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ADVANCED METHODS FOR NUCLEAR REACTOR-GAS LASER COUPLING

For the Period

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by

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and

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1. Introduction

During this abbreviated contract period (8 mos.), the following work was undertaken and/or completed:

- a) Radiation enhancement studies of a pulsed He-Ne Laser in the pressure range of 10-80 Torr operating on both 3.39 and 1.15 μ .
- b) Radiation initiation and enhancement of chemical lasers.
- c) Optical spectra measurements were extended to high-pressure helium mixtures irradiated by alpha particles.
- d) Bench tests of a pulsed Ne-O₂ laser were essentially completed.
- e) Bench tests of probe methods for measuring electron energy distribution in lasers with an external ionization source were initiated.
- f) Enhancement of CO₂ laser at low levels of irradiation.

Each of these areas is described in more detail below.

II. Enhancement of a Pulsed He-Ne Laser

Earlier studies showed that nuclear radiation could be used to enhance the output from a CO₂ laser operating via electrical discharge. In the present contract period similar experiments were undertaken with a pulsed He-Ne laser operating at 3.39 μ and 1.15 μ .

Prior to these studies, some theories suggested that radiation would harm rather than enhance He-Ne operation. However, as described in more detail below, a significant enhancement was found. Aside from direct practical application, this result, then should be significant relative to sorting out appropriate theories about radiation effects.

Some detailed notes written at the time of these experiments are reproduced below.

During March and April, 1973, studies were undertaken of the possible enhancement of an electrically pulsed He-Ne laser during irradiation in the Illinois TRIGA reactor.

In all these experiments, the gas pressure was 28 Torr, 9:1 He-Ne. The laser cavity was 0.9 cm i.d. and 87 cm in length. One cm lengths of aluminum tubing coated on the inside with B-10 were placed in the laser tube. Alternate aluminum tubes and glass spacers were used along the length of the cavity. The laser was operated electrically from a pulse modulation circuit, in which the voltage and pulse repetition rates could be varied. The laser operated on the $3.39\mu\text{m}$ line in He-Ne using one dielectric and one gold coated mirror. A IAU-600 Philo detector was used to observe the emitted light, which was then displayed on a Tektronic oscilloscope and photographed.

The first experiments at the reactor used \$3 neutron pulses to excite the laser while it was operating electrically at a repetition rate of 100 pps, voltage of 11 kV max and current of 300 amps max. During the neutron pulse, a 20% increase in laser output was observed. Just after the neutron pulse the laser output (and current) would go to zero and remain there until approximately 15 minutes later, then they would approach the original output. The voltage remained constant throughout the neutron pulse. A \$2 neutron pulse was found to produce a 26% increase in laser output under the same conditions as above.

The pulse modulation circuit was changed, incorporating a new pulse forming network and pulse transformer. The voltage across the laser was now 15 kV max. and the current at 200 pps was 7 amps peak. Under the above conditions the laser was placed in the reactor and square waved to 750 Kw. The laser output experienced a 100% increase while the current did not

experience any change but remained at 7 amps peak. Shortly after each neutron pulse or square wave the laser output went to zero and did not return to normal until approximately 10-15 minutes later. (More recent experiments have shown that this effect is due to impurities from the boron coating which are driven into the gas during irradiation. It is possible to overcome this problem with careful treatment and bake out of the tubes just prior to pulsing.)

A steady-state neutron flux was used to excite the laser while operating electrically as before. No laser gain was noted until 15 Kw where a slight gain was found. As the power level was increased to 150 Kw, the output first increased slightly then decreased rapidly to zero. Some 10 minutes later the laser pulse returned to its original level under electrical excitation.

The amount of electrical energy deposited in the gas was found to be 23 watts, the amount due to nuclear excitation was calculated to be about 7.4 mwatts for 28 Torr He-Ne. The laser output under these conditions was .75 mwatts.

