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Principal Investigator: Stephen R. Leone

Phone: (303)-492-5128

Present Address: Joint Institute for Laboratory Astrophysics
University of Colorado
Boulder, CO 80309

Contractor: University of Colorado

Scientific Officer: Dr. Robert Behringer, Office of Naval Research

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 -- 20C - PRINCIPAL INVESTIGATOR: LEONE, S R
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Technical Report Summary

A new class of diatomic lasers has been developed by optical pumping. These are the $S_2(B^3\Sigma_u^- - X^3\Sigma_g^-)$ and $Te_2(AO_u^+ - XO_g^+)$, $(BO_g^+ - XO_g^+)$, which lase on lines in the near ultraviolet and visible regions of the spectrum. Optical excitation studies show that these molecular systems have high efficiency, may be scalable to high average powers, are nondegradable, and offer promise for pumping by other excitation schemes, such as electric discharge. It is possible that one or more candidate systems in this category of lasers might be useful in generating blue-green radiation for underwater applications. Preliminary investigations with electric discharge excitation show good formation of glow discharges in sulfur dimer vapor. Materials problems and the difficulties of high temperature discharge technology appear to be the major drawbacks in pursuing such studies further for laser action.

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Technical Report

Introduction

There is substantial interest in developing high average power, visible and ultraviolet lasers. Numerous approaches have been employed, some of which are now very successful. The molecular excimer lasers, KrF, XeF, etc. offer promise of high efficiency with electron beam and discharge excitation of high-pressure mixtures. The mercury-halide, HgCl₂, HgBr, etc. lasers now operate on bound-bound transitions with e⁻ beam and discharge pumping. It is apparent that recent discoveries and exploration of these new molecular laser species are rapidly changing our concepts of potentially important visible and UV lasers. There is undoubtedly a promising and important field of research in new diatomic molecular lasers, which lies ahead for the next several years.

Our group has been engaged in exploratory work on new visible and ultraviolet lasers. The approach we have taken is to use a modicum of spectroscopic and literature insight to identify molecular species which may be potentially good laser candidates. The candidates identified must offer promise to be eventually efficient and practical laser devices, not just laboratory curiosities. Effective demonstration of the lasing capability has initially been achieved by optical pumping experiments. These techniques have been highly productive in the discovery process, and are likely to continue to be so. A series of dimer lasers formed out of the group VI elements (O, S, Se, Te) have been discovered. All of the possible homonuclear and heteronuclear diatomic species, with the exception of O₂, must be formed at high temperature from the elements. Again with the exception of O₂, most of the diatomics formed from these elements have bound levels well below their lowest dissociation limit.¹ Fortuitous displacement of their electronic potential surfaces makes population inversion with respect to high vibrational levels in the ground state

very easy.² Thus, these molecules offer the potential for highly efficient laser action. Electrons with energies of 4-5 eV could produce 2-3 eV photons by direct excitation techniques.

The S_2 and Te_2 dimers have both shown efficient lasing by optical pumping using a doubled dye laser and a dye laser respectively.²⁻⁵ The laser optical pumping experiments afford a unique opportunity to determine desirable and undesirable parameters, and to assess the potential of such systems. In addition, these experiments have pointed out the potential to pump these novel laser systems by other chemical, electrical, or energy transfer mechanisms. The S_2 upper lasing state has been observed in emission in countless experiments involving chemical reactions and discharges over the years.⁶⁻⁹ It may be possible to utilize one or more of these observations to develop an efficient laser system.

Optical Pumping Studies

Laser emission has been achieved in the sulfur dimer, S_2 ,^{2,4} on its $B^3\Sigma_u^- - B^3\Sigma_g^-$ transition, and in the tellurium dimer, Te_2 ,^{3,5} on the $A0_u^+ - X0_g^+$ and $B0_u^+ - X0_g^+$ transitions. In S_2 , stimulated emission has been observed from 365-630 nm, and in Te_2 from 565-620 nm. In addition, Te_2 and S_2 have been shown to laser cw.¹⁰

