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UV GAS LASER STUDIES

M. L. Bhaumik, et al

Northrop Research and Technology Center

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13. ABSTRACT

Laser oscillations have been observed at two wavelengths spaced 5Å apart and centered at 1730 ±10Å from high pressure xenon gas under pulse excitation by an 80 nanosecond, 0.5 MeV E-beam. The spectral line narrowing from 150Å to 2.5Å with over a thousandfold increase in intensity, combined with the well defined mode pattern burned into the mirrors, provides conclusive evidence that a molecular xenon laser has been achieved. The results show that laser oscillations in the vacuum ultraviolet can be achieved in spite of the effects of photoionization and also that a whole new class of lasers may be possible based on molecular association. The laser doublet, tentatively identified as due to transitions from $^1\Sigma_u^+$ and $^3\Sigma_u^+$ to the repulsive ground state $^1\Sigma_g^+$, may help to provide quantitative data for the xenon molecular energy levels.

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G. Hasserjian

G. Hasserjian, Manager
Laser Technology Laboratories

FOREWORD

This special report describes the experimentally observed laser oscillations in the vacuum ultraviolet obtained from high pressure xenon gas under E-beam excitation. The experimental work was performed jointly with the Los Alamos Scientific Laboratory, Los Alamos, New Mexico, and Maxwell Laboratories, Inc., San Diego, California. A preliminary report of this work was presented in a postdeadline paper at the Optical Society of America meeting, Denver, Colorado, March 1973.

TABLE OF CONTENTS

1.0	SUMMARY	1
2.0	INTRODUCTION	3
3.0	THE EXPERIMENTAL SETUP AND PROCEDURES	4
4.0	RESULTS OF THE EXPERIMENTS	7
5.0	DISCUSSION OF THE RESULTS	11
6.0	IDENTIFICATION OF THE LASER DOUBLET	15
7.0	REFERENCES	19

1.0 SUMMARY

The Northrop Research and Technology Center, Laser Technology Laboratories, has been under contract with ARPA/ONR since April 1972 to demonstrate the feasibility of achieving stimulated emission in the vacuum ultraviolet (VUV), using molecular xenon gas excited with a relativistic electron beam. Initial experiments were performed with an E-beam from the Northrop Febetron 705. The results of these experiments, reported in NRTC Report 72-14R dated November 1972, indicated that only incipient lasing was achieved probably due to the small (~1 cm) gain length achievable by the Febetron electron beam. Consequently, a gun with a larger beam cross section was identified to continue the experiments. The results of these experiments are described in this report. This E-beam gun has a peak current of 150 kA at 0.5 MeV, a pulse width of 80 nsec, and is the Maxwell Laboratories POCO electron accelerator. The circular E-beam was shaped by a 15 kilogauss magnetic field into a rectangular cross section of 9 cm x 3.5 cm. The average current density inside the laser cell was ~1200 amp/cm^2 providing about 1200 joules of input energy.

The E-beam entered the optical cavity transversely. The laser resonator consisted of a 5m total reflector on a pyrex substrate and a plane output mirror on a MgF_2 substrate spaced 12 cm apart. The mirrors were prepared by aluminum coating with a MgF_2 protective overcoat. The radiation from the output mirror was directed through an evacuated chamber to a SPEX 3/4 meter spectrograph (model 1500) equipped for the VUV region. The E-beam excited xenon gas at a pressure of 12 atm, with the mirrors removed, had a ~150Å wide spontaneous emission spectrum centered around 1730Å. When the mirrors were installed and the optical cavity aligned, the emission narrowed down to two 2.5Å wide lines separated by 5Å at a center wavelength of 1730 ±10Å. The intensity of the above lines was more than 1000 times the intensity of the spontaneous emission band.

The spectral line narrowing was accompanied by a well defined mode pattern burned into the mirrors. The simultaneous observation of line narrowing, increase in spectral intensity and the mode pattern were repeatable in every shot for six shots. When the resonator was misaligned, or one of the mirrors was taken out, there was neither a mirror burn spot nor any line narrowing. This demonstrates that the observed mirror burn spot attendant with line narrowing must occur due to laser oscillations and not due to damage by scattered electrons, pressure waves or superradiant emission. The laser energy was estimated from the heat necessary to evaporate the mirror coatings to be approximately 1J. This corresponds to energy extraction of $\sim 0.1 J/cm^3$ with $\sim 0.3\%$ efficiency.