During May, 1973 additional studies were undertaken with the same He-Ne laser, but at a higher gas pressure of 40 ton. Experiments were run using pulses from \$1.17 to \$3.60. The following comments summarize the results of these runs and relate them to the previous observations.

The first pulse, \$3.60, was an attempt at direct pumping. With the vertical at 0.1 v/div., no output was seen. A series of \$3 pulses were run to find their effect on laser output, current, and voltage. An increase of 22% in 3.39 μ laser output was noted 13 msec before the peak of the neutron pulse. The laser output steadily decreased throughout the neutron pulse once past the peak laser pulse but did not go to zero.

The current was rastered throughout the neutron pulse. The trend indicates that if the laser pulse had been rastered further, greater

enhancement would have been noted. Direct pumping was attempted at \$1.17 with no results.

The current was rastered throughout the neutron pulse and again no change with or without nuclear excitation was noted.

A summary of these results then is:

- a) Using \$3 pulse an enhancement of 22% was found, before a 20% increase was found.
- b) Greater enhancement was found for \$1.17 pulses, 290% before 100% was observed.
- c) No change in current or voltage was found while pulsing.
- d) Direct pumping on the $3.39\mu\text{m}$ transition produced no results.
- e) Extreme care must be made in preparing the laser for irradiation, the laser bore walls must be outgased with heater tapes and a diffusion pump just prior to the experiments.

In early June a series of experiments were undertaken with the same laser operating on the 115μ line. However, an impurity problem was again encountered and the results, while encouraging, were not reproducible. Thus it is planned to repeat these experiments in the near future.

III. Radiation Effects - Chemical Laser

In our previous proposal, we indicated the reasons why the nuclear initiated chemical laser appeared attractive. In spite of the optimism expressed there, we felt that it was necessary to provide an experimental system with maximum flexibility. Accordingly, we have started our work with the TEA laser design shown in Figure 1.

The anode of the laser is an aluminum rod covered with a Boron-10 coating to provide a flux of alpha particles and lithium ions upon irradiation with neutrons. The TEA geometry is particularly convenient for such experiments since one can electrically pulse the laser simultaneously with the irradiation provided by the reactor. In this manner, we can ascertain the effects of the external source of ionization on the performance of the chemical laser.

We have started our chemical laser experiments with the methane and sulfur hexafluoride mixture of gases even though this appears to be far less attractive than the $\{BF_3 \text{ or } UF_6\}; D_2; CO_2$ system. The $CH_4:SF_6$ mixture is very easy to handle and thus considerably simplifies the experimental apparatus. Aside from that consideration, the laser itself is a very interesting and practical device.

The results have been somewhat contradictory at this stage. One particular run was very encouraging as exemplified by the data shown in Figure 2. In the first case, 2a, the laser was pulsed without any radiation, whereas the second is with reactor pulsed to a power level of $\sim 2.00 - 250 \text{ Mw}$ corresponding to a neutron flux of $6 \cdot 10^{14} \text{ n/cm}^2/\text{sec}$.

Fig. 1. TEA laser Design

Fig. 2. Enhancement of HF chemical laser.

A. Shows the normal laser output before the reactor was pulsed.

B. Shows the output during the peak of a 2.00 pulse.

SF_6 CH_4 = 6:1 TOTAL pressure = 120 Torr. Electrical Pulse 15 Kv.

Notice that there is a large increase in the energy of laser output for this case. However, this may just be due to the change in coupling of the optical cavity since we were using a hole-coupled system for this series of experiments. To further clarify this question, the output mirror was replaced by an uncoated piece of Irtran IV. Here the optical cavity has the same coupling coefficient for all modes and thus the result could be interpreted in a much simpler manner.

But, in this case, lasering ceased during the reactor pulse, and more significantly, it would not lase after the reactor was turned off! The gas pressure had to be lowered or a new gas fill added before lasing could be achieved by electrical excitation. This was also true to some extent with the hole-coupled system described in Figure 2. In that case, the laser output was enhanced significantly during the reactor pulse, but was seriously degraded after the reactor was shut-down.