A previous report of an S_2 laser on the $1^1\Sigma_g^+ - 3^3\Sigma_g^-$ transition at 1.1 μm has been made.¹¹ The S_2 laser observed by our group operates on a different transition, primarily in the short wavelength visible and near ultraviolet. Figure 1 sketches the approximate potential surfaces for the known S_2 molecular states.¹² Although many details about these surfaces and the exact locations of other states are not known, several qualitative features of these

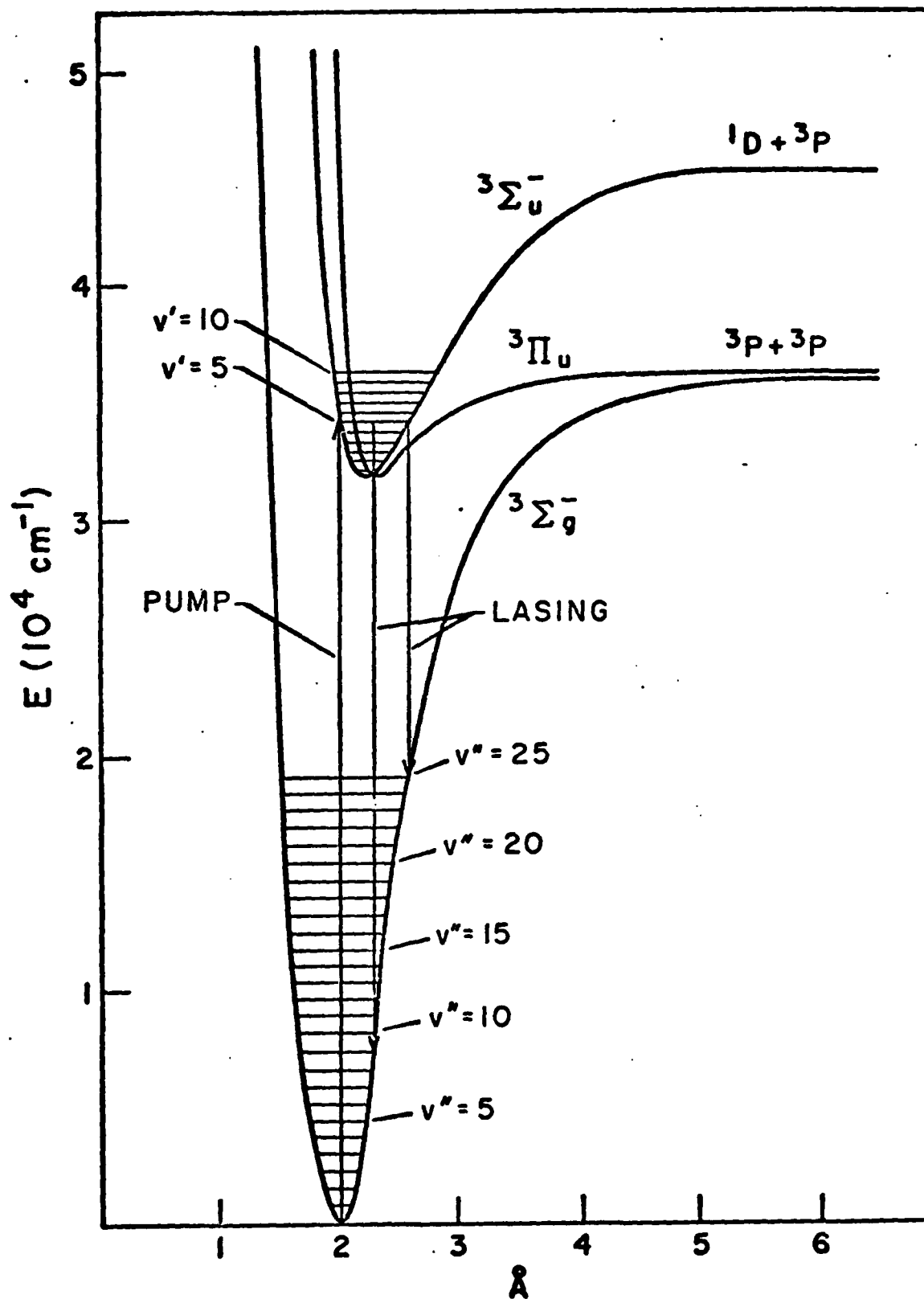


Figure 1

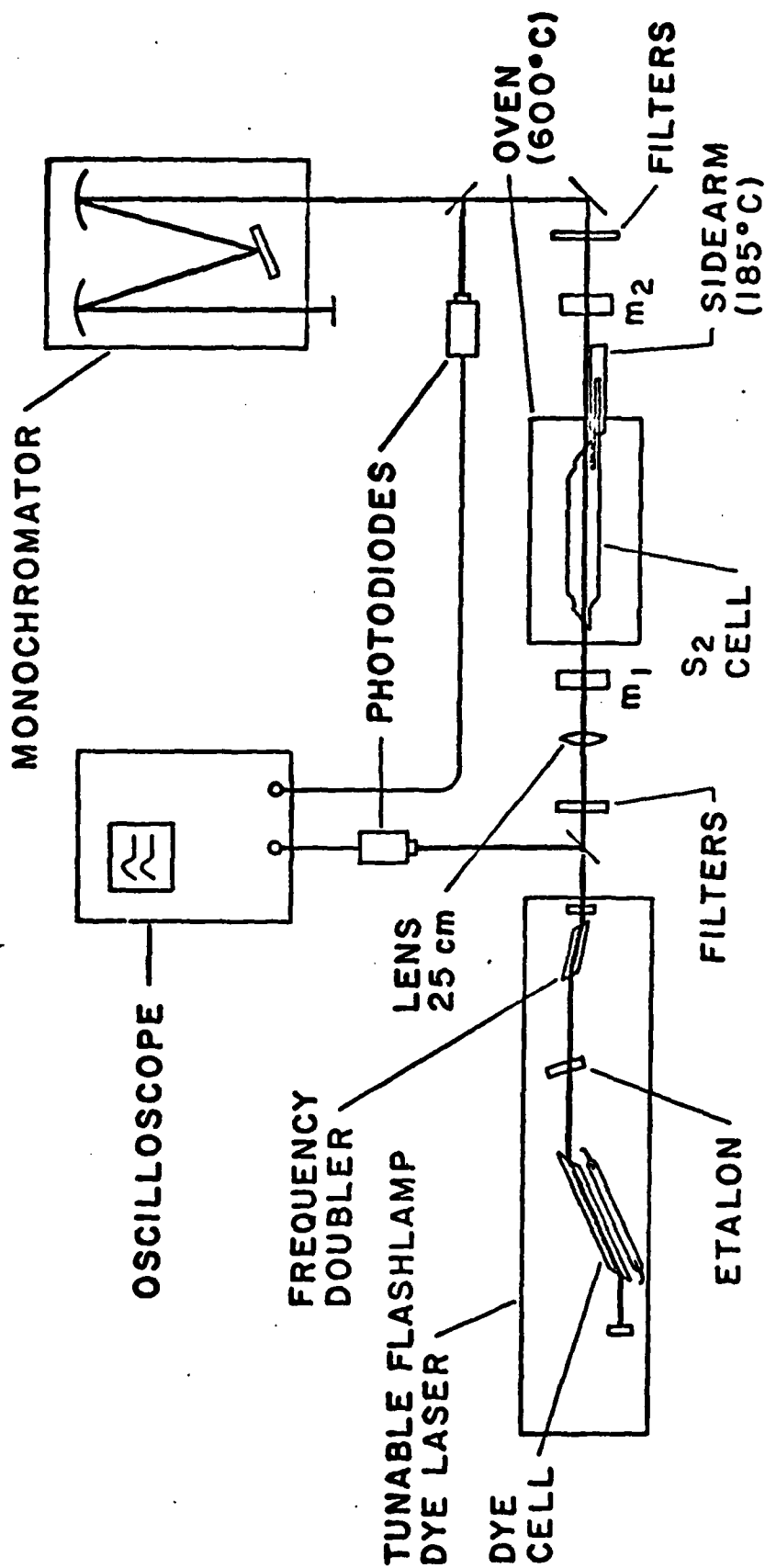
potential surfaces are most important. All of the lasing states observed, including $v' = 0-7$ are well below the dissociation limit of the S_2 molecule. Thus, the lowest vibrational levels of the $B^3\Sigma_u^-$ state, although heavily perturbed, are not predissociated.¹² The potential surfaces are fortuitously displaced so that the lowest vibrational levels of the $B^3\Sigma_u^-$ excited state are situated over high vibrational levels of the ground $X^3\Sigma_g^-$ state, which are not populated at the 500 C operating temperature. Thus a small fractional excitation into the B state will be an inverted population. The Franck-Condon factors for these B-X transitions are as large as 0.1 for many bands, and the measured radiative lifetimes of individual states fall in the range of 20-45 nsec.¹³⁻¹⁵ With stimulated emission cross sections in the range of $5 \times 10^{-14} \text{ cm}^2$, the S_2 system makes a high gain laser.