The observed laser doublet is tentatively identified to be due to the transitions from the upper electronic states $^1\Sigma_u^+$ and $^3\Sigma_u^+$ to the repulsive ground state $^1\Sigma_g^+$ of the xenon molecule. The doublet signifies that the laser builds up independently on the two transitions, indicating that the cross relaxation between the singlet and the triplet upper states during the pulse is insufficient. However, an examination of the relative intensities of the two lines clearly shows at least partial cross relaxation. Such a higher rate for the singlet-triplet intersystem crossing may be possible due to the strong spin-orbit coupling and the highly mixed wave functions.

These results leave little doubt that laser emission was observed from molecular xenon in the VUV region, in spite of competition from photoionization. The xenon laser doublet also provides a means for the first time to unravel some of the details of the excited states of the xenon molecule. The results of these experiments suggest that a new class of lasers should be possible based on similar molecular associations with output wavelengths from IR to VUV.

2.0 INTRODUCTION

The objective of the present program is to carry out a thorough investigation of the potentially new class of lasers, called the "molecular association" lasers. Since these "associative" molecules have a repulsive ground state, any population of the stable upper state leads to 100% inversion. This combined with their high quantum efficiency indicates the possibility of achieving a high overall laser efficiency. Furthermore, radiation from these molecules covers the spectral region from the near infrared all the way to the vacuum ultraviolet, suggesting the possibility of high efficiency lasers over a broad spectrum.

The associative xenon molecule was chosen as an initial candidate because of some recent encouraging results. Basov[1] and co-workers in Russia reported spectral evidence of stimulated molecular emission in liquid xenon with an indicated conversion efficiency of 50%. Subsequently, Koehler et al[2] presented evidence of stimulated emission from gaseous xenon at high pressures. These results were recently substantiated by Gerardo et al.[3] From calorimetric measurements of an E-beam excited high pressure xenon, Kolb et al[4] concluded that there must be superradiance in the medium.

We reported previously[5] the observation of line narrowing in high pressure xenon using a Febetron 705 for excitation. The data obtained in these experiments, using a small beam cross section, gave only a marginal evidence of lasing in xenon, probably due to the small ($\sim$ 1 cm) gain length available. During the follow-on program, a cooperative effort was undertaken, with the Los Alamos Scientific Laboratory and Maxwell Laboratories of San Diego, to conduct experiments with a larger gain length using the Maxwell Laboratories "POCO" machine.

3.0 THE EXPERIMENTAL SETUP AND PROCEDURES

The experimental arrangement, schematically shown in Figure 1, consisted of an optical cavity inside a high pressure gas cell excited transversely by an E-beam. The laser resonator was formed by a 5m total reflector on a pyrex substrate and a plane output mirror on a MgF_2 substrate spaced 12 cm apart. The mirrors were prepared by aluminum coating with a MgF_2 overcoat. The reflectivity of both the mirrors as well as the transmission per pass through the output coupler was measured at 1700Å by a VUV reflectometer. The optical cavity was housed within the pressure cell so that the static pressure on both sides of the cavity mirrors could be the same. This greatly reduced the mechanical requirements on the mirrors and their mounts. Each mirror was mounted on an adjustable mirror mount with an aperture of ~1 cm diameter. With larger apertures, the output mirror had a tendency to fracture since the strength of the substrate material was not sufficient to prevent damage from shock waves following the pulse excitation. The two sides of the flat output mirror were paralled to ~10 arc sec so that the resonator alignment could be accomplished simply by a He-Ne laser, first by aligning the inside surface of the total reflector and then the outside surface of the flat output coupler.

The E-beam was supplied by a Maxwell Laboratories POCO electron accelerator delivering ~150 kiloamps at 0.5 MeV. The circular beam was shaped by a 15 kilogauss magnetic field into a rectangular cross section of 9 cm x 3.5 cm. The E-beam entered the high pressure cell through a 1 mil thick titanium foil with an appropriate foil support structure. The average current density inside the high pressure, stainless steel, gas cell was estimated to be ~1200 amp/cm^2 providing approximately 1200 joules of input energy to the gas. The relevant parameters of the E-beam excited xenon laser experiments are given in Table I.