It is too early to state definitely why we achieved contradictory results with the nuclear radiation depending upon the type of optical cavity. The enhancement may be due to a shift in the laser mode pattern so as to couple more optical signal to the detector. On the other hand, the quenching of the laser radiation becomes more significant when it is coupled with the fact that a static fill of gas can be electrically pulsed 40-50-times or more before enough HF is formed to degrade its performance. Yet the reactor does this in just one pulse.

The key point here is that the reactor pulse that we have been using is typically 0.1 sec long, whereas the electrical pulses are $\sim 5 \mu\text{sec}$ long. Consequently it's not surprising that one long reactor

pulse can form enough HF (in the ground vibrational state) to prevent lasing on the $v = 1$ to $v = 0$ transition, when the system is excited electrically.

But if this line of reasoning is correct; i.e., the quenching of the laser is due to the HF being formed by the reactor, then this negative result is very encouraging. This would mean that we need a short intense neutron pulse to achieve a nuclear initiated chemical laser. We still have considerable more leeway here at Illinois to perform additional experiments and there are much faster reactors and neutron sources available throughout the country.

Furthermore, the $\text{BF}_3:\text{D}_2:\text{CO}_2$ nuclear initiated system should provide even greater leeway. In this case, the vibrational energy of the chemical reaction should funnel into the CO_2 (001) state with considerably less relaxation to the ground state.

IV. Optical Spectra of High-Pressure Helium Mixtures

Optical studies of helium in some mixtures irradiated in the polonium alpha facility were described in prior progress reports. The objective of this work is to obtain a basic understanding of the plasma kinetics which will help plan future incore laser experiments.

During the present contract period, studies of helium at 100 to 750 Torr with 10 ppm to 10% of N_2 , O_2 , H_2 and Ne additives. The results are described in C00-2007-37 which is reproduced below.

OPTICAL SPECTRA OF HIGH-PRESSURE HELIUM MIXTURES IRRADIATED BY ALPHA PARTICLES

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Measurements of optical lines from atomic and molecular states created by alpha-particle irradiation of He mixtures at 100 to 750 Torr are reported vs. the partial pressure of N_2 , O_2 , H_2 , and Ne additives (10 ppm to 10%) and applied electric field strength. While gross spectra of the impurity species have been reported⁽¹⁾, the present study is the first to include the helium spectra (3000 to 9000-Å). Also, except for a study of He-CO by Kunz et al.⁽²⁾, earlier work was not designed to probe the mixture kinetics.

The equipment and earlier high purity helium results are described elsewhere⁽³⁻⁶⁾. The mixtures were made using an inverse McLeod gauge measurement of the partial pressures⁽⁷⁾.

Figure 1 shows results for 740 Torr total pressure. (At lower pressure line intensities are essentially linear in total gas pressure for any given partial pressure.) For low concentrations the first negative N_2^+ bands (originating in the $B^2\Sigma_u^+$ state) increase linearly with the partial pressure of N_2 . Their origin is either from charge exchange with He_2^+ or resonant transfer (Jesse Ionization)⁽⁷⁾ from the He_2 molecular metastable. As the N_2 concentration is increased, the 6400 and 6594-Å bands of He_2 disappear, indicating quenching of the helium 2P atomic states by N_2 . At higher concentrations, the first negative system is also quenched by $N_2^{++} - N_2$ collisions (possibly forming N_4^+), causing the emission to peak at 200 ppm. For $N_2 > 1000$ ppm the electron energy distribution⁽⁸⁾ is

altered as is shown by the decreased He 7065-Å intensity and the increased N₂ second positive emission. The latter increased linearly until N₂(2P)-N₂ collisions depopulate this state resulting in a maximum 2P intensity at about 3% N₂.

