Wave Mixing and Amplified Spontaneous Emission in Pure Potassium and Mixed Sodium-Potassium Vapors

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Abstract. We report observations of several wave-mixing and stimulated processes in pure potassium and mixed sodium-potassium vapors which are excited by a pulsed laser operating in the range 680–800 nm. When the laser is tuned to the potassium two-photon $4S \rightarrow 6S$ transition, we observe stimulated emission on the various cascade transitions as well as four- and six-wave mixing. When the laser is tuned over the range 747 to 753 nm, which is well away from all atomic transitions, we observe strong forward and weak backward emission at the potassium $3D_{3/2} \rightarrow 4P_{1/2}$ transition wavelength (1.17 µm). However, this latter emission is only observed in the mixed sodium-potassium vapor. We present data on the excitation spectrum, forward to backward asymmetry, temporal dependence, and the laser power dependence of this emission. We speculate that two-photon photodissociation of the NaK molecule is responsible for this emission.

The last few years have seen a rapid growth in the number of studies involving non-linear optical processes in alkali vapors. Included in this are studies of purely atomic processes such as parametric four- and six-wave mixing, amplified spontaneous emission, optically pumped stimulated emission, and stimulated Raman and hyper-Raman scattering [1–18]. Additional coherent processes involving collisional energy transfer or photodissociation of alkali molecules have also been observed [19–25].

In the present work, we present experimental observations of various non-linear processes occurring in pure potassium or mixed sodium-potassium vapors excited by a high-power, pulsed laser operating in the range 680 to 800 nm. In particular, when the laser is tuned to either the potassium $4S \rightarrow 6S$ or $4S \rightarrow 4D$ two-photon transition, we observe amplified spontaneous emission on the cascade atomic lines, as well as several four- and six-wave mixing processes. Figure 1 presents partial energy level diagrams of potassium showing in schematic form some of the processes observed. The

~404 nm four-wave mixing signals, which are nearly resonant with the potassium $5P \rightarrow 4S$ transitions, were to the best of our knowledge first observed by Lumpkin [1] and Barak et al. [2]. In those experiments an off-resonant, two-step excitation scheme was used, and the $5P$ levels populated by a stimulated Raman process. Agostini et al. [3] observed similar four-wave mixing near 404 nm when pumping potassium vapor with a dye laser tuned to the two-photon $4S \rightarrow 4D$ transitions. Analogous processes have since been observed in sodium [4, 5], rubidium [6], and cesium [7]. Processes such as the optically pumped stimulated emission $(6S \rightarrow 5P)$ and cascade amplified spontaneous emissions $(5P \rightarrow 5S, 5P \rightarrow 3D, 5S \rightarrow 4P, \text{ and } 3D \rightarrow 4P)$ are created by population inversions which occur as the atoms cascade down through their various energy levels. Analogous processes have been observed in all of the alkalis under a wide variety of excitation conditions [4, 9–18]. Finally, when pumping the potassium $4S \rightarrow 6S$ transition we observe coherent forward emissions at 1.28 and 1.37 µm. By comparison
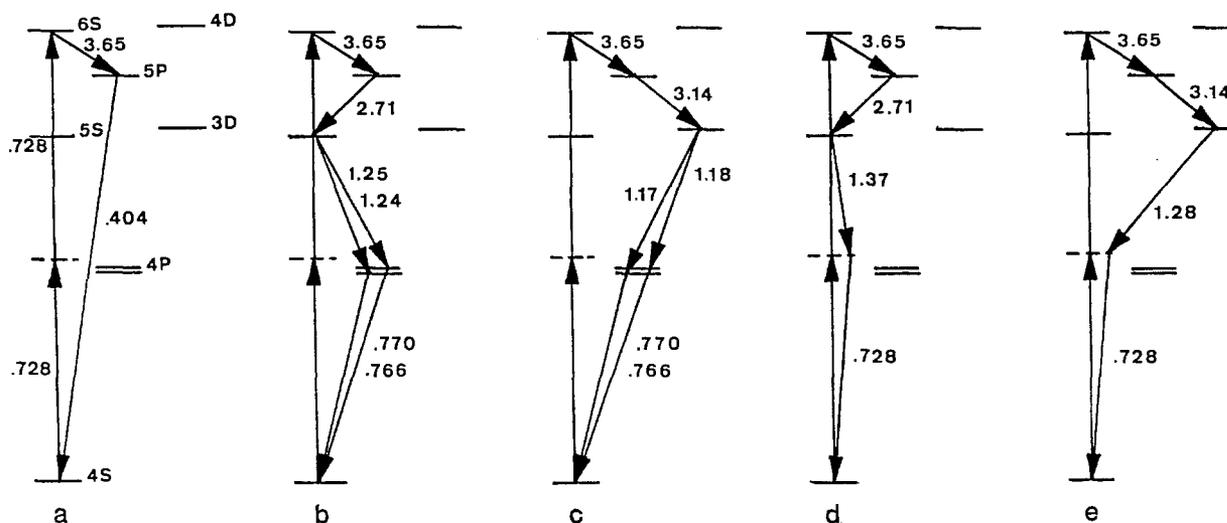
Fig. 1. Partial energy level diagrams of potassium showing the processes observed when the laser is tuned to the potassium 4S→6S two-photon transition. (a) Four-wave mixing near the 5P→4S transition wavelength. (b, c) Amplified spontaneous emission on the cascade transitions. (d, e) Six-wave mixing processes

with analogous processes in Na vapor [18], we can identify these as six-wave mixing processes. Our data on these various processes are presented in Sect. 2.