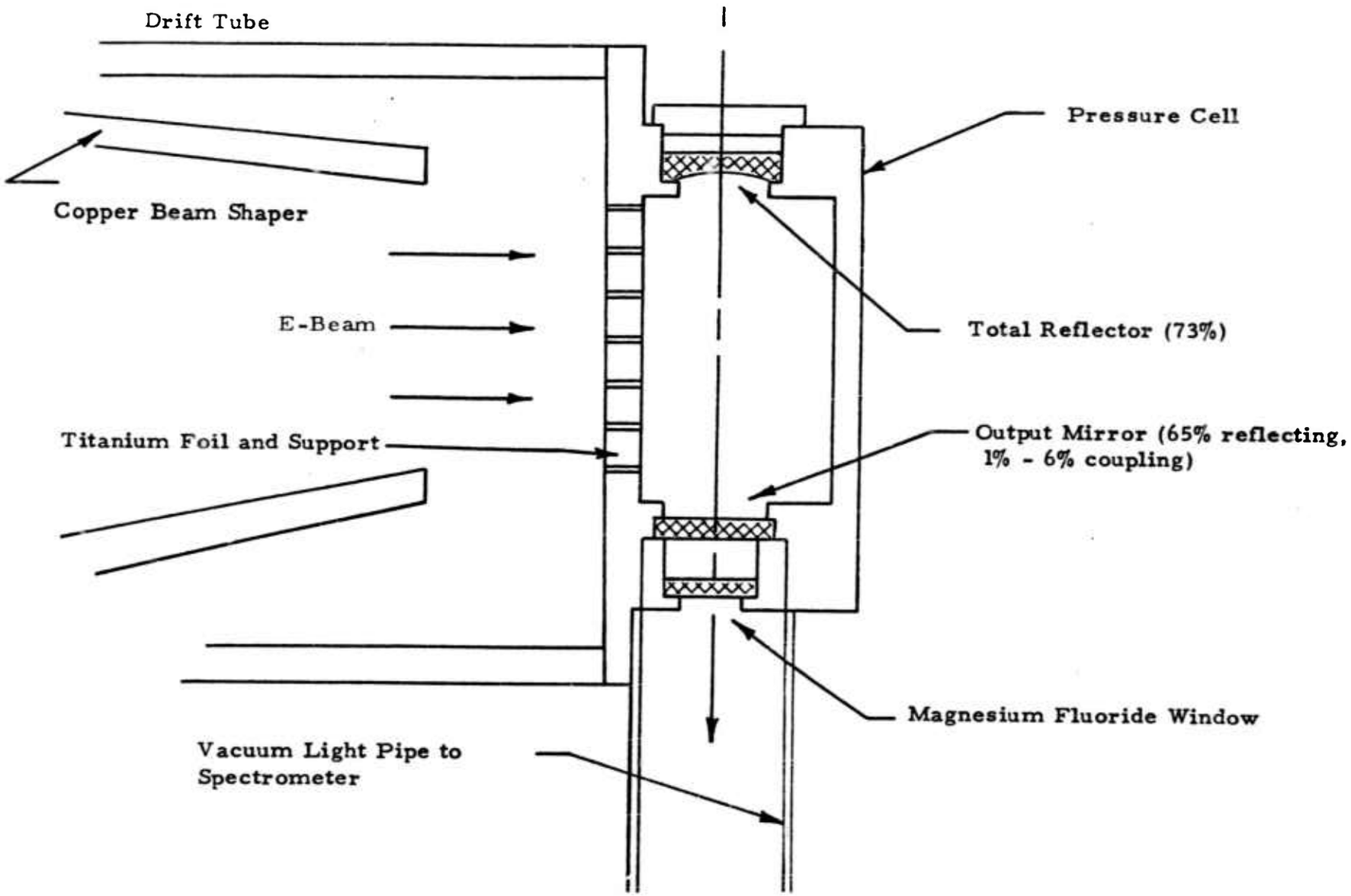

Figure 1. Schematic of the experimental arrangement of the xenon molecular laser.

TABLE I. Parameters for Experiments with Maxwell POCO Beam

Diode Voltage (Peak)	~ 500 kV
Cathode Current	~ 150 kA
Measured Beam Fluence Incident on Xenon Gas	40J/cm^2
Estimated Current Density at Cavity Window	~ 1.2 kA/cm^2
Usable Beam Cross Section	3.5 x 9 cm
Cavity Length	12 cm
Pulse Length	~ 80 ns
Pressure Range	1-20 atm

The laser diagnostics were primarily accomplished by spectral measurements to observe line narrowing. The spectral measurements were performed with a SPEX 3/4 meter VUV spectrograph Model 1500 provided with a 1200 lines/mm grating blazed at 1500Å. The dispersion of the spectrograph near 1700Å was ~10Å/mm with a maximum resolution of 0.1Å. The spectrograph was calibrated with a Xe atomic resonance line at 1470Å. Radiation from the laser cell was directed by a 45° mirror through an evacuated chamber to the spectrograph and was recorded on Kodak 101-01 special UV film. The internal pressure of the spectrograph was maintained at 10^{-5} torr to minimize absorption by residual gas in the optical path. In most of the spontaneous emission experiments the spectrograph slit width was kept at 400μ limiting the resolution of the spectrometer to nearly 4Å.