The results for hydrogen and oxygen were similar to those obtained for N₂. Atomic oxygen emission (e.g., 7774 and 8446-Å) was also detected.

The intense 2p-1s lines from 5400 to 8000-Å were found to increase linearly in Ne concentration similar to the N₂ second positive bands. As other emissions in the 3000 to 9000-Å region were very weak, direct electron excitation at high pressures appears to dominate as opposed to resonant transfer from He atomic or molecular metastables. This view is supported by differences between the electric field data for Ne and for molecular gases.

These results indicate that in high-pressure noble gases a significant fraction of the energy of ionizing radiation can be efficiently transferred to a few levels of a low concentration additive gas via the noble gas molecular metastable and ion. This is in general agreement with a model for He proposed by Hurst, et al. ⁽⁹⁾ and explored in mixtures of Ar, Kr, and Xe by Gedanken, et al. ⁽¹⁰⁾ However, they treat only resonant transfer from the molecular metastable, whereas the present data shows that above 1000 ppm additional mechanisms must be incorporated.

A variety of practical applications are suggested from this data. For example, it appears that population inversion in high-pressure He-N₂ using nuclear or e-beam pumping may be possible in both the second positive ⁽¹¹⁾ and in the first negative systems. Similarly, lasing in high pressure He-O₂ mixtures on 8446-Å is suggested. It also appears that a helium buffer might allow operation of the 5401-Å Ne laser at high

pressure. Relative to detector applications, it appears that either a 9:1 He-Ne mixture with a high-gain extended red tube, or a He-N₂ mixture with a high-gain UV tube, is well suited for gas scintillation counters.

References:

1. See the review published by the authors in Ref. 3.
2. C. Kunz, S. Dondes, and P. Harteck, "A Spectroscopic Study of the Alpha-Ray-Induced Luminescence in Gases; the Helium-Carbon Monoxide System," *Radiation Research*, 41, 288 (1970).
3. P. E. Thiess and G. H. Miley, "Spectroscopic and Probe Measurements of Excited State and Electron Densities in Radiation Induced Plasmas," *Proc., 2nd Symp. on Uranium Plasmas*, pp. 90-97, AIAA, Atlanta (November 1971).
4. P. E. Thiess and G. H. Miley, "A High-Vacuum Pure-Gas Alpha Irradiation Facility," to be published in *Trans. Am. Nucl. Soc.*, 16, No. 1 (June 1973).
5. P. E. Thiess, T. A. Visel, and P. A. Kwitkowski, "A Digital Photometer for Studies of Spectra Produced by Ionizing Radiation," *Trans. Am. Nucl. Soc.*, 15, 707 (1962).
6. P. E. Thiess and G. H. Miley, "Optical Spectra of High-Pressure Helium Irradiated by Alpha Particles," *Trans. Am. Nucl. Soc.*, 15, 706 (1972).
7. W. P. Jesse and J. Sadauskis, "Ionization by Alpha Particles in Mixtures of Gases," *Phys. Rev.* 100, 1755 (1955).
8. M. A. Uman, *Phys. Rev* 133, A 1266 (1964); 123, 399 (1961); and 120, 1542 (1960).
9. G. S. Hurst, D. M. Bartell and E. B. Wagner, "Model for VUV Emission and Energy Pathways Following Proton Excitation of Helium," to be published, *Bul. Am. Phys. Soc.*, March 1973.
10. A. Gedanken, J. Jortner, B. Raz, and A. Szöke, "Electronic Energy Transfer Phenomena in Rare Gases," *J. Chem. Phys.* 57, 3456 (1972).
11. T. Kobayasi, M. Takemura, H. Shimizu, and H. Inaba, "TEA UV Nitrogen Laser and Its Application to High-Sensitive Remote Pulsed-Raman Spectroscopy of Atmospheric Pollutants," *IEEE J. Quantum El.*, QE-8, 579 (1972).