While observations of these types of atomic stimulated and wave-mixing processes are now becoming fairly commonplace, more complicated processes involving the diatomic molecules in the vapor are less well understood. In particular, several observations of atomic stimulated emissions have been made when alkali transitions coupling two excited states have been pumped with high power lasers [19–23]. The lower state of the transition (typically the sodium 3P or the potassium 4P state) is in each case excited by either one photon dissociation of the diatomic molecule or by collisional excitation transfer from the molecule. Another source of stimulated emissions on atomic transitions is due to two-photon photodissociation of molecules into one ground state and one excited state atom. Population inversions can then exist between the excited level and lower unpopulated states. Such a "photodissociation laser" was observed in mercury vapor in 1977 by Komine and Byer [26]. Similar processes were observed more recently in sodium [5, 22] and potassium [19, 21]. The work of Wang and coworkers [19, 21] is of particular interest in connection with our present findings. Finally, there have been recent reports of stimulated emissions on diffuse molecular bands following excitation of atomic transitions [24, 25].

In this paper we also report observations of an interesting process where strong forward emission at the potassium 3D→4P transition wavelength is detected when sodium-potassium mixtures are pumped

over a broad wavelength range near 750 nm. Since this process cannot be observed in pure potassium vapor, we suspect that either Na_2 or NaK molecules are involved. Since Na_2 absorbs only weakly in this wavelength region, we believe we are observing two-photon photodissociation of NaK. Using the known well depth of the NaK ground state, [27] we find that we have just sufficient energy to reach the $Na(3S) + K(3D)$ dissociation limit from the $v = 1$ level of the ground state well. However, the 3D→4P emission appears primarily in the forward direction which may indicate that either a parametric wave mixing process is involved, or that the gain is asymmetric. The excitation spectrum for this 3D→4P emission shows a broad hump centered near 750 nm which is similar to the broad hump centered on 640 nm observed by Wang et al. [19] in the excitation spectrum for potassium 5P→5S and 5P→3D emission. In the present case, the 750 nm hump has a series of sharp dips superimposed on it. We found that these dips are the result of depletion of the potassium 3D state population by laser-induced transitions to Rydberg F levels. Details concerning these observations are presented in Sect. 2.

1. The Experiment

The experimental apparatus is shown in Fig. 2. A vapor containing a mixture of sodium and potassium was contained in a 4-way cross, stainless-steel, heat-pipe oven. The initial mixture of potassium and sodium was ~2 to 1 although temperature cycling and subsequent reloadings caused the ratio of potassium to sodium
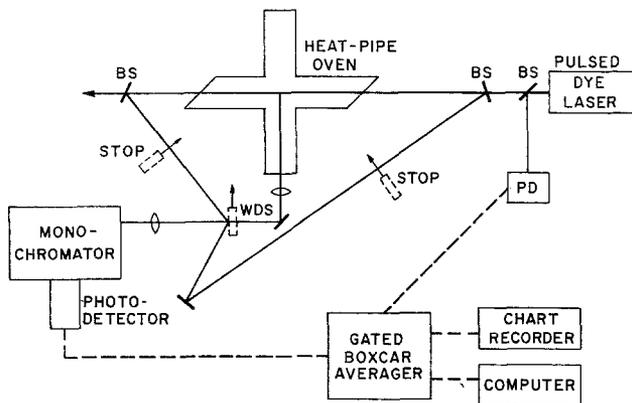
Fig. 2. Experimental set-up. In the figure, PD, BS, and WDS represent photodiode, beam splitter and white diffusing surface, respectively. The white diffuser can be translated out of the way so that side fluorescence can be imaged onto the monochromator slits

atoms in the vapor phase to vary with time. The oven pressure (determined by the argon buffer gas pressure) was varied over the range 1.3 to 9 Torr, while the temperature range studied extended from 385 to 440 °C. To verify the molecular species involved in the non-resonant process, an oven containing pure potassium was occasionally substituted for the Na–K oven without adjusting any of the associated optics.

The vapor was excited by a pulsed, frequency-doubled Nd:YAG laser pumped dye laser. The dye LDS 765 was used in the range 740 to 810 nm, while LDS 722 was used to cover the range 680 to 760 nm. Peak laser powers of ~1 MW at 750 nm were typical. In most cases the laser beam entering the oven was unfocused.

