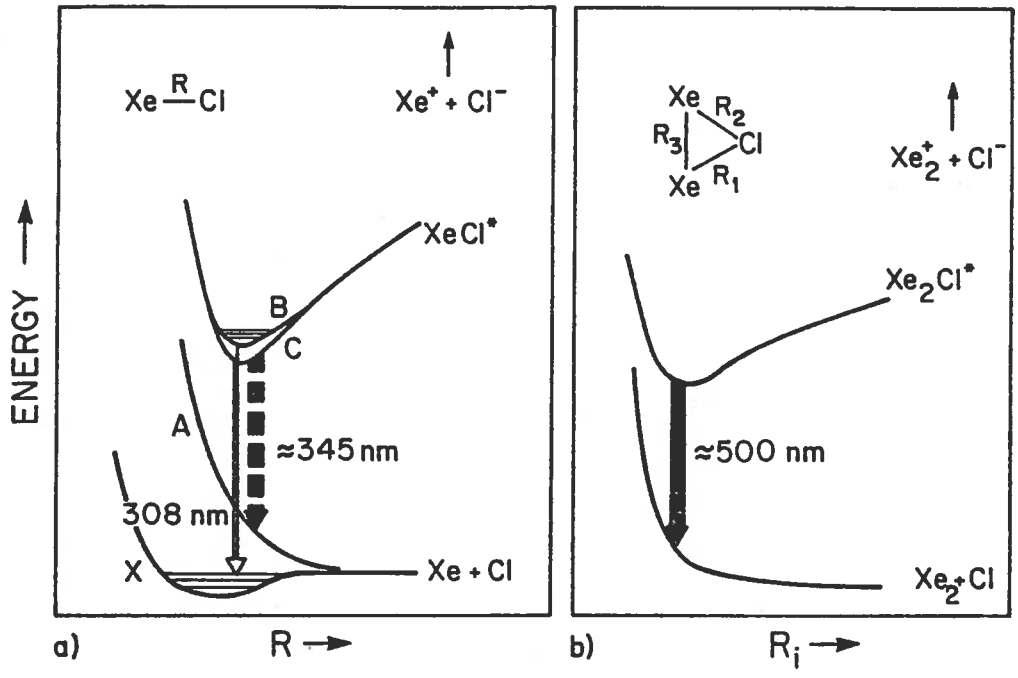

Xe—R—Cl
Xe⁺ + Cl⁻
ENERGY
XeCl*
B
C
A
≈345 nm
308 nm
X
Xe + Cl
a)
R
Xe
R2
R3
Cl
Xe
R1
Xe2⁺ + Cl⁻
Xe2Cl*
≈500 nm
Xe2+Cl
b)
Ri