Attempts were made to maintain the gas purity by carefully pumping all gas containers to less than 10^{-5} torr and using metal seals wherever possible. The spectrograph camera was shielded to keep the x-rays from fogging the film.

4.0 RESULTS OF THE EXPERIMENTS

The densitometer traces of the xenon laser spectrum at 12 atm gas pressure are shown in Figure 2a. The corresponding spontaneous emission spectrum, obtained without the mirrors, is shown in Figure 2b. The intensity of the spectrum in 2a was reduced by a factor of at least 1000 compared to that in 2b. A slit width of 50μ was used in Figure 2a giving 0.5Å resolution on the film. These densitometer traces show that the laser emission occurs at two wavelengths spaced 5Å apart and centered at 1730 $\pm$10Å while the spontaneous emission width is ~150Å centered also around 1730Å. The separation between the two lines is ~160 cm^{-1}.

The mode structure as depicted by the symmetric burn pattern on both mirrors is shown in Figure 3. The pattern is obviously multimoded since the fundamental mode waist is calculated to be ~0.2 mm. The mirror burn spot became smaller and moved toward the E-beam entrance window when the pressure was raised to 15 atm showing possible reduction in electron range due to scattering in the high pressure gas. This is in agreement with the experimental observations of Koehler et al[2] where, at pressures over 150 psi, a 0.5 MeV electron beam did not transit a volume defined by a collimator of 1.27 cm diameter.

The mirror burn spot attendant with line narrowing was repeatable in every shot. When the mirrors were misaligned, or one of the mirrors was taken out, there was neither a burn spot on the mirror nor any line narrowing. This shows that the observed mirror burn spot could not occur due to damage by scattered E-beam, pressure wave or superradiant emission.

In these experiments the laser energy was not measured; therefore an attempt was made to estimate the laser energy from the mirror burn spots. The mass of the evaporated, 1.25 cm^2 x 800Å coating on the semitransparent window