Fluorescence emitted perpendicular to the pump laser axis was monitored through a cross-arm of the heat-pipe oven. The light was resolved using a 0.22 meter monochromator in conjunction with a liquid nitrogen cooled intrinsic germanium detector. Long-pass, color-glass filters were used to eliminate signals due to second order scattering from the diffraction grating. Light emitted approximately parallel (which we will call forward emission) or antiparallel (backward) to the pump laser beam could be directed to the same monochromator by a set of partially reflecting and highly reflecting mirrors. In these cases the intense emissions were scattered off a white diffusing surface in front of the monochromator. The white diffuser was positioned on a translating mount so that forward, backward and side scans could be taken without changing the optical alignment. Note that since collimating optics were not used in the forward and backward directions, only coherent emissions could be observed in those directions. For observations in the

forward direction, the laser beam was always blocked with an appropriate color-glass filter before the white diffuser. Calibration of the forward to backward detection efficiency was obtained by first observing the unblocked laser beam in the forward direction (with the oven cold), and then retroreflecting the beam and observing in the backward direction. In this manner it was found that the forward to backward detection efficiency was ~1.

Several types of measurements were performed. For the first type of measurement, the monochromator was set to pass either the potassium $3D_{3/2} \rightarrow 4P_{1/2}$ emission at 1.17 μm or the $3D_{5/2} \rightarrow 4P_{3/2}$ emission at 1.18 μm. The dye laser was then tuned through part of the dye profile and the resulting atomic emission monitored in the forward, backward, or side directions. These we call excitation scans. Measurements of this type were carried out for a range of pressures and temperatures.

In the second type of measurement, the dye laser was set to a particular wavelength and the monochromator scanned, usually from 800 to 1700 nm. These we call emission scans. Since an intrinsic germanium detector was used most of the time, sensitivity was poor below 800 nm although strong emission near 404 nm corresponding to $5P \rightarrow 4S$ could be weakly detected.

In some cases the germanium detector was replaced with a photomultiplier tube (S-1 cathode) or an indium-antiminide detector, so that observations could be made at shorter or longer wavelengths, respectively. Using the three detectors, the entire spectral range between 0.4 and 4 μm could be monitored. Additionally, since the photomultiplier has an ~1 ns time response and a small, but adequate, sensitivity at 1.17 μm, it could be used to study time-dependences of the $3D \rightarrow 4P$ emissions. The angular divergence of the potassium $3D \rightarrow 4P$ forward emission was studied using an iris diaphragm.

Finally, the power dependences of the $3D \rightarrow 4P$ and other emissions were determined using a series of neutral density filters placed between the dye laser and the heat-pipe oven. The filters were calibrated in place using the photodiode shown in Fig. 2.

2. Results and Discussion

Figure 3 shows the near-infrared forward and backward emission scans when the laser wavelength in air was set to 728.36 nm, which corresponds to the potassium $4S \rightarrow 6S$ two-photon transition. Here it can be seen that the atomic $3D_{3/2} \rightarrow 4P_{1/2}$ (1.17 μm), $3D_{5/2} \rightarrow 4P_{3/2}$ (1.18 μm), $5S_{1/2} \rightarrow 4P_{1/2}$ (1.24 μm), and $5S_{1/2} \rightarrow 4P_{3/2}$ (1.25 μm) emissions are all seen in both the forward and backward directions. (In all cases the
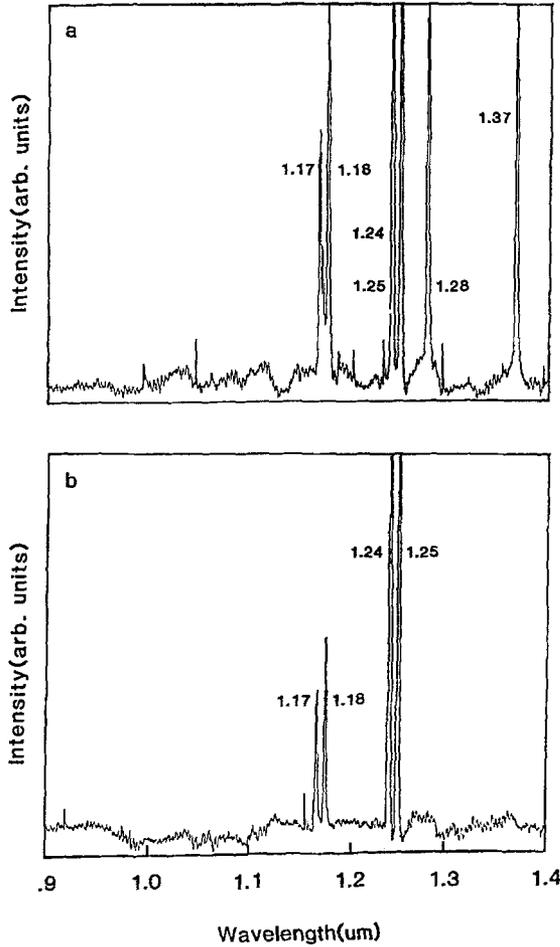
Fig. 3. (a) Forward and (b) backward near-infrared emission scans obtained when the laser was tuned to the potassium $4S \to 6S$ two-photon transition in the mixed sodium-potassium vapor. The 1.28 and 1.37 μm emissions in the forward direction correspond to six-wave mixing processes while the 1.17/1.18 and 1.24/1.25 μm doublets correspond to potassium $3D \to 4P$ and $5S \to 4P$ transitions, respectively

forward to backward intensity ratios are in the range between 1 and 2.) These lines clearly represent amplified spontaneous emission (ASE) resulting from population inversions that exist as the atoms cascade down through their energy levels in allowed transitions.