FIG. 9

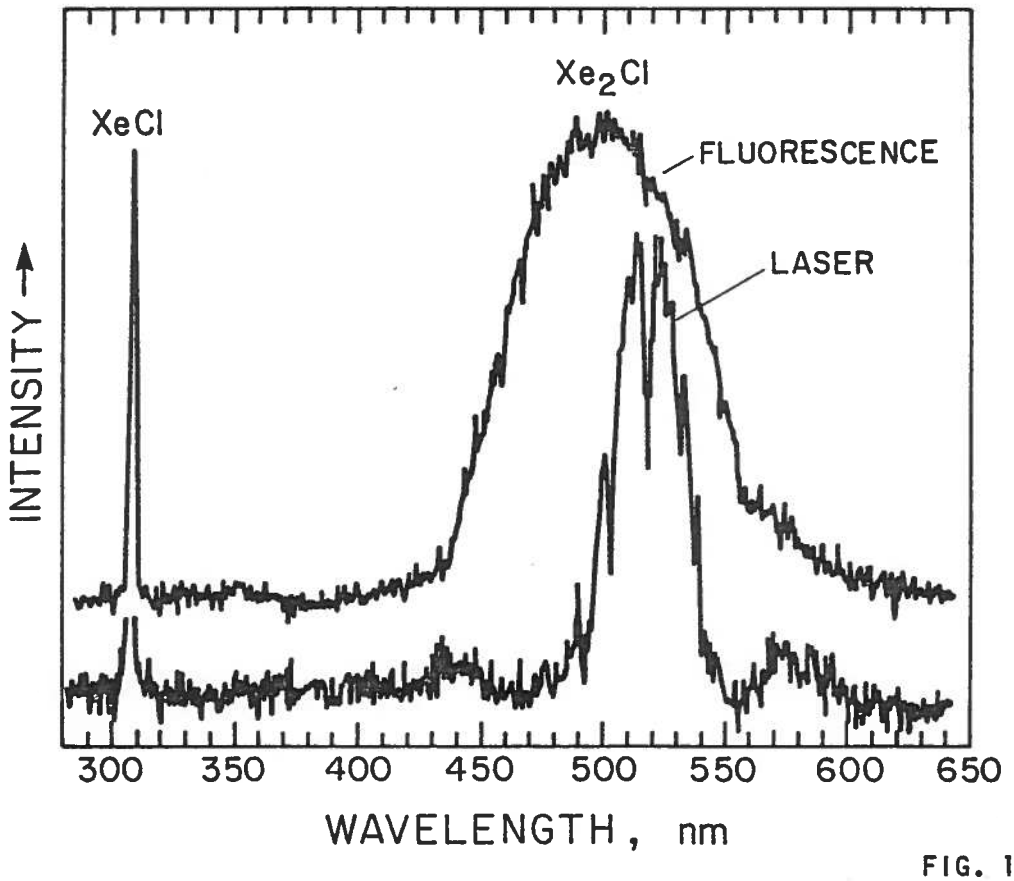

Xe2Cl
XeCl
FLUORESCENCE
LASER
INTENSITY
300
350
400
450
500
550
600
650
WAVELENGTH, nm

FIG. 10

Fluorescence measurements were used to determine the optimum composition of a gas mixture for enhanced Xe_2Cl emission as compared to XeCl emission, which is always present, since it can be considered as a precursor in the reaction chain finally leading to the formation of Xe_2Cl [15]. The range of pressures of argon, xenon, and CCl_4 evaluated was 4 to 9 Atm., 100 to 750 Torr, and 0.5 to 10 Torr, respectively. Low xenon pressures (< 100 Torr) favored increased XeCl excited state production. A time integrated fluorescence spectrum obtained with the optical multichannel analyzer for a typical mixture of 6.5 Atm. Ar, 200 Torr Xe and 1 Torr CCl_4 is shown in Fig. 10. The Xe_2Cl spontaneous emission has a spectral bandwidth of approximately 100 nm (FWHM) and exhibits weak absorption features.

On the other hand, the Xe_2Cl laser spectrum which has a center wavelength of 518 nm shows significant spectral narrowing from 100 nm (spontaneous emission) to 20 nm (laser). In addition, greatly enhanced intracavity absorption features are observed. The laser output spectrum is red shifted relative to the fluorescence spectrum by approximately 20 nm. Since the reflectivity of the laser mirrors remains nearly constant over this spectral range, cavity pulling effects can be excluded. Instead, the observed shift is due to the spectral dependence of the stimulated emission cross-section. Furthermore, the cavity enhanced absorption features shown in Fig. 10 can be compared to the visible atomic and molecular absorption characteristics for the electron beam pumped argon diluent reported in Ref. 11. Two observed absorption peaks at 519 nm and 537 nm are in good agreement with the published data on molecular absorptions in

argon [11]. In fact the intracavity laser enhancement of absorption features reveals additional features at 493, 503, and 532 nm which may be attributed to enhanced Ar absorption or to transient absorptions of the other components (Xe, CCl_4) not observed by the single pass absorption measurements.

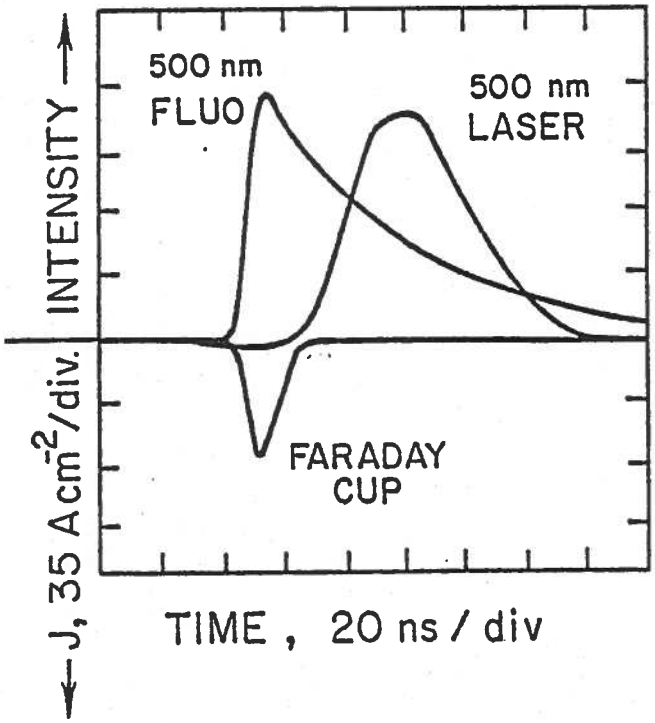
Also, the spectrally resolved temporal behavior of Xe_2Cl has been studied. In Fig. 11a the Xe_2Cl fluorescence and laser pulse shape as monitored by the photodiode via a 500 nm interference filter is depicted. The electron beam pulse as monitored by a Faraday probe is shown for calibration. From this data it is apparent that laser emission occurs with a delay of some 40 nsec after the excitation pulse and onset of fluoresence, as a result of the visible absorption effects referred to in connection with Fig. 10. Fig. 11b depicts the xenon pressure dependence of this laser delay and the pulsewidth of the laser emission. The FWHM pulsewidth of the Xe_2Cl stimulated emission is typically 50 ns as compared to a fluorescence bandwidth of 80 ns.

