NUCLEAR-PUMPED LASERS II

Final Contract Report

by

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and

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INTRODUCTION

This final contract report will summarize research on nuclear-pumped lasers accomplished under NASA Grant NSG-1396 from March 1977 to August 1979. This research was carried out in cooperation with the nuclear-laser group at the NASA Langley Research Center, under the direction of Frank Hohl. Reactor experiments were performed at the Army Pulse Radiation Facility, Aberdeen Proving Grounds, Aberdeen, Maryland.

The two main research objectives of this contract were to demonstrate high power nuclear lasering (>1 kWatt) and demonstrate nuclear pumping with fission fragments from $^{235}$U$_6$. The progress made towards these objectives will be discussed in this report.

BASIC LASER AND GASEOUS ELECTRONICS RESEARCH

Using a high-pressure electrically-pulsed afterglow discharge in the noble gases, it has been experimentally demonstrated that the electrically-pulsed afterglow and the nuclear-pumped discharge are equivalent for the noble gas lasers. Lasing occurs at the same wavelengths and minority species concentrations for both discharges. Thus, it is possible to study noble gas lasers, optimizing the minority species concentration, before using the reactor for excitation. This result may also apply to other non-noble gas systems, but further research is needed here.
NUCLEAR PUMPED MULTIPLE PASS BOX LASER

- Polyethylene Moderator
- Back Cavity Mirror
- He-Ne Alignment Laser
- Gold or Aluminum Plane Mirror
- Fast-Burst Reactor
- Brewster Angle Window
- Dielectric Output Mirror
- Neutral Density Filter
- InAr Array Detector

Fig. 1
He-Ar Nuclear Pumped Box Laser Beam Profile

800 Torr He-Ar, 1\%Ar
20\% T Output Mirror

Laser Beam
Dielectric Output Mirror
Brewster Angle Window, 1 cm x 5 cm

Peak Laser Output (rel. units)

\begin{center}
\begin{tabular}{c c c c c c c c c c c}
InAr Channel Number & Channel Spacing \\
1 & 1.5 & 3.0 & 4.5 & 6.0 & 7.5 & 9.0 & 10.5 & 12.0 & 13.5 & 15.0 & mm \\
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Fig. 2
NUCLEAR PUMPED MULTIPLE-PATH BOX LASER

Since the power deposition per cubic centimeter is low, as compared to E-beam excitation, it is important to demonstrate scaling of nuclear laser output with increasing volumes of excited gas. Thus, a large volume multiple path box laser was constructed as shown in fig. 1. Two box lasers were constructed, box laser I was 2 cm thick while box laser II was 3 cm thick, all other dimensions were the same. Both lasers were made of stainless steel with aluminum side cover plates on which were attached polyethylene moderators. Gold plane mirrors were mounted inside the laser to reflect the laser light back and forth between the external dielectric coated laser cavity mirrors. The mirrors were aligned using a helium-neon laser. The laser radiation was detected by an InAs linear array detector which was used to resolve the laser beam profile. The beam profile of box laser I is shown in fig. 2. Note that the profile is not Gaussian indicating that the mode pattern is complex.

In fig. 3 is plotted the measured experimental results of our cylindrical and box laser power outputs. With Box Laser II a peak power of ~100 Watts has been achieved at low thermal neutron fluxes (lower than with cylindrical laser configurations) demonstrating the improved coupling of the laser gas to the reactor neutron flux.
FISSION FRAGMENT LASING OF Ar-Xe

Thermal Neutron Flux
$4.8 \times 10^{16} \text{ n/cm}^2 \cdot \text{sec}$

600 Torr Ar-3% Xe
No added UF$_6$

600 Torr Ar-3% Xe
1 Torr added UF$_6$
Fission Fragment Pumped Ar-Xe Lasing

- 3% Xe, 600 Torr Ar-Xe
- $7.7 \times 10^{16}$ n/cm$^2$-sec
- 2.65μm filter

Channel
1
2
3
4
5
6
7
8

Ar-Xe Laser Output

Neutron Pulse

100 μsec / Div

Fig. 5
FISSION FRAGMENT LASING IN Ar-Xe and He-Xe

- 3% Xe, 600 Torr Ar-Xe
- 20% Xe, 600 Torr He-Xe
- $8 \times 10^{16}$ n/cm$^2$-sec Thermal Flux