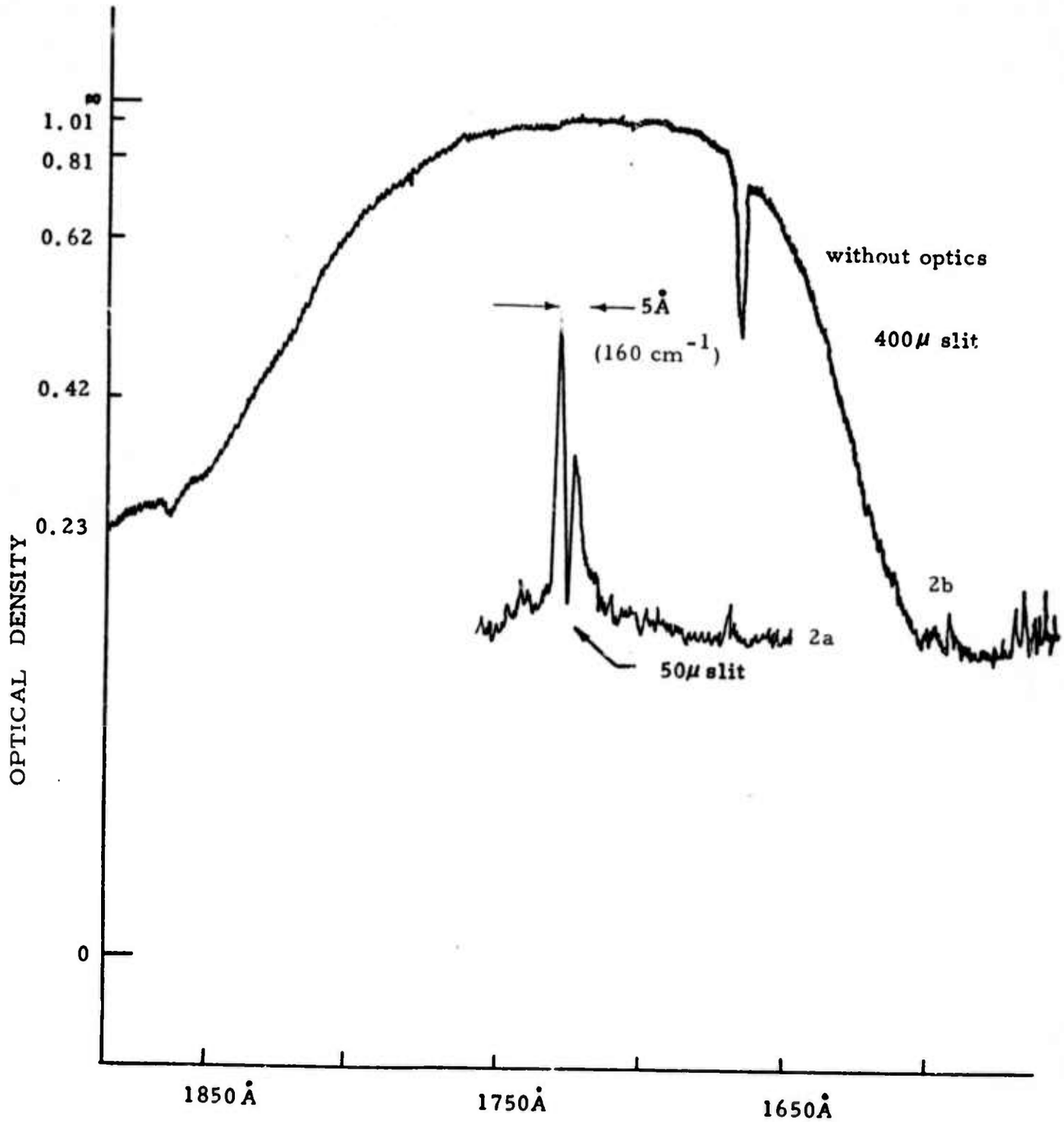

Figure 2a. Xe laser spectrum (attenuated by ~1000 compared to Figure 1b). Spectrometer slit width 50μ → 0.5Å resolution. Output coupling ~1%. Absorption losses 34% and 27% for the flat and spherical mirrors, respectively.

Figure 2b. Typical fluorescence spectrum for xenon gas at 12 atm without optics (400μ slit → 4Å resolution).

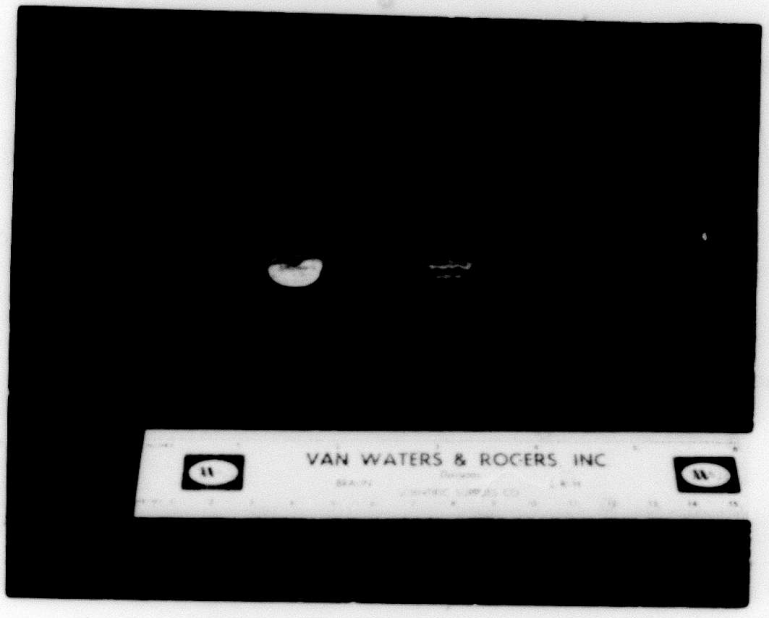

Figure 3. Photographs of the output mirror (magnesium fluoride) on the left and the spherical (5 meter radius) total reflector on the right. The electron beam direction is from bottom to top in this picture.

is 2.7×10^{-5} gm. The heat absorbed by the coating for evaporation was estimated to be nearly 0.38 joules. The coating on the total reflector was thicker and did not evaporate completely. But since the absorption by the coating and the spot size are nearly the same on both mirrors, the total reflector should have absorbed another 0.38 joules. Considering also the energy necessary to burn off the MgF_2 overcoats, the total laser energy was estimated to be approximately 1 joule. This corresponds to an energy extraction of 0.1J/c.c. The extraction of laser energy was from a volume less than 25% of the total volume excited because of the mirror apertures. Since the measured E-beam energy inside the cavity was 1200J, the laser efficiency is estimated to be on the order of 0.3%.

The separation between the lines of the laser doublet was quite large compared to the resolution capability of the spectrograph. When the slit width was reduced from 400μ to 50μ, the doublet became sharper and well resolved. Contribution of any absorption causing the appearance of a doublet may be eliminated. Transmission studies show that there is no absorption at 1730Å by the optics or the windows. This leaves only the laser medium itself as a possible source of absorption. No trace of a strong line absorption of this nature was evident in the spontaneous emission spectrum. In a properly aligned resonator, however, the light transits the laser gas several times which could effectively increase the total absorption. But in this case there is also a high gain at the same wavelength which should compensate for the loss due to possible absorption. Therefore the observed doublet must be a genuine feature of the laser emission spectrum.