The two lines at 1.28 and 1.37 μm are only observed in the forward direction. These emissions, which have also been observed at Oak Ridge [28] are analogous to emissions at 795 and 840 nm observed in sodium vapor pumped at the two-photon $3S \to 5S$ and $3S \to 4D$ transitions, respectively [18]. From the explanation of [18] and the diagrams in Fig. 1d and e, it can be shown that these lines represent six-wave mixing processes involving three photons at the laser wavelength and two near-infrared photons produced in the vapor by stimulated hyper-Raman scattering and ASE. Due to

phase-matching conditions, these emissions are only in the forward direction. A side emission scan verified that although the various atomic lines and molecular emission could be clearly seen, the 1.28 and 1.37 μm emissions were absent as expected.

A photomultiplier tube was used in place of the germanium detector so the emission scans could be made down to 400 nm. Near 404 nm, very strong forward emission was observed. No emission near this wavelength could be detected in the backward direction. This emission, which has been studied extensively in the past [1-3, 28], represents a parametric four-wave mixing process.

These various processes observed when pumping the two-photon transition at 728.36 nm can be summarized by the following set of equations:

4-wave mixing

$$\omega_{5P \to 4S} = 2\omega_{laser} - \omega_{6S \to 5P(4D \to 5P)}, \quad 2\omega_{laser}$$
$$= \omega_{4S \to 6S(4S \to 4D)}$$

6-wave mixing

$$\omega_{1.28(1.37)} = 2\omega_{laser} - (\omega_{6S \to 5P} + \omega_{5P \to 3D(5P \to 5S)} + \omega_{laser})$$

amplified spontaneous emission

$$\omega_{3D \to 4P(5S \to 4P)} = 2\omega_{laser}$$
$$- (\omega_{6S \to 5P} + \omega_{5P \to 3D(5P \to 5S)} + \omega_{4P \to 4S}).$$

Measurements of the angular divergences of these emissions for the laser tuned to 728.36 nm were not carried out in the present experiment. However, Wang et al. [18] found that the analogous emissions in sodium display roughly the same angular divergence as the pump laser beam. We note also that apart from effects due to pressure and temperature differences in the two ovens, results obtained with the pure potassium and the sodium-potassium mixtures were essentially identical when pumping at 728.36 nm.

When we replaced the germanium detector with the indium-antimonide detector, several other coherent emissions could be observed. These include infrared emissions at 3.65, 3.14, and 2.71 μm which correspond to the $6S \to 5P$, $5P \to 3D$, and $5P \to 5S$ transitions, respectively. The $6S \to 5P$ emission appeared in both the forward and backward directions with an intensity ratio of 3:8, while the $5P \to 3D$ and $5P \to 5S$ emissions, which compete with the $5P \to 4S$ emission at 404 nm, were observed with a forward to backward intensity ratio of ~10:1.

Raising the pressure in the heat-pipe oven we observed a change in the 1.17 and 1.18 μm emission when pumping at 728.36 nm. At low temperatures and pressures, these emissions appeared at 1.169 and 1.177 μm and were observed with approximately equal intensities in the forward and backward directions. At

higher temperatures and pressures a third emission at 1.172 µm, which was only observed in the forward direction, began to build in. As the temperature and pressure were raised still further, the 1.172 µm emission was the only one of the three that could be observed. This observation represents a competition between amplified spontaneous emission and a parametric six-wave mixing process. This competition is strongly influenced by phase-matching conditions involving the anomalous dispersion near the transition frequency. This in turn is influenced by the temperature, pressure and composition of the vapor. For example, under our conditions the 1.169 and 1.177 µm lines were difficult to produce clearly in the pure potassium heat-pipe but easily observable in the mixed vapor oven.

In the course of investigating the various coherent emissions described above, we also recorded several laser excitation spectra. We found that when the laser was tuned away from the two-photon $4S \rightarrow 6S$ and $4S \rightarrow 4D$ transitions we could still observe strong forward and weak backward emission on the potassium $3D_{3/2} \rightarrow 4P_{1/2}$ transition at 1.17 µm, and under certain conditions we could also see weak emission at 1.18 µm corresponding to the $3D_{5/2} \rightarrow 4P_{3/2}$ transition. These emissions could only be observed in the heat-pipe oven containing the sodium-potassium mixture. No forward emission could be observed in the pure potassium oven when the laser was tuned away from the atomic transitions despite extensive searches over our range of pressures, temperatures and laser wavelengths.