The experimental set-up is shown in figure 2, and consists of a pump laser, an S_2 cell, and associated measurement devices. The S_2 molecules are generated simply by heating sulfur in an evacuated, all fused quartz Brewster angle cell. Table I shows some typical data for the partial pressures of

Table I¹⁶
Formation of S_2 in the Equilibrium Vapor

	600 C 92.5 mm	600 C 740 mm	400 C 93 mm
S_2	75.3%	36.0%	4.4%
S_3	3.6%	3.2%	0.5%
S_4	14.6%	19.1%	1.5%
S_5	2.9%	10.2%	7.8%
S_6	2.3%	15.0%	32.8%
S_7	1.2%	13.7%	34.6%
S_8	0.1%	2.8%	18.4%

APPARATUS FOR OPTICALLY PUMPED S₂ LASER



CHARACTERISTICS: 0.3 cm⁻¹ LINEWIDTH
1 mJ/PULSE, 30 Hz
~300 nm

m₁: R ~ 99 %
m₂: R ~ 50 %

Figure 2

various S_n molecules as a function of temperature and total pressure. At typical working vapor pressures between 1 and 10 torr, more than 95% of the sulfur exists as S_2 at 600 C. The S_2 species is even more predominant at higher temperatures. Sealed cells of sulfur and tellurium have operated indefinitely in our set-up without degradation. The main body of the cell is maintained at 600 C with a tube furnace, and the vapor pressure of the sulfur inside is adjusted independently with a sidearm at a lower temperature.

The S_2 laser is pumped with the output pulses of a frequency-doubled, tunable dye laser. Such a pump laser offers the advantage of being able to tune to almost any transition desired for all types of molecular systems. The dye laser has characteristic pulsed energies in the ultraviolet of 0.5 mJ, 2 μ sec duration, and a 0.5 cm^{-1} band width using a low-finesse ($R = 30\%$ etalon). The dye laser is usually mildly focused to a beam radius of 100 μ m. Typical pump wavelengths are 295-305 nm.

All of the $^3\Sigma_u^-$ vibrational states which have been excited exhibit strong lasing action throughout the short wavelength visible and near ultraviolet. Sharp P- and R-branch doublets are observed for each lasing transition. Gain was observed to be superfluorescent on the strongest lines at higher S_2 pressures.

With typical 99% R and 50% R mirrors, the energy conversion was measured to be 2% for the total visible S_2 laser output. From a number of standpoints, the optically pumped S_2 laser was not fully optimized. The 0.5 cm^{-1} pump spectrum is ten times broader than individual S_2 lines, and much of the pump light was not being used to produce the lasing state. Proper mode matching of the focused pump beam to the S_2 laser was not attempted. The 3-m-radius-of-curvature mirrors on the S_2 laser resonator sustain a beam area inside the cavity which is an order of magnitude larger than the pump zone. Bright

spontaneous emission is observed from the side of the S_2 cell because of the short radiative lifetime. Even when the best lasing was obtained, this emission was not dramatically reduced in intensity. More proper mode matching should enhance the output efficiency nearly to the theoretical limit.

The 308 nm line of the $XeCl_2$ excimer laser overlaps with one line of the (2,0) band of S_2 (B-X), providing an efficient pump source for the molecular S_2 laser. A series of strong laser transitions from S_2 occur in the 480-500 nm region, and are listed below (Table II). In addition, the 337.1 nm line of the nitrogen laser overlaps with the (4,2) band of S_2 , and results in successful laser action.