5.0 DISCUSSION OF THE RESULTS

The excitation mechanism of the xenon laser is believed to be the following: The unexcited xenon gas consists mostly of free xenon atoms (Xe) together with some loosely bound (0.025 eV) Van Der Waal molecules (Xe_2). But when the Xe atom is either ionized or excited by electron impact, stable excited molecules are formed through a series of interactions summarized below (together with their rate constants):

$$Xe^+ + 2\,Xe \rightarrow Xe_2^+ + Xe; \qquad k \sim 10^{-31}\ cm^6/sec$$

$$Xe_2^+ + e \rightarrow Xe^* + Xe; \qquad k \sim 10^{-6}\ cm^3/sec$$

$$Xe^* + 2\,Xe \rightarrow Xe_2^* + Xe; \qquad k \sim 10^{-32}\ cm^6/sec$$

A qualitative energy level diagram of Xe_2 as constructed by Mulliken[6] is represented in Figure 4.

The laser emission is due to transitions from the lowest vibrational level of the lower excited states of Xe_2^*. The population density of the laser upper state is estimated, on the basis of the above processes, to be $4 \times 10^{17}/cm^3$ for a 12 atm gas excited by 1000 amp/cm^2, 0.5 MeV E-beam. The stimulated emission cross section for the laser transition is given[7] by

$$\sigma = \frac{\lambda^2}{8\pi^2 \tau \Delta\nu}$$

where τ is the spontaneous decay time and $\Delta\nu$ is the halfwidth at half maximum intensity. σ is thus calculated to be 3×10^{-18} cm^2. The small signal gain for a population density of $4 \times 10^{17}/cm^3$ should therefore be 120% per cm. But, since the emitted photon has enough energy to ionize the Xe_2^* molecule, photoionization will compete with stimulated emission leading to a lower gain. The photoionization cross section $\sigma_{p.i.}$ for Xe_2^* has been calculated[8] to be

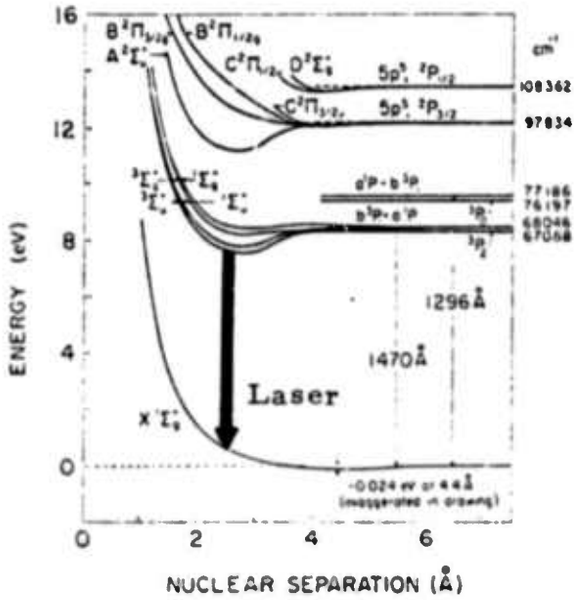

Figure 4. Energy level diagram of the Xe_2^+ which is similar to those of Xe_2.

$\sim 2 \times 10^{-18}$ cm^2; the net gain should therefore be ~40%/cm. A somewhat lower gain is expected due to processes like $Xe_2^* + Xe_2^* \rightarrow Xe_2^+ + 2\,Xe + e$ with an estimated[3] rate constant of 3×10^{-9} cm^3/sec. Therefore the effect of this process will be to reduce the population density and hence the gain by nearly 10% at these population densities. Thus the net gain is estimated to be 35%/cm. This is consistent with the observation of only incipient lasing using a Febetron 705 where the population density, and consequently the gain coefficient, was similar but the gain length was ~1 cm with a ~35% mirror absorption loss.

Since the terminal state of the molecular xenon laser decays by dissociation within the time period of a molecular vibration (typically 10^{-12} sec), the relaxation rate of the upper state determines[7] the saturation parameter. The primary mechanism of decay of the upper state is radiative.. Assuming the decay time τ to be 20 nsec, the saturation parameter I_s is given by

$$I_s = \frac{h\nu}{\tau\sigma} \approx 20\ \mathrm{MW/cm^2}$$

where σ is the cross section for stimulated emission (3×10^{-18} cm^2). Therefore the energy extraction from the xenon laser should be

$$\alpha_o I_s \approx 7\ \mathrm{MW/cm^3}$$

The estimated extraction of 0.1J/cm^3 of the experimental results implies a laser pulsewidth of ~15 nsec, which is reasonable in this experiment.