Figure 4 shows several excitation spectra for the $3D_{3/2} \rightarrow 4P_{1/2}$ forward emission taken in the Na–K oven at various pressures. In this case the dye LDS 765 was used. Excitation spectra obtained with the dye LDS 722 over the same wavelength range were quite different and will be discussed below. It can be seen from Fig. 4 that the 1.17 µm emission can be produced over a broad range of pump wavelengths between 747 and 753 nm. Although each excitation spectrum appears to be a set of spikes, it really consists of a broad hump with a series of sharp dips superimposed. The dips can be easily identified as occurring at wavelengths corresponding to $3D_{3/2} \rightarrow nF_{5/2}$ transitions. Here n is the range 26 through 29. Under conditions where forward emission at 1.18 µm can be observed, the excitation spectra reveal a series of sharp dips corresponding to $3D_{5/2} \rightarrow nF_{7/2}$ transitions. In each case it is clear that the emission intensity is being reduced by the coincidental pumping of the upper state of the forward emission to a high-lying Rydberg F level.

Under the same conditions as those that produced Fig. 4, weak backward emission at 1.17 µm could also be observed. The ratio of forward to backward emission at 1.17 µm was on the order of 25:1.
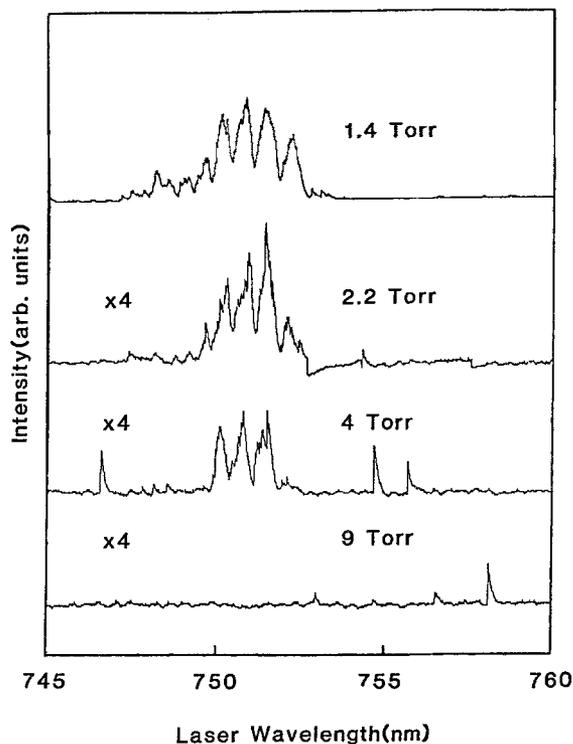
Fig. 4. 1.17 µm (potassium $3D_{3/2} \rightarrow 4P_{1/2}$) excitation spectra obtained at several pressures in the mixed sodium-potassium vapor. The extraneous spikes at ∼747, 755, and 756 nm in the 4 Torr scan, and at ∼758 nm in the 9 Torr scan are not reproducible and are due to cosmic rays to which the germanium detector is sensitive

Figure 5 shows emission scans in the forward and side directions for laser excitation at 751.6 nm. It can be seen that the forward 1.17 µm emission is stimulated since spontaneous emission (as observed in the side direction) produces more light at 1.18 than at 1.17 µm. The spontaneous emission spectrum also shows K_2 and NaK molecular emission near 1 µm and potassium $5S \rightarrow 4P$ emission at 1.24 and 1.25 µm.

Time dependence measurements shown in Fig. 6 reveal that the forward 1.17 µm light is produced during the laser pulse duration of ∼8 ns, while the side direction spontaneous emission is recorded over a period greater than 100 ns, which is characteristic of the $3D$ state lifetime (∼40 ns) lengthened by radiation trapping. Using the three available detectors, we conducted a search for other forward emissions in the wavelength range 0.4 to 4 µm, but no other emissions were observed.

Figures 4 and 7 show the effects of pressure and temperature on the 1.17 µm forward emission. As can be seen, increasing temperature enhances the emission, while increasing pressure quenches the process.

Figure 8 shows the dependence of the 1.17 µm emission on laser power. Clearly the emission displays a