The output coupler of the S_2 laser can be replaced with a grating for line tuning. When pumping a single line and lasing on a single line, the S_2 laser pulse was observed to follow the entire time duration and shape of the 2- μ sec doubled dye laser pumping pulse. Thus, no bottlenecking problems have been observed in these low energy optical excitation experiments. It is known that $S_2^* - S_2$ collisions can quench the excited B state, but that collisions of S_2^* with rare gases can serve to relax the excited B state vibrationally and rotationally without quenching.¹⁷ It appears most reasonable that a steady-state collisional relaxation of the vibrational manifolds in the ground and excited state will allow considerable input and extraction of energy.

Table II

Line (nm)	Tentative lasing band
~474	(2,17)
~486	(2,18)
~501	(2,19)
~517	(2,20)

The performance of the dimer lasers is most sensitively dependent on collisional effects. At very low pressures, the stimulated emission process occurs in the collisionless regime. Although this serves to prevent collisional relaxation of the upper excited state, there is no collisional removal mechanism for the lower laser level. At higher pressures, relaxation of both upper and lower laser levels occurs. Depending on the magnitudes of the pump, stimulated emission, and relaxation rates, an optimum pressure is observed.¹⁸ A kinetic balance can be achieved between relaxation of molecules back into the originally pumped ground state level, relaxation out of the lower laser levels, and the pumping and stimulated emission rates.

In the limit of high pumping flux, it should be possible to observe stimulated emission from upper laser states populated by collisional energy transfer. In all reported cases, however, lasing is only observed from the initially pumped state. In an extensive series of studies attempting to achieve lasing on collisionally populated neighboring excited states in S_2 , no lasing other than from the initially excited level was observed. Collisional relaxation of the initially pumped level populates many other states, so that the gain of any individual collisionally excited upper level is too small to lase. Similarly, when pumping a single velocity group, lasing is also only observed from that group.

Modeling of optically pumped lasers has generally been successful in predicting gain parameters, but observed output powers are sometimes lower than expected.^{19,20} The loss processes most likely responsible are population build-up in the lower laser level, incomplete repopulation of the initially pumped state, absorption of the pump from upper laser levels, and absorption of the dimer laser by excited levels populated through collisional relaxation of the lower laser level.

A detailed understanding of the collisional relaxation processes is an important goal. In interpreting the results of both the S_2 and Te_2 pulse pumped dimer lasers, the observations of quasi-continuous laser operation can be explained only if the collisional relaxation rates of the upper and lower laser levels are essentially gas kinetic. The difficulty of designing good experiments to probe the details of vibrational, rotational, and velocity changing collisions should be apparent. Rather than simply observing the output characteristics of optically pumped lasers, various pump-laser experiments using the dimer laser as a probe will be possible in the future. This should provide more detailed information about the numerous inelastic collision processes occurring in dimer systems.

Discharge Studies

Construction of a device for direct electric discharge pumping of the S_2 molecular laser was attempted. As mentioned above, the S_2 molecule has fortuitous placement of its potential surfaces, which allow population inversions to be achieved readily. By close analogy to the data available for electron collisions with O_2 , it is likely that electron collisions with S_2 will produce significant excitation on the $B^3\Sigma_u^- - X^3\Sigma_g^-$ transition.²¹ As observed in the optically pumped laser, small fractional populations produced in the $B^3\Sigma_u^-$ excited state will be inverted with respect to the high vibrational levels of the ground $X^3\Sigma_g^-$ state. The exceptionally high gain of the S_2 stimulated emission makes the prospect for attaining laser action by direct electron excitation very favorable.

In consultation with Richard T. Weppner at the Joint Institute for Laboratory Astrophysics, a number of designs for direct electron excitation of

the S_2 laser were considered. A design is necessary which pushes the current state of technology. The S_2 molecules are ideally contained in a 600 C, meter-long tube, with transverse electrodes to produce a very fast (~ 10 nsec risetime), 50 kV discharge uniformly over its entire length.

