A CHEMICAL PROCESS PRODUCING A CONTINUOUS LASER AMPLIFIER IN THE VISIBLE REGION

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In a study of the reaction dynamics of the Na₃-X (Cl, Br, I) metatheses, Na^{*}₂ is formed in several excited electronic states, the fluorescence from these states resembling in some respects that characteristic of optically pumped alkali dimer lasers. Optical gain through stimulated emission demonstrates population inversion involving several excited state levels of Na₂ and hence an amplifying medium for a visible chemical laser. A maximum laser gain of 1% of the input laser power has been observed at 5270 Å, corresponding to a gain coefficient of $\approx 2 \times 10^{-3}$ cm⁻¹. An explanation is provided for this efficient inversion process and comparisons are made with optically pumped laser systems.

1. Introduction

We have been concerned with the study of sodium cluster-halogen atom reactions (Na_n+Cl, Br, I). At sufficiently low pressures, the short-lived electronically excited molecules formed as the nascent products of these highly exothermic processes can be studied under "single collision" conditions. A major focus of these efforts has involved the study of the nascent chemiluminescent emission from the highly exothermic reactions [1]

$$Na_3 + X \rightarrow Na_2^* + NaX \quad (X = Cl, Br, I) . \tag{1}$$

The electronic emission spectra which result from these reactions (see fig. 1 and ref. [1]) encompass the Na₂ $A^{1}\Sigma_{u}^{+}-X^{1}\Sigma_{g}^{+}$, $B^{1}\Pi_{u}-X^{1}\Sigma_{g}^{+}$, $C^{1}\Pi_{u}-X^{1}\Sigma_{g}^{+}$, and $C'^{1}\Sigma_{u}^{+}-X^{1}\Sigma_{g}^{+}$, band systems are characterized by *sharp well-defined emission regions* superimposed on a weaker but perceptible background.

The rather sharp nature of certain B-X, C-X, and C'-X Na₂ emission features and their analogy in certain regions to the fluorescence characteristic of optically pumped Na₂ laser systems [2] (e.g. 5282 Å (v', v'') = (6, 14) B-X) suggested the possibility that stimulated emission analogous to that obtained in alkali dimer optical pumping experiments might have been observed. Therefore, we have carried out laser gain measurements to demonstrate that stimulated visible emission has been observed and that several of the sharper emission features correspond to the establishment of a population inversion at relatively low pressures in a high gain system.

2. Experimental conditions

Fig. 2 presents an overview of the apparatus and experimental configuration used to study the Na₃-X reactions and investigate the nature of the Na₂ produced as the product of this metathesis. All experiments, including both studies of the chemiluminescent emission (fig. 1) resulting from the Na_3-X reaction and laser gain probes of this reaction, were carried out in a two-chamber differentially pumped system (oven and reaction chamber) both chambers being evacuated to a base pressure of 10⁶ Torr using 1500 l/s diffusion pumps operated with liquid nitrogen trapping. Sodium trimers required for the experiment were produced using a supersonic expansion operated under conditions which have been considered in detail elsewhere [3,4]. During the experiments, the typical chamber running conditions ranged from a low of 10⁻⁵ Torr to a high of 10^{-2} Torr. Because of the extremely short radiative



Fig. 1. Chemiluminescent emission resulting from the reaction $Na_3 + X \rightarrow Na_2^* + NaX$ with X = Br, I. Spectra were taken at a resolution of 6 Å. See text for discussion.

lifetime of the Na₂ excited electronic states ($<10^{-7}$ s) considered in this study, nascent state product formation is monitored, even at the upper pressure limits in this system, via the emission of a photon long before subsequent collisions occur. However, at the high pressure limit, self-absorption by unreacted vibrationally excited ground state sodium dimers produced in the emission process was also considered and determined not to be of significance. This is likely due to the high cross section for reaction of vibrationally excited sodium dimers with halogen atoms [5].

In order to produce the spectra depicted in fig. 1, the sodium source, loaded with > 100 g of sodium, was used under conditions which produce sodium atoms, dimers, and trimers, at the upper limit of operation for these experiments, in a ratio of approximately 1:1:0.03 [3,4]. Typical run times to expend the 100 g sample generating the emission spectra depicted in fig. 1 ranged from 1 to 3 h. Experiments in which laser gain was measured ranged in duration from 20 to 40 min, since these experiments were generally conducted at the highest achievable source flux levels. The supersonic beam source, which consists of a rear oven chamber and smaller frontal nozzle, was operated ($T_{oven} \approx 950-1100$ K, $T_{nozzle} \approx$ 1000-1250 K) in a manner which, at the lower operating temperatures, produced an insignificant concentration of the higher sodium polymers Na_n $(n \ge 4)$ which can be obtained from such expansion sources ^{#1}. However, even under conditions producing higher sodium polymer concentrations, the dominant spectral emission features (fig. 1) are readily attributable to the reaction of sodium dimers and trimers.