The stimulated emission cross-section can be estimated based on the fluorescence spectrum and decay time. The excimer gain cross-section is given by [15,20] as

$$\sigma = \frac{1}{4\pi c\tau} (\ln 2/\pi)^{\frac{1}{2}} \frac{\lambda^4}{\Delta\lambda}$$

where τ is the spontaneous decay time and $\Delta\lambda$ is the FWHM of the fluorescence spectrum. For Xe_2Cl: $\lambda \sim 500$ nm, $\Delta\lambda \sim 80$ nm, and $\tau \sim 80$ nsec. Longer ?

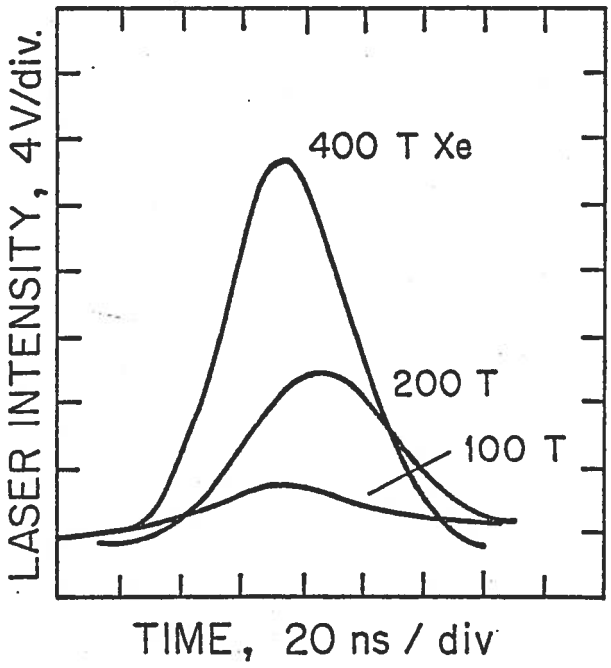
This gives a $\sigma_{Xe_2Cl} \sim 1.1 \times 10^{-17}$ cm^2, which is considerably less than $\sigma_{XeCl} = 4.5 \times 10^{-16}$ cm^2 for the main line XeCl transition. The gain



$C Cl_4$ = 2 Torr
Xe = 400 Torr
Ar = 6 atm

$J_{EB} \approx$ 10 ns
$J_{FL} \approx$ 80 ns.
$J_{LA} \approx$ 45 ns
$J_{DEL} \approx$ 40-50 ns

FIG. 11a



$C Cl_4$ = 2 Torr
Ar = 6 atm

FIG. 11b

coefficient g_o may be expressed by $g_o = \sigma N^*$, where the value of the upper laser state density may be determined by comparing it to a previously analyzed laser system, such as XeF ($C \rightarrow A$) or N_2 ($C \rightarrow B$). For $N^* = 10^{16}$ cm^{-3}, $g_{calc} = 0.02$ cm^{-1}, which is in agreement with g_{thresh} obtained by establishing the laser threshold reflectivity of the cavity output coupler (~ 92%).

At present laser operation is close to threshold and limited by the low signal gain, the short time available for the build-up of stimulated emission and stable cavity oscillations, and by visible absorption effects. The peak laser power as measured by a fast photodiode was several kW at 518 nm. In order to obtain broad band tunability of an excimer laser system it is necessary to develop an intracell resonator with an adjustable low loss dispersive element such as a prism, grating, a narrow band filter, or by injecting tunable dye laser radiation [16]. Using an intracell resonator composed of a Littrow prism reflector and a curved output coupler we have obtained a tuning range of 30 nm for the Xe_2Cl laser. D.J. Eckstrom of Stanford Research Institute SRI recently succeeded in continuous tuning of the XeF ($C \rightarrow A$) laser over a range of 80 nm [8], which represents a major technological advance for excimer lasers which have been so far fixed wavelength laser sources.

Summary

Most electron beam lasers including the Rice University lasers described above utilize transverse electron beam excitation. Only a small fraction of the electron beam energy is actually deposited in the optically active region. Although long cathode (0.5 to 1 m) electron beam machines are commercially available these are characterized by long (> 40 nsec) pulse dura-

tions. Hence, for low gain laser systems a very promising pumping method would employ longitudinal excitation as shown in Fig. 12 which has been explored by Sandia and Wurzburg University [17 - 19]. We plan to construct such a longitudinal electron beam pumping configuration that utilizes the Rice University Physics International Pulserad 110 accelerator in the near future.