Fig. 7. Temperature dependence of the 1.17 μm (potassium $3D_{3/2} \rightarrow 4P_{1/2}$) forward emission observed when the mixed sodium-potassium vapor is excited over the range 745 to 760 nm. The pronounced dips in the emission hump correspond to depletion of the $3D_{3/2}$ level by laser excitation to Rydberg $nF_{5/2}$ levels (with n in the range 26 to 30)
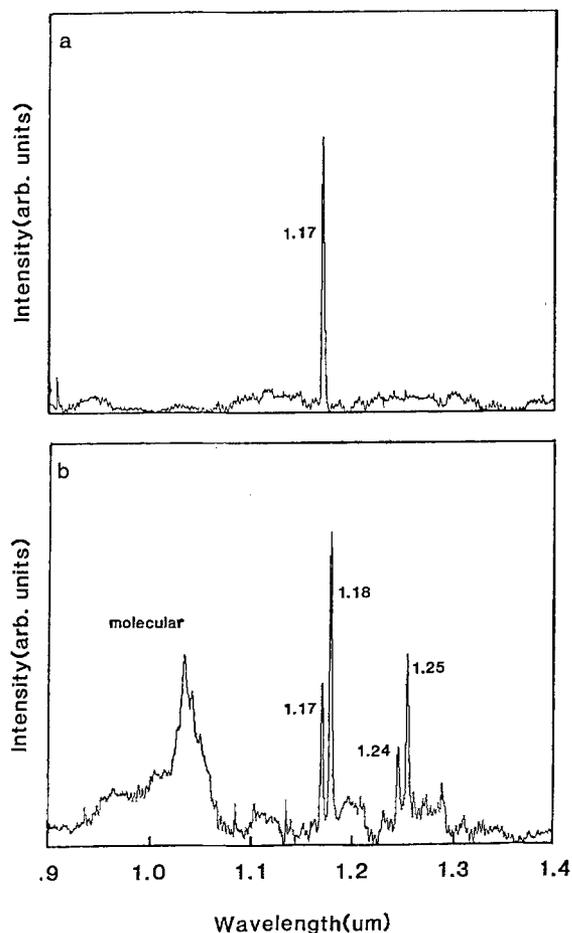
Fig. 5. (a) Forward and (b) side near-infrared emission scans obtained when the laser was tuned to 751.6 nm in the mixed sodium-potassium vapor. The molecular emission observed in the side direction between 0.95 and 1.05 μm is due to NaK and K_2 $A^1\Sigma \rightarrow X^1\Sigma$ transitions
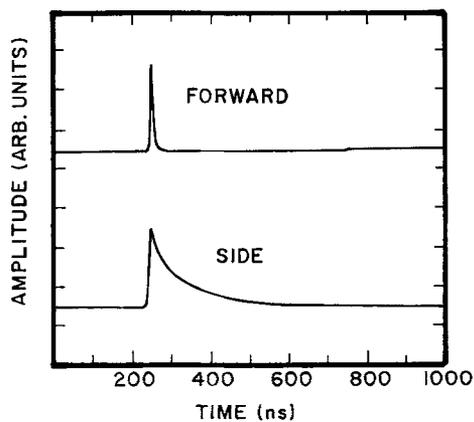
Fig. 6. Time dependence of the forward and side emission at 1.17 μm. The forward emission has a full width half maximum of ∼8 ns which corresponds to the laser pulse duration. The side emission persists for more than 100 ns which represents the 3D state natural lifetime of ∼40 ns lengthened somewhat by radiation trapping

Fig. 8. Laser power dependence of the 1.17 μm emission when the mixed vapor is excited at 751.6 nm. Laser power of 100 corresponds to full laser power (approximately 1 MW) in an unfocused beam of ∼0.1 cm diameter

threshold behavior. Above threshold the emission follows a linear response with some indication of saturation at high intensities. Finally, the angular divergence of the 1.17 μm emission produced with LDS 765 dye pumping of the NaK vapor at 751.6 nm was studied using an iris diaphragm. These studies showed that the 1.17 μm emission has a somewhat larger angular divergence than the pump laser beam.

We interpret these results as indicating that two-photon excitation of the NaK molecule populates a state that can either directly dissociate or predissociate to produce excited potassium atoms in the $3D_{3/2}$ state; i.e.

$$NaK(X^1 \Sigma) + 2h\nu \rightarrow NaK(?) \rightarrow K(3D_{3/2}) + Na(3S).$$
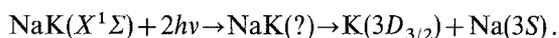
We identify the important molecular species as NaK by a process of elimination. K_2 can be ruled out by the absence of the emission in pure potassium vapor and Na_2 is an unlikely candidate since it is a poor absorber near 750 nm and a collisional step would also be required to populate $K(3D)$ in that case. The requirement of sodium in the oven makes it difficult to imagine that the process could be strictly atomic, although some hybrid sodium-potassium collisional mechanism might be invoked. However, the molecular dissociation mechanism is further supported by the absence of other stimulated infrared emissions. Two laser photons would carry the potassium atom to well above the $5P$ level, which could be populated by stimulated hyper-Raman scattering or by collision from some excited state of sodium. However, any such mechanism would be likely to produce stimulated emission on either the $5P \rightarrow 3D$ or $5P \rightarrow 5S$ transitions. Such emissions were observed by Sorokin and Lankard [8] pumping at 694.3 nm, and also by Wang et al. [19] who used laser excitation over the range 600 to 700 nm. Because of the binding energy of the NaK ground state, it is not possible to reach $5P$ through photodissociation of the molecular states. However, the $K(3D) + Na(3S)$ dissociation limit can just be reached from low lying rovibrational levels of the NaK ground state with two laser photons. Collisional excitation transfer from molecules to atoms (i.e. $NaK^* + K \rightarrow NaK + K^*$ where NaK^* and K^* represent an excited molecule or atom, respectively) cannot be entirely ruled out. However, collisions would be expected to occur over a time much longer than the laser pulse, and would be expected to populate other levels such as $5P$, $5S$, and $3D_{5/2}$ just as strongly as $3D_{3/2}$. Photodissociation, on the other hand, can be a very selective process. In cesium, for example, the $5D_{3/2}$ state is found to be populated more than twice as efficiently as $5D_{5/2}$ when Cs_2 is photodissociated over the pumping range 460–520 nm, [29] and the $6P_{3/2}$ state is 200 times more strongly populated than $6P_{1/2}$ over the range 560–620 nm [30].