The beam of sodium vapor formed from the expansion source was directed out of the oven chamber, through a liquid-nitrogen-cooled heat shield to a fluorescence zone in the adjoining reaction chamber, the beam density under conditions used to generate the spectra in fig. 1 ranging from 10^{12} trimer molecules/cm³ in this region to notably higher values for the laser gain studies. The alkali beam reacted with halogen atoms in a merged flow near beam-gas configuration to produce the chemiluminescence depicted in fig. 1. The halogen atoms, primarily bromine and iodine for this particular study, which filled the reaction chamber were produced through ther-

^{#1} All reactions of the alkali atoms and dimers (Na, Na₂) have been studied in this system so as to account for background processes where necessary (see also ref. [1]). It has also been determined that the Na^{*}₂ emission spectrum in fig. 1a cannot be associated with the interaction of effusively or supersonically expanded sodium and CCl_x (x=1-4) compounds formed via reaction of chlorine atoms or molecules with the surface of the hot carbon gun.



Fig. 2. Schematic of (a) apparatus for the study of the chemiluminescent Na_3-X reaction and (b) the arrangement of the experimental configuration from the Na_3-X metatheses. See text for discussion.

mal dissociation of the halogen molecules in a halogen gun-graphite tube furnace, the output from which was directed vertically and perpendicular to the alkali metal flux. Typically, the halogen gun operated at temperatures of ≈ 1600 K ^{#2} as measured by optical pyrometry. Simple halogen molecule sources which will be discussed elsewhere were used for the bromine and iodine molecules – the vapor pressure of bromine is sufficient to produce the required flux to the inlet of the dissociation gun, however, iodine was heated to 70° C in order to maintain the required vapor pressure.

The amount of halogen entering the tube furnace was regulated manually via a valve attached to the halogen reservoir bulb. The flow was adjusted to produce optimum gain conditions. Care was taken not to increase the flow for maximum chemiluminescent intensity, since other competing processes

^{#2} At this temperature the halogen molecules should be >95% dissociated.

produced an intense yellow flame ^{#3} which made the measurement of laser gain impossible by increasing the opacity of the reaction zone. The gain was most clearly defined and optimized over the wavelength range studied thus far when a slightly lower halogen flow rate, which may also provide a slightly more efficient halogen dissociation, was employed.

3. Spectral simulations

We are currently engaged in extensive spectral simulation studies of the A-X, B-X, C-X, and C'-X Na₂ band systems whose emission spectra characterize the Na₃-X reactions. We find that all of the sharp emission features depicted in fig. 1 and associated with the Na₂, B, C, and C' state emission regions cannot be readily explained on the basis of purely dimer fluorescent phenomena which will produce much broader optical signatures. The possibility that the sharp emission features in the Na₂ B-X fluorescence region (fig. 3) signal a population inversion was made evident through a detailed simulation #4 of the Na₂ B-X emission spectrum. As a comparison of the experimental and simulated spectra for the Na₃+Br reaction over this region demonstrates (fig. 3), the intense spectral feature near 5275 Å is unexpected if the fluorescence follows the known Franck-Condon factor profile and the transition moment behavior for the Na₂ B-X emission system [6] #5.

4. Laser gain measurements

In our initial experiments, we wished to establish the probability of a population inversion and measure laser gain in the Na₂ B-X region using an argonion-pumped coumarin 540 dye laser operating in the range 5230-5300 Å. The dye laser was tuned using



Fig. 3. Comparison of (a) observed and (b) calculated emission spectra for the Na₂ B-X emission system. The experimental spectrum corresponds to the chemiluminescence from the Na₃-Br reaction. The calculated spectrum, which was obtained for a rotational temperature, $T_{rot} \approx 1000$ K, represents an estimate of *effective* rotational temperatures in a system operated under single collision conditions and therefore not at equilibrium. Relative vibrational populations input for Na₂ B-X, v' = 0-6 were in the ratio 1.00: 1.17: 1.33: 1.50: 1.58: 1.67: 1.54. See text for discussion.