A brief discussion of the parameters relevant to discharge excitation of the S_2 molecule should be considered. From spectroscopic measurements, values for the transition strengths of various S_2 lines have been given. More recent measurements of the $S_2(B^3\Sigma_u^-)$ lifetime give 45 nsec.¹⁵ Taking $A_{V''J''}^{V'J'}$ for a single line to be $1 \times 10^6 \text{ sec}^{-1}$, we can obtain a coefficient for stimulated emission. For a single S_2 emission line at line center, $\lambda = 500 \text{ nm}$, Doppler width $= 0.054 \text{ cm}^{-1}$ and $T = 600 \text{ C}$, the stimulated emission coefficient is $1.5 \times 10^3 \text{ cm}^{-1} \text{ torr}^{-1}$ ($\sigma = 4.6 \times 10^{-14} \text{ cm}^2$). The density of S_2 molecules in a single vibration-rotation level of the $B^3\Sigma_u^-$ state needed to achieve a gain of e^5 in a 100 cm path length is conservatively estimated to be $1 \times 10^{12}/\text{cc}$. This number agrees well with what is observed empirically in our optical pumping experiments. Because the S_2 system is intrinsically high gain, stimulated emission can be achieved in a small number of passes. The short spontaneous lifetime (45 nsec) favors the use of fast electrical discharges initially. There is no reason to preclude that longer current pulses cannot eventually be used. Indeed, the optical pumping experiments produced stimulated emission over the full duration of the 2 μsec pumping pulses. A gain of e^5 in a 1 meter path length will require only 4-5 transits for full laser action to be achieved. This is a conservative requirement for current pulses in the initial range of 20-50 nsec duration and optical cavities of 1.5 meter path length.

On electron bombardment, excitation to the $B^3\Sigma_u^-$ level will lead to a distribution of vibration-rotation states. If we neglect the weaker triplet

splittings and recognize that only even rotational levels are allowed in the $^3\Sigma_u^-$ state,¹ there will be approximately 500 levels available at 600 C ($V' = 0-9$, $J' = 0,2,4\dots100$). We expect that an excited state density of approximately $5 \times 10^{14}/\text{cc}$ distributed among all levels is adequate to produce laser stimulated emission. A density of $5 \times 10^{14}/\text{cc}$ excited molecules is an entirely reasonable expectation for discharge excited lasers.²² Reasonable starting densities from discharge considerations will be in the range of 5×10^{16} to 5×10^{18} S_2 molecules/cc. Choosing $5 \times 10^{17}/\text{cc}$, then an excitation of only 0.1% of the ground molecules to the $B^3\Sigma_u^-$ state is required.

Under the condition where there are few collisions during the excitation pulse, the S_2 molecule would lase on thousands of lines simultaneously. In the optical pumping experiments, as many as 10-12 doublets lase simultaneously when exciting only a single level. In the electric discharge laser, a distribution of perhaps as many as 500 levels in the $B^3\Sigma_u^-$ state will lase. By addition of He or Ar it is expected that this large number of excited states can be compressed into a few of the lowest vibrational levels.¹⁷ Using rare gases to produce rapid rotational and vibrational relaxation, significant fractions of the total available energy could be extracted on single frequencies. In addition, relaxation of lower laser levels is important to prevent the problem of bottlenecking and premature termination of the laser.

Electrons with energies in the range of 4-5 eV are expected to excite the $S_2(B^3\Sigma_u^- - X^3\Sigma_g^-)$ transition. From close analogy to the O_2 system, excitation of this transition can be made a dominant fraction of the total cross section at appropriate values of E/N (V/cm-torr).²¹ Electron excitation of the S_2 molecules to high vibrational levels of the ground state will have very small cross sections, and thus for lower laser levels above $V'' = 15$ there is no appreciable excitation by either electron collisions or the 600 C temperature.