Both the forward to backward intensity ratio and the larger angular divergence of the 1.17 μm emission compared to the pump beam argue that the observed process involves some type of wave-mixing. However, it is hard to reconcile the wave-mixing idea with a photodissociation process. Photodissociation converts some excitation energy into kinetic energy so it is difficult to imagine how the photon energies could add up as required in wave-mixing. In addition our studies indicate that no other nearly collinear photons are emitted in the range 0.4 to 4 μm. It is impossible to devise a four- or six-wave mixing process involving two laser photons and one photon at 1.17 μm that does not also produce at least one other photon in this 0.4 to 4 μm range. Nevertheless, it is possible to imagine that a population inversion between $3D_{3/2}$ and $4P_{1/2}$ could be produced by photodissociation, and the subsequent amplified spontaneous emission could trigger a wave mixing process involving two laser photons, one 1.17 μm photon, one $4P_{1/2} \rightarrow 4S$ photon at 769.9 nm (which is highly trapped in the optically thick vapor and therefore cannot be observed), and several IR photons beyond 4 μm. This would explain the small emission in the backward direction as a residual of the ASE, while most of the forward emission would result from the wave-mixing process. An alternate explanation is that we are observing a simple photodissociation produced lasing which has a strong forward to backward gain asymmetry. This type of gain anisotropy is observed in alkali dimer lasers, for example [31, 32]. However, it should be emphasized that at this point all such discussion is primarily speculation, and remains to be fully tested.

It was mentioned above that pumping the sodium-potassium vapor using the dye LDS 722 produced a very different excitation spectrum at 1.17 μm than that obtained with the dye LDS 765 over the same wavelength range. The LDS 722 excitation spectrum is shown in Fig. 9. Not only is the intensity of the 750 nm hump approximately 12 times larger than the corresponding LDS 765 peak (even though we had less power at 750 nm using LDS 722), but also a weaker hump centered about 741 nm and a very weak hump near 730 nm can now also be seen. Because of this surprising discrepancy, we carried out detailed measurements of the spectral composition of the laser pulse for each of the two dyes. When the laser wavelength was set to 751.6 nm, the LDS 722 dye is working near the edge of its gain profile. Under such conditions background ASE from the dye is expected to be relatively strong. We found that in addition to the main laser peak, the LDS 722 produced a 15 nm wide lobe located 30 nm to the blue of the main peak which

Laser Wavelength(nm)
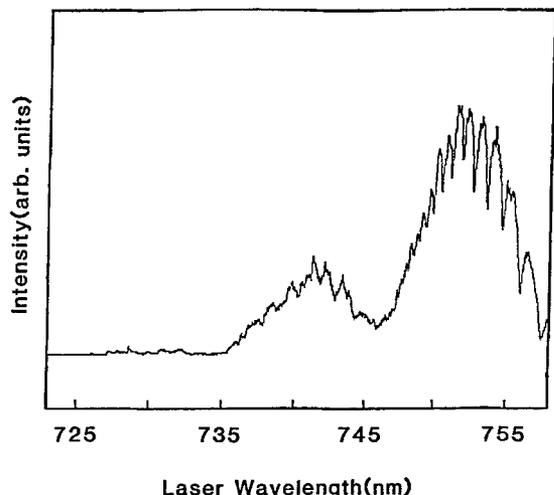
Fig. 9. 1.17 μm forward excitation spectrum obtained in the mixed vapor using unfiltered LDS 722 dye laser pumping. In this case the dye laser emission is contaminated by an ASE lobe located at ~720 nm and containing approximately 0.5% of the total laser intensity. Although the intensity scales in this figure and Figs. 4 and 7 are arbitrary, our measurements indicate that the peak intensity is roughly a factor of 12 greater when the ASE lobe is present. Additionally, a second hump in the excitation spectrum can now be seen centered on 741 nm and a third very weak hump can just barely be seen near 730 nm. The dips on the main hump correspond to depletion of the $3D_{3/2}$ level by pumping to $nF_{5/2}$ levels with n in the range 21 to 32

contained approximately 0.5% of the total intensity. A similar, but 20 times smaller, lobe was observed approximately 25 nm to the long wavelength side of the main peak. When we used the dye LDS 765, the short wavelength lobe was not observed although the long wavelength lobe was still present.