a home-built [7] three-stage birefringent filter operating at ≈ 0.5 cm⁻¹ resolution (fwhm). Constant dye laser power was maintained using a Spectra Physics model 373 light stabilizer. The laser beam was chopped and split into two branches; one branch was directed into a reference photodiode (B), the other delivered to the alkali-halogen reaction chamber as indicated in fig. 2. The beam entered the vacuum chamber with ≈ 10 mW of power, collimated at a diameter of ≈ 5 mm, and oriented so as to be perpendicular to both the flow of halogen atoms and the alkali vapor beam. After passing through the reaction zone whose length is estimated at between 2 and 4 cm, the beam exited the reaction chamber, was collected by a 50 mm focal length plano convex lens, and was focused onto a second photodiode detector (A). To discriminate against blackbody radiation from the halogen dissociation furnace, the beam was

⁴³ The intense yellow flame is due in large part to the Na D-line emission.

^{#4} This program was written by W.H. Crumley and modified by J.S. Hayden and S.H. Cobb. RKR and Franck-Condon factor programs to perform the calculation input to the simulation program were written by Dr. Brian Wicke.

^{#5} This profile is readily generated using RKR and Franck-Condon factor programs written by Dr. Brian Wicke.

chopped and detected with a lock-in amplifier. The amplifier was operated in the A-B mode, providing the opportunity to detect changes in laser beam intensity occurring only in branch A as the laser beam passed through the alkali-halogen reaction zone. The output from the lock-in was fed to a DSP Technology 2001 digital transient recorder equipped with a model 4100 signal averaging module. The averaged output from this recorder was passed to an IBM PC-XT where the data were monitored and stored.

Laser gain was first observed by setting the dye laser at the wavelength of the most intense feature in the Na₂ chemiluminescence spectrum, 5270 Å (fig. 3). The halogen was manually introduced into the system via a needle valve assembly, and the photodiode detectors monitored changes in the A-B signal strength. This process was repeated several times, each time opening and closing the halogen inlet to initiate and halt the chemical reaction (1) responsible for the Na₂ population inversion. Gains of close to 1% of the input laser power were repeatedly observed at 5270 Å. Proper allignment of the sodium beam, the halogen atom source, and laser beam proved to be critical and great care was taken to ensure that the beam passed directly through the center of the reaction zone. The experiment was repeated several times while varying the size and intensity of the laser beam delivered to the reaction zone. In each case, gains of close to 1% were recorded.

In a typical experimental measurement of the laser gain, the sodium source was brought to a temperature at which it outgassed hydrogen [3]. The source was then brought quickly to those conditions producing a requisite Na₃ concentration so as to employ the $Na_3 + X$ reaction to produce the gain condition. As the temperature of the sodium source was increased prior to the gain measurement, the increasing Na flux produced a decrease in the signal measured by detector A due to scattering or absorption of the laser photons by sodium atoms, dimers, and trimers (see section 5). Laser gain was observed only when a sufficient trimer flux was generated and only when the Na₃+X reaction was initiated. During the laser gain experiments, once the Na₃ concentration was established and gain observed, the sodium source temperature was continually increased to maintain the vapor pressure (flux) required for the



Fig. 4. Wavelength dependence of laser gain measure of superradiance over the range 5230-5300 Å for the Na^{*}₂ product of the Na₃+Br reaction depicted in fig. 3a. See text for discussion.

gain condition. Typical run times ranged from 20 to 40 min.

Measured gains of 1% were consistently recorded under the optimum conditions indicated previously for generating the Na₃ trimer molecule. It is to be emphasized that the gain condition and the spectra in figs. 1 and 3 do demonstrate a perceptible temperature dependence. The sharp fluorescence features increase relative to the Na₂ background fluorescence whose simulation is attainable within the constraints of a non-equilibrium environment (fig. 3). In other words gain reduction can be demonstrated by lowering the source temperature and hence the Na₃ concentration. The gain observed in these systems thus depends both on the Na₃ and on the halogen atom concentration. Further, as the laser power is changed, the photodiode signal (A-B) is found to vary linearly with laser power indicating that we are in the small signal gain regime.