The results presented here leave little doubt that laser oscillation was observed from molecular xenon in the VUV region. Since the emitted photon has enough energy to ionize an excited xenon molecule, it was not obvious that a significant laser emission in the VUV could actually be achieved in high pressure xenon.

These results demonstrate that sufficient net gain for laser oscillations from molecular xenon is possible in spite of the competition with photoionization.

The results also demonstrate the possibility of obtaining stimulated emission from other molecular associations. Numerous molecular associations of this type are known with emission wavelengths ranging from the VUV to IR region. These emissions are inherently broadband indicating the possibility of tunable lasers over a wide range of frequencies. Since these emissions usually have high quantum efficiency, high overall efficiencies should be possible in addition to tunability.

The efficiency of energy deposition into a gaseous medium by relativistic electron beams can be as high as 30-50%. With a quantum efficiency of 80-90%, an overall efficiency of 20-40% may be expected. However, due to the effects of photoionization and other similar loss processes, actual efficiencies may be in the order of 10%.

6.0 IDENTIFICATION OF THE LASER DOUBLET

The observed xenon laser spectrum has been concluded to be a well resolved doublet separated by nearly 160 cm^{-1}. This doublet appears to be due to transitions from the lowest vibrational level of upper states $^1\Sigma_u^+$ and $^3\Sigma_u^+$ to the ground state $^1\Sigma_g^+$ (see Figure 5). However, another possibility that might give rise to a doublet should be considered. If the $^3\Sigma_u^+$ is split into 1_u and 0_u^- with a separation of nearly ~ 160 cm^{-1}, the transitions from them could give a doublet. But the transitions from 0_u^- are expected to have orders of magnitude lower probability. Therefore, once the laser starts to oscillate with transitions from 1_u, the population of 0_u^- would cross relax since 1_u and 0_u^- belong to the same triplet manifold, preventing any lasing from 0_u^-.

On the other hand, since the transitions $^1\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ and $^3\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ have much closer probabilities and singlet-triplet intersystem crossing is somewhat slow, simultaneous laser oscillations on both transitions may be possible at least under certain conditions. However, one may question as to why the separation between the lines is so small compared to the anticipated upper state singlet-triplet separation. The explanation may be that the ground state repulsive potential curve is actually much steeper than shown by Mulliken[6] and the $^1\Sigma_u^+$ has a slightly smaller equilibrium internuclear distance than the $^3\Sigma_u^+$. Since electron transition follows the Franck-Condon principle, the difference in energy for the two transitions would be smaller than the actual separation of the singlet and the triplet upper state, as illustrated in Figure 6. An accurate identification of the doublet has to await computation of the singlet-triplet separation as well as a more accurate determination of the repulsive potential curve of the ground state.

Koehler et al[2] reported the observation of two different decays, one with a 2 nsec and the other with a 20 nsec decay time. It appears that the former could be due to the singlet-singlet transition while the latter may be due to