In summary, a new diatomic blue-green XeF ($C \rightarrow A$) laser has been operated centered at 486 nm with a spectral bandwidth of 12 nm and a gain coefficient of 0.05 cm^{-1}. A peak laser power of 10 kw with a pulse duration of 25 ns ($\tau_{FL} \approx 45$ ns) was obtained from electron beam excited $Ar/Xe/NF_3$ mixtures. More recently, the first triatomic Xe_2Cl laser has been operated. Laser emission centered at 518 nm with a spectral bandwidth of 20 nm and a peak power of about 5 kw in a pulse duration of 45 nsec ($\tau_F \approx 90$ ns) was obtained from an electron beam excited Ar/Xe/CCl mixture.

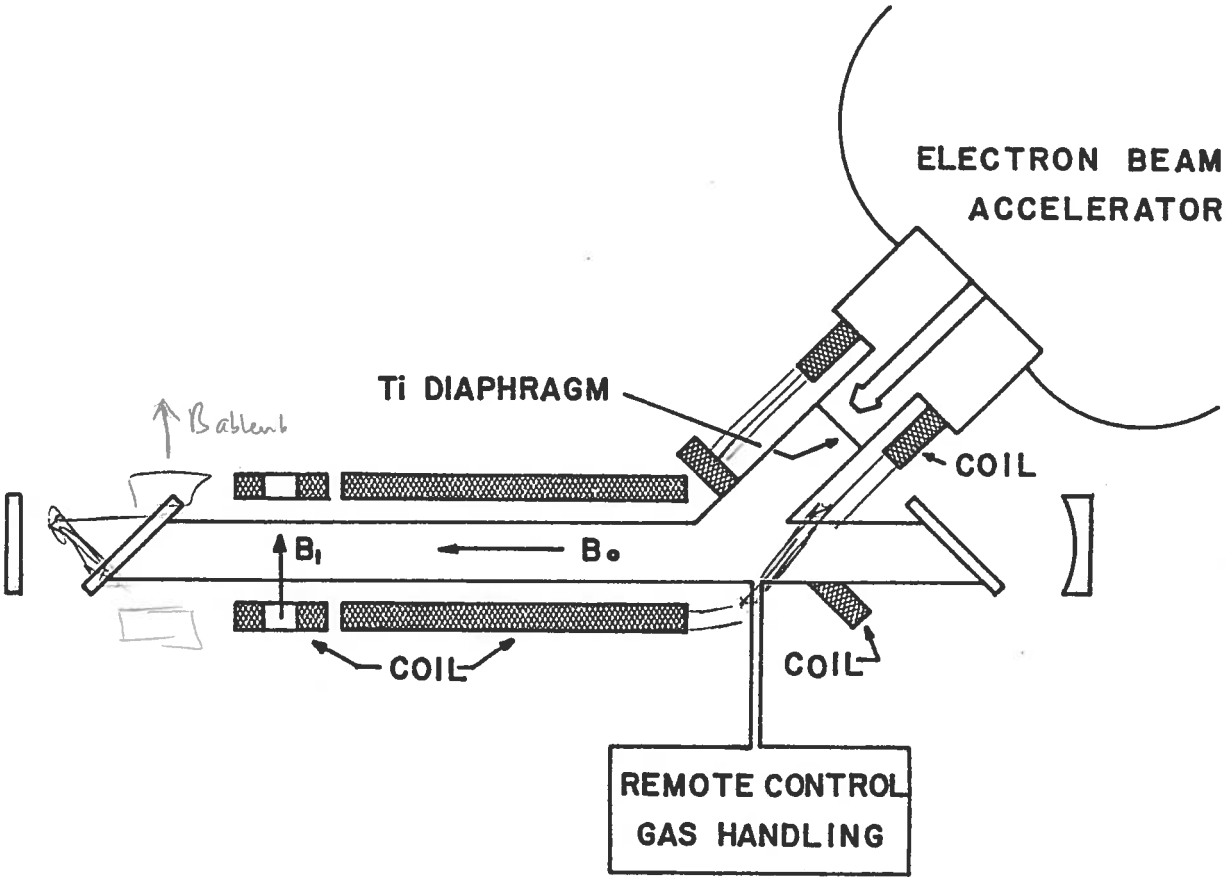

ELECTRON BEAM
ACCELERATOR
Ti DIAPHRAGM
COIL
B_1
B_o
COIL
COIL
REMOTE CONTROL
GAS HANDLING

FIG. 12

References

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