To measure the wavelength dependence of the laser gain in the region 5230–5300 Å, the experiment as described above was performed while tuning the laser to several discrete wavelengths. Fig. 4 depicts the gain monitored at several wavelengths. The observed gain was found to drop off less sharply on the short wavelength side of the maximum, this being associated with the substantial spectral emission feature at 5240 Å and the gain associated with the transition corresponding to this feature. In order to probe additional sharp Na₂ fluorescence features depicted in fig. 1, we have used argon ion pumped coumarin 480 and stilbene 420 dye lasers of similar bandwidth operating in the ranges 4600-5000 Å and 4220-4610 Å respectively. With these dye lasers, we have probed the sharp features at 4920, 4605, 4360, and 4260 Å.

Laser gains of 0.3% and 0.8% were detected at 4920 and 4605 Å respectively. No gain was measured for the notably less intense 4360 and 4260 Å features, indicating that a threshold population inversion level may not have been reached or more likely, a limit in our ability to detect gains less than 0.1%. The 1%, 0.8%, and 0.3% laser gains measured at 5270, 4605, and 4920 Å corresponding to the peaks in the observed sharp fluorescence features are in the approximate ratio of their intensities.

5. Discussion

The laser gain measurements outlined here strongly suggest that the experimentally observed spectral intensity distributions (figs. 1 and 3) can be explained on the grounds that the monitored luminescence in certain spectral regions results from a population inversion. That is, a gain condition due to stimulated emission exists which enhances the intensity of the observed radiative transitions. Thus a population inversion involving certain levels of the Na₂ ground and excited electronic states must be produced by the chemical reaction (1).

There are three important factors which distinguish the Na₃-X reactive system: (1) The cross section for this reaction is extremely high, (2) the cross section for removal of ground state Na₂ molecules is substantial, and (3) the stimulated emission cross section for those excited state Na₂ molecules formed under the reactive conditions outlined, in a near collisionless environment for nascent product excited state formation, may not be limited by the thermal Doppler width but in fact may be associated with a linewidth lying between the thermal Doppler and natural linewidth limited values.

Because of the low Na₃ ionization potential and the high halogen atom electron affinities [5,8] the Na₃-halogen atom reactions are exposed to proceed via an electron jump mechanism with extremely high cross sections certainly exceeding 200 Å² ^{#6}. Thus they have the potential for producing substantial Na₂ excited state populations. We believe that the created population inversion producing the most pronounced effects monitored thus far and involving the Na₂ B state is sustained by the large number of free halogen atoms reacting with the Na₃ molecules in those ground state levels (v'' = 13, 14) on which the lasing transition emanating from the v' = 5, 6 levels of the Na₂ B state terminates. The cross section for reactions of vibrationally excited ground state Na₂ is expected to be comparable to, if not substantially greater than, that corresponding to collision-induced vibrational deactivation of the Na₂ manifold (see footnote 6). In other words extremely efficient reactions rapidly deplete the lower state levels in this system allowing one to sustain a population inversion.

There are several specific comparisons which can be made between the present system and optically pumped sodium dimer laser systems both in heat pipe and supersonic beam configurations. Wellegehausen et al. [2,10] have created several three-level systems by pumping sodium dimer in a heat pipe oven at temperatures approaching 800 K (6.84 Torr). Using the 488 nm line of the argon ion laser operating single mode, these authors excite the pump transition $X^{1}\Sigma_{g}^{+}(3,43) \rightarrow B^{1}\Pi_{u}(6,43)$. Lasing is observed for transitions from the v' = 6 level to levels v'' = 9, 10, 13, 14, 15, and 16 of the Na₂ ground state, the dominant lasing transitions being those to v'' = 13, 14 in the 525-530 nm range. Threshold for this pump cycle has been accomplished with pump powers as low as 1 mW and focused intensities of 1 W cm^{-2} , the observed gain for a 2 W cm⁻² pump being approximately 2×10^{-1} cm⁻¹ [2,10]. By comparison, our focused intensity is 0.05 W cm^{-2} and our observed (maximum) gain at 5270 Å is on the order of $(2-5) \times 10^{-3} \text{ cm}^{-1}$.