Fig. 6
Fig. 7

FISSION FRAGMENT LASING IN HE-XE
-20\% 600 torr He-Xe
-8 \times 10^{16} \text{n/cm}^2\text{-sec}

He-Xe Lasing With No Filter
Neutron Pulse
Lasing At 3.5 \mu m
In He-Xe

Laser Output (rel units)

50 \times \text{sec/div}
FISSION FRAGMENT PUMPING OF Ar-Xe-235UF₆

Research was also started in attempting to pump a noble gas laser system by fission fragments from 235UF₆ fission. The Ar-Xe (3% Xe) system was chosen from laboratory experiments where up to 5% 238UF₆ could be added to Ar-Xe(3%) before laser quenching at 2.65 µm would occur.

After repeated fillings of 235UF₆, at the reactor, a UF₅ and UF₄ solid coating covered the interior wall of the quartz laser cell. When Ar-Xe(3%) at 600 Torr was placed in the laser cell, lasing was achieved as shown in fig. 4. Since the UF₅ coating was not homogenous, the laser output is very erratic. When 1 Torr of gaseous 235UF₆ was added to the above mixture, lasing also occurred although at reduced output as shown in fig. 4. When 4 Torr of 235UF₆ was added, no lasing occurred.

Figure 5 shows the InAs linear array detector output. The beam profile is resolved and is complex in nature.

Figure 6 shows the total lasing output for two gas mixtures Ar-Xe(3%) and He-Xe(20%) using the same gold hole-coupled (2mm dia. hole) mirror set. Note that the Ar-Xe output is larger probably due to the higher stopping power of argon for fission fragments as compared to helium. Figure 7 shows that most of the lasing probably occurred at 2.027 µm since there is very little output at 3.5 µm. Although lasing did not occur with gaseous 235UF₆ in the laser cavity, if higher reflectivity mirrors are used, possibly more 235UF₆ could be added to the point where the Ar-Xe will be pumped entirely by gaseous 235UF₆.
MODELING OF NUCLEAR PUMPED LASERS

Modeling of the $^3$He-Ar nuclear pumped laser has been completed and is described in a paper by Wilson, DeYoung, and Harries. In this system charge transfer and Penning ionization of Ar by $\text{He}_2^+$ and He(met) respectively create a high density of Ar$^+$. Since the electron temperature is low, collisional radiative recombination occurs at a fast rate eventually populating the upper laser level and lasing occurs at 1.79 µm in ArI. If too high a concentration of Ar is used in He-Ar, then formation of Ar$_2^+$ occurs rapidly, which upon dissociative recombination, populates the lower laser level quenching lasing. All the noble gas laser systems (except 3.5 µm XeI) appear to operate as described above.

CONCLUSION

During this contract period progress has been made toward the goal of understanding the basic processes active in noble-gas nuclear-pumped lasers. We have demonstrated that recombination plays the dominant role in pumping such lasers. A peak power output from $^3$He-Ar of ~100 Watts has been achieved with a large volume multiple path box laser. Also, Ar-(3%)Xe has been nuclear pumped by fission fragments from UF$_5$ and UF$_4$. One Torr of $^{235}$UF$_6$ gas could be added to the laser before quenching occurred.