It was clear that the short wavelength lobe had a profound effect on the 1.17 μm excitation spectrum. To demonstrate this we used a set of long pass filters to prefilter the laser beam before the heat-pipe oven. When a Schott RG695 filter was used in the LDS 722 beam, no change in the excitation spectra was seen other than that due to the power loss from reflections at the glass surfaces. However when an RG780 filter was used, the 741 nm hump disappeared from the excitation spectra and the 750 nm peak was reduced by a factor of 7. In fact the filtered LDS 722 spectrum closely resembled the LDS 765 spectrum, but the latter was ~0.6 times as intense. A look at the filtered laser spectrum showed that the short wavelength lobe was reduced in intensity by ~95% over the unfiltered case, while the main laser peak was reduced by only a factor of 2. Filtering the LDS 765 output had little effect on the results. Note that the 1.17 μm output pumping at 750 nm with LDS 722 dye is much stronger than the same wavelength emission when pumping the two-

photon $4S \rightarrow 6S$ atomic transition at 728.36 nm. The latter can just barely be seen in Fig. 9.

Another interesting but as yet unexplained result is that the dips appearing in the LDS 722 excitation spectrum near 750 nm (which we have identified as corresponding to $3D_{3/2} \rightarrow nF_{5/2}$ transitions) are more pronounced in the backward than in the forward direction.

The data from LDS 765 shows that two 750 nm photons are sufficient to produce strong forward emission at 1.17 μm. However the additional presence of even a very weak source near 720 nm enhances the process dramatically. Note that the LDS 722 dye excitation spectrum (Fig. 9) demonstrates that 2 photons at 720 nm are not, by themselves, particularly efficient at producing 1.17 μm light. No other atomic lines were observed in the unfiltered LDS 722 case, indicating that the enhancement due to the 720 nm ASE lobe is not simply due to a resonant atomic process. Note also that the data presented in our previous paper [33] were all taken with LDS 722 dye with the contaminating ASE lobe at 720 nm. Thus the earlier published results were taken under the conditions of the present Fig. 9 and therefore are quite different from the results presented here in Figs. 4 and 6.

The conclusion from these studies using the filtered and unfiltered output from the LDS 722 dye laser is that one or the other step in the two-step NaK molecular excitation is greatly enhanced near 720 nm. Since the NaK $A^1\Sigma^+ \leftarrow X^1\Sigma^+$ absorption peaks near 750 nm (according to the potentials of Refs. [27] and [34]), we expect that it is the second step which is enhanced. In particular, the $v = 0$ level of the ground state cannot be pumped to the $K(3D) + Na(3S)$ dissociation limit with two 750 nm photons, but can be with one 750 and one 720 nm photon. The final explanation of these processes can only be obtained by two-laser experiments which are now underway in our laboratory.

3. Conclusions

In this paper we have reported our observations of forward and backward stimulated emission in pure potassium and mixed sodium-potassium vapor excited in the range 680–800 nm. We have found that when either vapor is excited at 728.36 nm, corresponding to the potassium $4S \rightarrow 6S$ two photon transition, we observe optically pumped stimulated emission on the $6S \rightarrow 5P$ transition. In addition we observe amplified spontaneous emission on cascade transitions, and four- and six-wave mixing signals which are nearly resonant with the $5P \rightarrow 4S$, $5S \rightarrow 4P$, and $3D \rightarrow 4P$ transitions. These results for resonant excitation are

analogous to previous results obtained in other alkalis as well as in potassium.

We have also found that when the sodium-potassium vapor is excited off resonance over the range 747–753 nm we can observe stimulated emission at 1.17 μm (potassium $3D_{3/2} \rightarrow 4P_{1/2}$). This emission, which cannot be observed in pure potassium vapor, has a strong forward to backward asymmetry (25:1). A weaker signal at 1.18 μm (potassium $3D_{5/2} \rightarrow 4P_{3/2}$) can also be observed under certain circumstances. We believe the large $3D_{3/2}$ population is created through a two-photon photodissociation of the NaK molecule. Sharp dips in the excitation spectra are due to coincidental depletion ot fhe upper $3D_{3/2}$ level via laser excitation to Rydberg F levels. The forward to backward asymmetry in this emission is still unexplained, but may result from either a wave-mixing process triggered by the photodissociation lasing or simply an asymmetric gain in the latter. Finally we note that the 1.17 μm forward emission is enhanced by more than an order of magnitude when the pump dye laser contains a weak (0.5%) ASE contamination near 720 nm. We believe that this enhancement results from a two-step sequence in which NaK $A^1\Sigma^+$ is first excited from the ground ($X^1\Sigma^+$) state with a 750 nm photon and then is photodissociated by a second photon near 720 nm. Note that two laser photons at 720 nm produce no emission at 1.17 μm. Further work on this two-step process is currently in progress.

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