At 800 K, the concentration of species in the sodium heat pipe oven is $\approx 10^{17}$ cm⁻³ compared to a total alkali concentration in the present reaction zone which for the laser gain studies reported here is well in excess of 10^{14} cm⁻³. However, the concentration of sodium dimers in the heat pipe is $\approx 1\%$ of the total concentration or 10^{15} cm⁻³ and only 15% of these dimers are in the v=3 level. Thus, the Na₂ concentrations in the heat pipe and the reaction zone of the present experiments differ by less than an order of

^{#6} Given Na₂ and Na₃ ionization potentials of 4.87 and 3.97 eV [9] and an electron affinity of 3.363 eV for atomic bromine, we determine a very substantial electron jump cross section $\sigma \approx \pi [14.38/(3.97-3.36)]^2 = 1746 \text{ Å}^2 (1.75 \times 10^{-13} \text{ cm}^2)$ for the Na₃-Br reaction and $\sigma \approx \pi [14.38/(4.87-3.36)]^2 = 285 \text{ Å}^2$ $(2.85 \times 10^{-14} \text{ cm}^2)$ for the Na₂-Br reaction.

magnitude. Nevertheless, collisional relaxation and thermalization effects dominated by *sodium atom* collisions are expected to play a much more dominant role in the heat pipe configuration than in the present reactively dominated configuration.

Under the typical operating conditions which correspond to the onset of laser gain, 99.75% of the expanded sodium dimers are at an effective vibrational temperature less than 100 K ($T_{\rm vib} \approx 45-50$ K, $T_{\rm rot} \approx 25-30$ K) [3]. At lower operating fluxes, the fraction of sodium dimer molecules which are not effectively expanded and cooled may be in excess of 0.25%. Although the overwhelming number of sodium dimers in the system will be in the v'' = 0 level $(N_{\nu''=3}/N_{\nu''=0}=10^{-3}$ at $T \approx 100$ K), experiments in our laboratory have demonstrated that the very small hot dimer concentration can be pumped to the Na₂ $B^{1}\Pi_{\mu}$ state (figs. 4 and 5 in ref. [3]) with a laser operating over the range 5240-5300 Å. Thus absorption by the small fraction of hot Na₂ molecules which are not effectively cooled in supersonic expansion (fraction decreasing with increasing stagnation pressure) or are collisionally heated in their passage to the fluorescent zone can account for losses in laser power observed as the expansion is brought to the threshold for laser gain. Note, however, that the pump intensities (0.05 W cm^{-2}) are far below that required for optical pumping.

If we envision the $Na_3 + X$ reaction as an efficient pumping source for electronically excited sodium dimers, both the Na₂ optically pumped laser and the present amplifier operate in a three-level configuration. Both schemes face a bottleneck in the lower ground state Na₂ level on which the superradiant transitions terminate. In the heat pipe configuration which operates at much higher pressures, one relies on collisional deactivation on this lower laser level. However, this efficient removal through Na₂-Na collisions ($\sigma \approx 1 \times 10^{-14} \text{ cm}^2$) [2,10] is matched by an even more efficient excited state deactivation cross section $(6.4 \times 10^{-14} \text{ cm}^2)$ which certainly must be considered in the pumping cycle and gain equations. By comparison, the present amplifier relies primarily on the high cross sections associated with alkali-halogen atom chemical reactions to remove the lower metastable sodium dimers formed in the emission process. Collisional deactivation is not a major factor in the present experiments especially for the Na₂ excited states formed in reaction whereas it is tantamount to the continuous wave operation of the optically pumped heat pipe laser systems.

Given a Na₃ concentration in the fluorescence reaction zone well in excess of 3×10^{12} molecules cm^{-3} *⁷, we can estimate a conservative bound to the Na₂ excited state density of 6.3×10^9 cm⁻³ *⁸. If we assume that 10% of the Na₃ reactions (see footnote 6) produce Na₂ in the B state and that 15% of these constitute the B state emitters in the laser gain wavelength region (figs. 3 and 4), the Na^{*}₂ concentration would be $\approx 1 \times 10^8$ cm⁻³. With an experimental amplification length of 2–4 cm, a laser gain of 10^{-2} (1%) at 5270 Å thus corresponds to a maximum cross section $\sigma \approx (2.5-5) \times 10^{-11}$ cm² *⁹.

It must be emphasized that this cross section is associated with the region of peak gain probed in the present experiments whose focus has been the Na₂ B state region. Further, the Na₂ excited state concentration used to estimate this concentration is based upon a Na₃ concentration of 3×10^{12} cm⁻³, a lower bound for the laser gain experiments.