Additional information and experimental results are discussed in the papers shown on the "List of Journal Articles and Conference Presentations Under Contract NSG-1396."
List of Journal Articles and Conferences  
Presentations under Contract NSG-1396


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LaRC Results on
Nuclear Pumped Noble Gas Lasers

R. J. De Young

All the noble gas except He have now lased with nuclear excitation. The noble gas nuclear lasers are the best understood systems thus far and lasing has been achieved with both $^3$He and fission fragment pumping. The noble gas nuclear lasing transitions are shown in figure 1. Nuclear pumping of neon (not shown) at 632.8 nm has been achieved by the nuclear laser group of the University of Florida.[1] As shown in figure 1, lasing has occurred for wavelengths of 1.79 µm to 3.65 µm, at pressures from 400 Torr to 4 Atm, and for noble gas concentration from 0.01% to 30%.[2] It is the purpose of this paper to review the recent experimental and theoretical results obtained for noble gas systems.

In figure 2 the power deposited in either $^3$He or $^{235}$UF$_6$ is shown for the $^3$He(n,p)$^3$H and $^{235}$UF$_6$(n,ff)FF reaction respectively assuming no loss of charged particles to the container walls. From the figure it can be seen that nuclear pumping cannot match the power deposited from typical E-beam machines; thus potential nuclear laser systems must have
reasonable gain at low power deposition rates. This is exactly the case for the He noble gas mixture lasers.

The gain of a laser medium can be written as:

\[ \gamma(v) = \frac{\lambda^2}{8\pi \tau_{\text{spon}}} \left[ (N_2 - N_1(g_2/g_1)) g(v) \right] \]  
\hspace{1cm} (1)

where \( g(v) = \frac{(\Delta \nu/2\pi)}{(v - v_0)^2 + (\Delta \nu/2)^2} \)  
\hspace{1cm} (2)

is the Lorentzian lineshape function. At line center the gain becomes

\[ \gamma_0 = \frac{\lambda^2}{4\pi^2 \tau_{\text{spon}}} \left[ N_2 - (g_2/g_1) N_1 \right] / \Delta \nu \]  
\hspace{1cm} (3)

where \( \Delta \) is the full width at half maximum of the laser transition.

By noting eq. 3 we can understand why the noble gas lasers are so easily pumped under nuclear excitation. First, all the lasing wavelengths are in the infrared maximizing the gain by \( \lambda^2 \); second, the inversion density \( [(N_2 - N_1(g_2/g_1))] \) is easily maintained since for all the transitions the upper laser level lifetime is longer than the lower laser level lifetime and minimal pumping of the lower laser level occurs. Third, \( \Delta \nu \) for all noble gas laser transitions is small (as compared to excimer systems for example). Fourth, \( \tau_{\text{spon}} \) the radiative lifetime is short but not shorter than the lifetime of the lower laser level. Thus, it is clear why the noble gas systems can be easily pumped with nuclear or electrical excitation.
Noble Gas Laser Excitation Mechanisms

Noting again figure 1, it is observed that the noble gases form a Penning mixture with He metastables. Also, charge transfer from He$_2^+$ can efficiently ionize the minority Ar, Kr, or Xe lasing species. In either case a high density of noble gas atomic ions is produced. Calculations were undertaken for the $^3$He-Ar nuclear laser which are summarized in the "generalized energy block diagram as shown in figure 3 for $^3$He-Ar but are equally valid for He-Kr, or He-Xe.[3] Here the efficient production, by charge transfer and Penning ionization, of Ar$^+$ is shown. Loss of Ar$^+$ occurs from three body association to form Ar$_2^+$ or from collisional-radiative recombination which after radiative cascade eventually pumps the upper laser level of the 1.79 μm Ar transition. The lower laser level is pumped predominately by dissociative recombination of Ar$_2^+$; thus, the minority gas species must be kept low to retard formation of Ar$_2^+$, Kr$_2^+$, or Xe$_2^+$. The favorable lifetimes of the upper and lower laser levels ensure a population inversion if pumping from Ar$_2^+$ dissociative recombination is kept small.

Figure 4 shows a schematic diagram of the Ar$^+$ collisional-radiative model used to calculate the flow of energy after recombination. The atomic states within the Saha region are in equilibrium with the free electrons and the states there are assumed to be completely collision dominated. States below the Saha region are assumed to be completely radiative dominated, and
the radiative energy flow into the upper laser level is calculated using the Ar transitions probabilities. Using this model, reasonable gain and power output for the Ar 1.79 μm laser transition were calculated and compared favorably to experimental measurements.