Figure Legends

Fig. 1 Observed Line or Band Intensities of Excited Species in Helium Mixtures Detected by Single Photon Counting.⁽³⁾ [Intensities should be multiplied by the scale factors where indicated. The He_2 6400-Å band is shown for the case of an applied field yielding 1 μ a collected current which allowed more accurate measurements to be made.⁽³⁾ The resolution used was 22 to 26-Å FWHM which was sufficient to prevent overlap with other spectral lines⁽⁵⁾ (e.g., He 5876 and 3889).]

OBSERVED LINE OR BAND HEAD INTENSITY, cpm

V. Bench Tests of Ne-O₂ Laser

As explained in the companion proposed technical program, this laser is considered to be a prime candidate for direct pumping. A laser with appropriate boron lining has been constructed and bench tests undertaken. Little experience with this system is reported in the literature, so considerable work is necessary to optimize operation. Lasing has been obtained, but with fairly low power outputs. This work is continuing and incore experiments are planned for the near future.

VI. Probe Study of Electron Energy Distributions.

Prior work on CO_2 lasers had indicated increased efficiency and power when irradiated by the neutron flux. Although some of the enhancement could be attributed to the effects of external ionization, there also appeared to be a fundamental change in the excitation rate of the lasering levels by the electrons. For instance, increased power was obtained from a CO_2 laser even though the voltage and the current were the same as without radiation.

Inasmuch as the voltage across a discharge is a measure of the average electron energy, and the current is a measure of the number density of electrons, the results are quite puzzling. They were tentatively explained by a change in shape of the electron energy distribution. Previous work by Ganley with double probes tended to give some credence to the hypothesis, but a detailed study was not completed.

We are now attempting to do this by the use of a single Langmuir probe which, in principle, measures the entire electron energy distribution (as opposed to the double probe sampling only the high energy tail). Inasmuch as most of the effects of the nuclear radiation is to provide a source of ionization which is independent of the plasma electrons, any other source of external ionization will provide equally valid data. Accordingly, we have constructed a simple gas discharge for these probe studies and are using the beam electrons from another discharge to provide the external ionization.

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The set-up is shown in Figure 3. The positive column of the primary discharge path is the one to be studied. The length of the auxiliary discharge was chosen so that the anode well is within the negative glow. Consequently, the electrons accelerated by the cathode fall potential are ballistic in nature and can travel through the mesh anode and into the positive column of the primary discharge. Thus, we have an easily controlled external source of ionization which is independent of the process occurring in the main discharge.

The experimental apparatus has been constructed and the associated electronic equipment assembled. Preliminary results have been obtained, but it is too early for detailed analysis.

Fig. 3. Discharge Tube for Probe Studies.

Fig. 4. Enhancement of CO_2 laser at low reactor power levels.

- A. Illustrates a typical laser pulse with the reactor shut-off.
- B. The laser output with reactor at 50 kW.

Note the vertical scale change between A and B. There is a factor of ~ 27 in enhancement of laser energy output.

$p = 600$ Torr of $\text{He:N}_2:\text{CO}_2$ (8:1:1), 15 kV electrical pulse.

VII. Enhancement of CO₂ lasers at low level of radiation

It is a simple matter to interchange gases in the course of our work on chemical lasers. Indeed, the use of the CO₂:N₂:He mixture facilitates the alignment of the laser-detector combination.

We have found that very low levels of nuclear radiation significantly enhances the pulsed CO₂ laser output. For instance, Figure 4a shows the laser output without any radiation, whereas 4b illustrates the laser output with the reactor power of only 50 Kw. The enhancement in laser energy is sizeable, being a factor of 27 or more. The electrical energy input stays the same or decreases in such a circumstance. Furthermore, it appears that the laser may be run in a quasi-CW manner at these high pressures.

These results have some very significant technological implications. They suggest the possible use of an isotope for pre-ionization in high pressure lasers (the enhancement improves with pressure). However, more experiments are needed before a definite statement can be made.