The Doppler-limited stimulated emission cross section for a thermalized Na₂ B state emitter is between 1×10^{-12} and 3×10^{-12} cm² for typical temperature ranges [2,11]. As Byer and co-workers [11] have noted, the cross section is very large even compared to the 10^{-16} cm² rhodamine 6G dye laser cross section and the sodium dimer laser represents an extremely efficient system. While it might be plausible to ascribe the difference between our *maximum* cross section at 5270 Å and the Doppler-limited value to an underestimate of concentration, we suggest an alternate possibility associated with the near collisionless environment in which the excited states of sodium dimer produced in the non-equilibrium reactive encounter are formed.

In a strictly collisionless environment, the limiting linewidth may be taken as the natural linewidth, approximately 50 times smaller than the thermalized Doppler width. While a determination of the emission lineshape associated with the current experi-

^{#7} The concentration at the onset of readily measurable laser gain.

⁴⁸ From the estimate $[Na_2^*] \leq \tau_{rad} [Na_3] v_{flow}/(diameter of reaction zone) with <math>\tau_{rad} = 7 \times 10^{-9}$ s, $[Na_3] = 3 \times 10^{12}$ cm⁻³, $v_{flow} = 1.5 \times 10^5$ cm s⁻¹, d = 0.5 cm.

^{#9} From $\sigma \approx (\text{measured gain})/[\text{Na}_2^* B^1 \Pi_u](\text{gain length}) = 10^{-2}/(1 \times 10^8) \ (L=2-4 \text{ cm}).$

ments is not at present feasible, we assert that the appropriate linewidth limit lies between the thermalized Doppler and natural linewidths. The magnitude of our estimated maximum cross section is well within expected bounds. In fact, the estimated cross sections are not surprising, for Bergmann, Wellegehausen and co-workers [12] have succeeded in optically pumping a sodium dimer supersonically expanded molecular beam.

In the absence of collisional losses with an intensity of 1 W cm⁻² [2,10] and an absorption coefficient $\alpha_p = 10^{-1}$ cm⁻¹ [2,10], the optically pumped sodium dimer laser can produce $\approx 1.79 \times 10^9$ Na^{*}₂ cm⁻³. If we consider both deactivation by spontaneous emission and collisions and take $G=5 \times 10^{-2}$ cm⁻¹ [2,10] with a 2.5 W cm⁻² pump we determine $\sigma \approx 6 \times 10^{-11}$ cm^{2 #10}. It is noteworthy that the number of excited Na₂ B state molecules formed in the reactive process and with a 1 W cm⁻² optical pump are comparable as is our maximum cross section at 5270 Å for stimulated emission and that corresponding to the optical pump conditions outlined.

At the resolution (6-10 Å) which must be used to record the fluorescence spectra depicted in figs. 1 and 3, it is not possible to discern how many rotational levels are associated with each of the sharp fluorescence features. Certainly the laser used in these experiments, operating at a resolution of 0.5 cm^{-1} , can interact with more than one rotational transition. However, the sharp nature of the observed fluorescence features at the resolution used to obtain the spectra depicted in figs. 1 and 3 indicates that a limited number of rotational levels are involved [13] and that the rotational dilution may well correspond to a factor much less than 10^2 .

In carrying out the present gain measurements, we are of course sampling a volume region which is difficult to define precisely. However, we feel that the ratio of the gains for the present system and those associated with the Na₂ optically pumped laser [2,10] are significant. It would appear that our gain for the currently unoptimized system is a factor of between

*10 From $\sigma \approx (5 \times 10^{-2} \text{ cm}^{-1})(7 \times 10^8 \text{ s}^{-1})(6.324 \times 10^{-20})$ (2 π)/(10⁻¹ cm⁻¹)(2.5 W cm⁻²) \approx (G)(γ_3)($h\omega_p$)/ $\alpha_p I_p$. Here we have used the expressions given by Wellegehausen in ref. [2], γ_3 based on collisional deactivation in the B state [10] and the observed gains and pump powers recorded in table 1 [2]. 20 and 100 smaller than that reported for the single mode 488 nm optical pump [2,10]. This ratio seems reasonable in view of the cross sections for stimulated emission determined for the optically [2,10]and reactively pumped amplifiers and the rotational dilution for the reactively pumped system which we estimate from the experimental data.

We hope to extend our studies of the sharp Na₂ fluorescence features which encompass the visible and ultraviolet spectral regions. Based on preliminary data obtained on the basis of spectral simulations, we feel that it may be possible to generate additional laser amplifiers, the most attractive of these being those in the regions close to ≈ 4125 , ≈ 3950 , ≈ 3650 , and ≈ 3500 Å.

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