Figure 5 is a comparison between calculated and experimental laser inversion density vs. argon concentration. The results are quite good and should be equally adaptable to the He-Kr and He-Xe systems.

Large Volume Noble Gas Nuclear Pumped Lasers

As noted in figure 2, the pumping power densities for nuclear lasers are small; thus, in order to achieve high power outputs, it is necessary to use large volumes of excited gas. This can be done easily with nuclear pumping since neutrons can penetrate deeply into a high pressure gaseous medium.

Figure 6 shows a nuclear-pumped multiple pass box laser presently used for large volume lasing experiments with noble gases.[4] The laser frame is made of stainless steel with aluminum cover plates on which a polyethylene moderator is attached. Gold plane mirrors are aligned internally to reflect the laser beam back and forth through the excited gaseous medium. External dielectric coated mirrors form an optical cavity for laser experiments, alternatively, this configuration could be used as an amplifier by using an external oscillator. Lasing is detected by a multiple element InAs array.
The volume here is 4800 cm$^3$ as compared to 200 cm$^3$ for the cylindrical noble gas nuclear lasers. In figure 7 the box laser and cylindrical laser results are compared and show the advantage of the multiple pass large volume systems. Approximately 100 watts peak power has been achieved from box laser II, thus far, from $^3$He-$^4$Ar lasing at 1.79 $\mu$m. In figure 8 another plot is shown of the comparison between box and cylindrical laser output as a function of time indicating the progress made with noble gas systems.

**Fission Fragment Pumped Ar-Xe Lasing**

Experiments were undertaken on the Ar-Xe system with $^{235}$UF$_6$ fission fragment pumping.[5] This system was found to lase well in the laboratory at 2.65 $\mu$m in Xe with up to 5% added UF$_6$. Since Ar has a stopping distance of 7 cm compared to a He distance of 28 cm for fission fragments, it was thought that the Ar-Xe system would be the most ideal candidate for noble gas fission fragment pumping.

Initial experiments at the reactor used 600 Torr Ar-3% Xe (3% Xe was found to be the optimum concentration for high-pressure electrically pulsed lasing at 2.65 $\mu$m) with from 5 to 20% $^{235}$UF$_6$. No lasing was observed. A pressure transducer was attached to the laser cell with a response time fast enough to detect the gas pressure pulse during the reactor neutron pulse. The output of the pressure transducer is shown in figure 9. Also
shown is the thermal neutron pulse. The pressure pulse is made from shock waves created by the thermal neutron pulse, as shown at the bottom of figure 9. As the $^{235}\text{UF}_6$ concentration was lowered in Ar-Xe, it was noted that the pressure pulse did not change. Thus, Ar-Xe was then pulsed with no added $^{235}\text{UF}_6$ and lasing was observed from the Ar-Xe mixture at 2.65 $\mu$m. The origin of the excitation energy is shown in figure 10. After many $^{235}\text{UF}_6$ fillings, UF$_5$, UF$_4$, etc., was deposited on the inner laser cell wall, thus creating a source of fission fragments. No more than 1 Torr of $^{235}\text{UF}_6$ could be added to the Ar-Xe before laser quenching would result. Since the fissionable coating was not homogeneously deposited on the wall, uneven excitation of the gas medium would result. This was observed as shown in figure 11 where a typical Ar-Xe laser output is compared to the thermal neutron pulse. Also shown is the laser output at 2.027 $\mu$m, which was considerably lower than the 2.65 $\mu$m laser output. The erratic nature of the excitation source is readily observed.

This is the first laser system to be pumped with fission fragments from $^{235}\text{UF}_6$ and with a higher Q optical cavity it may be possible to add sufficient gaseous $^{235}\text{UF}_6$ to actually pump Ar-Xe directly.