The first discharge tests were made in a 1.5 meter, longitudinal discharge cell, heated to 600 C to provide the sulfur dimer. With current risetimes of 500 nsec, the discharges in sulfur were completely homogeneous, glow discharges, at up to 2 Torr of the dimer. Addition of helium sustains the uniform glow discharge up to sulfur dimer pressures of 6 Torr. Much higher pressures are feasible when scaled to a few cm transverse electrode gap. The light emission from the sulfur discharges is extremely bright. Extensive spectral studies of the emission light have been made by other workers.²³ The predominant emission is from the B-X transition of the S₂ molecule. However, it is still not known whether the S₂(B) state is produced by direct electron impact.

In order to test the possibilities of discharge lasing, a meter long, transverse, high temperature discharge laser device was constructed. The device consists of a quartz laser tube, with 10 pairs of tantalum wire electrodes, each 10 cm long, spaced 2 cm apart. The tantalum electrodes were gold-brazed to tungsten feedthroughs which were fused directly into the quartz with GS-1 glass. Ten 2.7 nF capacitors on each side of the laser tube formed a voltage doubling circuit with approximately 15 Joules of energy storage. The experimentally determined discharge parameters were: 50 kV with a 15 nsec current risetime which was measured optically. These characteristics were highly effective in producing the usual 3371 Å laser transition in nitrogen, with 3 mJ of energy.

The entire laser tube was housed in a high temperature oven, consisting of 120° sector ceramic potted heaters and hollowed out firebricks. The 15 nsec risetime achieved was excellent,, considering the long current return paths necessitated by the bulky oven. The laser tube itself sustains 800 C operation under high vacuum (10^{-6} Torr). The tungsten feedthroughs are maintained

at a 250 C lower temperature than the main body of the tube in order to reduce corrosion rates with the hot sulfur vapor.

Introduction of sulfur vapor into the laser tube was found to be complicated by several problems. Contact of the hot sulfur vapor with the tantalum and tungsten electrode materials generated several new species, one of which is volatile at room temperature, and another which has significant vapor pressure at high temperatures. Even though gross macroscopic decomposition of the electrode materials was very slow, the generation of these new gases was rapid each time the sulfur was introduced into the tube. These new vapors had two serious effects. The high temperature compound absorbs in the visible, appearing to be an extremely strong, broad-banded absorber. The brilliant blue discharges observed in test cells of S_2 vapor were modified to give only weaker intensity red or yellow discharges. There were several differences in the test cell designs from the final laser tube. In one test cell, a longitudinal discharge was used, so that the electrodes were always maintained at low temperatures (200 C) while the main length of S_2 vapor was at 700 C. In the second test cell, a transverse geometry was used, with solid tantalum bar stock for electrodes and gold plating on stainless steel feedthroughs. In both of these cells, the generation of new compounds was not observed on heating, nor were the reddish-yellow discharges. Intense blue discharges were always observed from these test cells. There were several different features in the meter long laser, primarily with regard to the tungsten feedthroughs, which were required to make reliable seals, and the use of tantalum wire electrodes.

The observation of severe material problems points out how serious the metals compatibility is for the corrosive S_2 vapor. It appears that there may not be good metal materials which are fully compatible with sulfur vapor at the temperatures of 600 C. It is conceivable, but unlikely, that the problem

of the strong visible absorber is linked to other species in the sulfur vapor composition. For example, S_4 is known to absorb in the visible. Normally, however, at low vapor pressures (~few torr) and high temperatures (600-700 C), the concentration of S_4 is very small and has never posed a problem in all quartz cells. The behavior of the visible absorbing compound is not at all characteristic of what would occur if it were S_4 . Thus, it is likely that the visible absorber is some metal sulfide, which becomes volatile at higher temperatures.

So far, none of the physics involved would indicate that an electric discharge S_2 laser is not feasible. Other schemes might be employed to generate S_2 at lower temperatures. For example, a variety of mercury, cadmium, etc., sulfides decompose to give pure S_2 species. It might be possible to generate S_2 in an all quartz region and flow the vapor through the metal electrodes zone at low temperatures (200 C). However, this would prevent a sealed-off, continuous use system from being developed, and involves additional technological effort. Other workers have shown that the dimer can be produced at lower temperatures by dissociation of the larger S_8 , S_7 , S_6 molecules with the electric discharge itself.²³ Possibly sustained discharges with pulsed excitation on top will provide optimum conditions for lasing experiments.

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