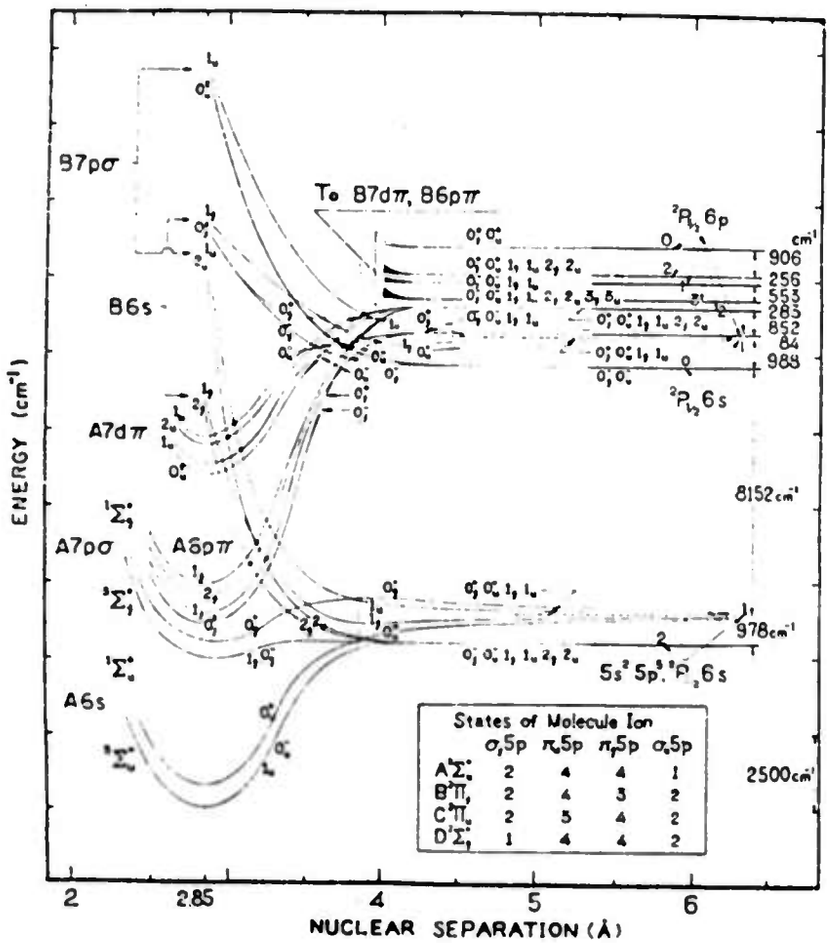

Figure 5. The estimated potential energy diagrams of the upper electronic states of Xe_2. (Reference 6)

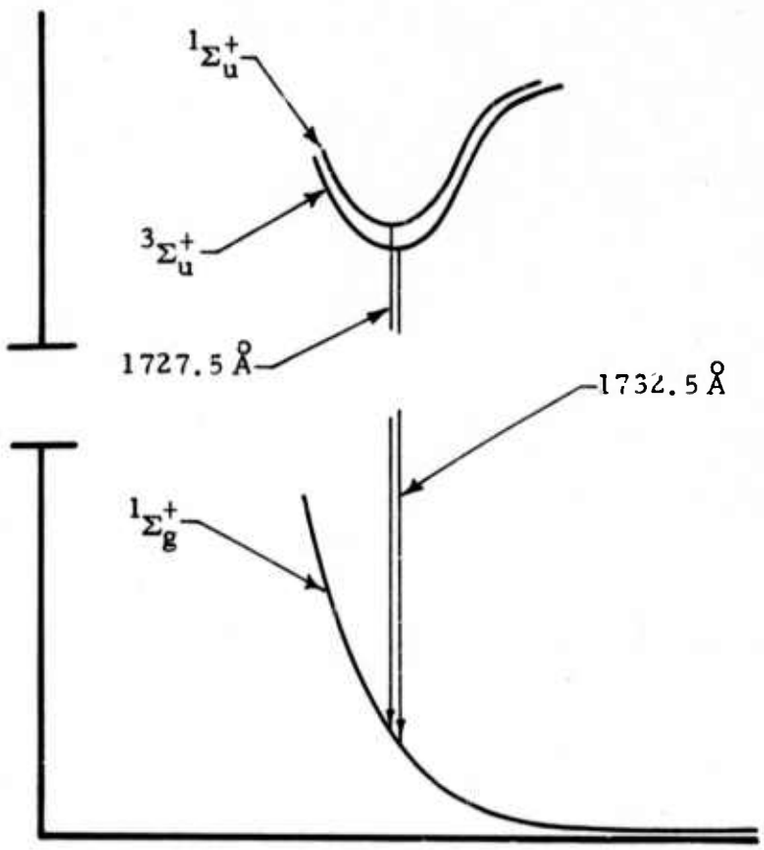

Figure 6. Proposed qualitative feature of the Xe_2 energy level diagram that could lead to a smaller energy separation between the transitions $^1\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ and $^3\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ compared to the actual separation of the upper levels.

triplet-singlet transition. The $^1\Sigma_u^+$ Xe_2^* state, which corresponds to the $^3P_1$ Xe atomic level, should have an allowed transition with a transition probability of the same order as that of the atomic level. Wilkinson[10] reported the lifetime of the $^3P_1$ state to be $(3.74 \pm 0.3) \times 10^{-9}$ sec. Therefore a decay time of 2 nsec for the transition $^1\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ is not unexpected. Also, only an order of magnitude longer decay time for the transition $^3\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ is reasonable since the wave functions are highly mixed in this case.

In the absence of any cross relaxation, the transition $^1\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$ may have the higher intensity since the transition probability and consequently the rate of energy extraction for this transition is higher; this is based on the reasonable assumption that nearly an equal population would reach the two states $^1\Sigma_u^+$ and $^3\Sigma_u^+$ through the chain of processes associated with the excitation mechanism. However, an examination of the spectrum in Figure 2a clearly shows a higher intensity for the longer wavelength line which is due to the transition $^3\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$. This indicates at least a partial cross relaxation between the singlet and the triplet upper state during the excitation pulse. Such a higher rate for the singlet-triplet intersystem crossing may be possible due to the strong spin-orbit coupling and the highly mixed wave functions.

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