Conclusions

It has been shown that the noble gas lasers are among the easiest systems to pump by nuclear excitation and as a result all
the noble gases except He have lased under nuclear excitation. The noble gas systems are not ideal for high-power applications but they do give valuable insight into the operation and pumping mechanisms associated with nuclear lasers. At present, the Ar-Xe system is the best noble gas candidate for $^{235}\text{UF}_6$ pumping. It appears that the quenching of Ar-Xe lasing is a result of the fluorine and not the uranium or fission fragments themselves. Thus, to achieve lasing with UF$_6$, a fluorine compatible system must be found.
References


Energy Level Diagram of $^3$He DNP Lasers

- $^3\text{He}$
  - $2^1S$
  - $2^3S$

- Ar$^+$
  - $15.755$
  - $3d(3/2)^0$
  - $4p(3/2)^{0.5}p(1/2)^{0.5}$
  - $4S(3/2)$

- Kr$^+$
  - $13.996$
  - $3d(1/2)$
  - $4d(1/2)^0$
  - $5p(3/2)$

- Xe$^+$
  - $12.127$
  - $5d(3/2)^0$
  - $5s(3/2)^0$
  - $6p(3/2)^0$

Laser Wavelengths:
- $1.792\mu m$
- $3.65\mu m$
- $3.5\mu m$

Fig. 1
POWER DEPOSITED vs. NUCLEAR AND E- BEAM EXCITATION

Fig. 2
Collisional Processes in $^3$He-Ar DNP Laser

Fig. 3
STATES TREATED IN ARGON RECOMBINATION MODEL

Ar$^+$

Saha Region (states are completely collision dominated)

Radiative Region (states are completely radiative dominated)

upper laser level

1.79 m Laser

lower laser level

Fig. 4
Comparison of calculated and experimental $^3$He-Ar laser output

- $P = 0.54$ atm
- $f_0 = 6 \times 10^{16}$ n/cm$^2$/s
- Cell i.d. = 2 cm

Fig. 5
PEAK LASER OUTPUT FROM CYLINDRICAL AND BOX LASERS

- **Box Laser I**
  - $\text{He-0.5\%Xe, 2.027\mu m}$
  - $2.3 \times 10^{16}$ Av. Flux
  - 20\%T Output Mirror
  - Cylindrical Laser

- **Box Laser II**
  - $\text{He-2\%Ar}$
  - $2.75 \times 10^{16}$ flux
  - Box Laser II

- **3\text{He-l\%Ar, 1.79\mu m}**
  - $6 \times 10^{16}$ Av. Flux
  - 20\%T Output Mirror
  - Box Laser I

- **3\text{He-l\%Ar, 1.79\mu m}**
  - $1 \times 10^{17}$ Av. Flux
  - 1\%T Output Mirror
  - Cylindrical Laser

**Fig. 7**
PROGRESS WITH NUCLEAR PUMPED LASERS

BOX LASER I

BOX LASER II

CYLINDRICAL LASER

1976
1977
1978
1979
1980

DATE

Fig. 8
FISSION FRAGMENT PUMPED Ar-Xe PRESSURE PULSE

- 600Torr, Ar-3% Xe

Thermal Neutron Pulse
$8.6 \times 10^{16} \text{n/cm}^2\text{-sec}$

Pressure Transducer
Output

45lbs

0.5 millisec/div

Fig. 9
Ar-Xe ALMOST \( ^{235} \text{UF}_6 \) PUMPED NUCLEAR LASER
- 600 Torr, Ar-3% Xe
- \( 8 \times 10^6 \text{ n/cm}^2 \text{-sec} \)

Quartz Laser Cell

\( ^{235} \text{UF}_6 \) Coating

Fission Fragments

Thermal Neutron Flux

Fig. 10
FISSION FRAGMENT LASING IN Ar-Xe

- 3% Xe, 600 Torr Ar-Xe
- $7.7 \times 10^{16}$ n/cm²·sec

Lasers at 2.65 μm in Ar-Xe
Lasers at 2.027 μm in Ar-Xe

Neutron Pulse

Laser Output (rel. units)

50 μsec/div

Fig. II