Advances in Laser Science - III
ed. A. C. Tam, J. L. Gole and W. C. Stwalley, American Institute of Physics - New York 1988

WAVE-MIXING PROCESSES IN SODIUM-POTASSIUM VAPOR

B. K. Clark, M. Masters and J. Huennekens, Lehigh University, Bethlehem PA 18015

ABSTRACT

We report observations of 4-wave and 6-wave mixing processes based upon two-photon pumping of the potassium 4S → 6S and 4S → 4D transitions in a sodium-potassium mixture. Additionally we observe coherent emissions at the potassium 3D → 4P transition frequencies for pump wavelengths between 725 and 760 nm. The excitation spectra for these emissions show several broad peaks, each of roughly 10 nm width. A series of sharp dips is superimposed on the most intense peak due to depletion of the 3D population by laser-induced absorption to Rydberg F levels. Although the 3D → 4P emissions can be observed in both the forward and backward directions, they are ~50 times more intense in the forward direction. We believe the forward emission is primarily due to 6-wave mixing while the backward emission can most likely be attributed to amplified spontaneous emission following two-photon molecular photodissociation. However, at present, the exact mechanisms responsible for these 3D → 4P emissions are not fully understood.

THE EXPERIMENT

The experiment was carried out in a crossed heat-pipe oven containing a 2:1 mixture of potassium and sodium and operated at ~435°C with an argon buffer gas pressure of ~2 Torr. The alkali vapor was excited with a Nd:YAG pumped dye laser operating in the 690 to 760 nm range with LDS 722 dye. The laser typically delivered ~10 mJ in a 10 ns pulse, and the beam was in all cases unfocussed. Forward, backward and side fluorescence was imaged onto a monochromator equipped with either a photomultiplier or an intrinsic germanium detector.

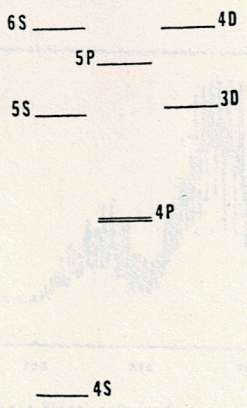

Fig. 1 - Potassium energy levels

RESULTS AND DISCUSSION

With the dye laser wavelength tuned to the 4S → 6S two-photon transition at 728.4 nm we observed K_2 and NaK molecular fluorescence in the side direction along with various sodium and

potassium atomic lines. In the forward direction we observed coherent emission at the potassium 5P → 4S, 3D → 4P and 5S → 4P transition wavelengths (see Fig. 1) and at 1280 and 1370 nm which do not correspond to any atomic transitions. In the backward direction, only the 3D → 4P and 5S → 4P emissions were observed. The 5P → 4S forward emission was also observed when pumping the 4S → 4D two-photon transition. The various coherent emissions can be understood as resulting from the following processes:

4-wave mixing

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

6-wave mixing

$$\omega_{1280(1370)} = 2\omega_{laser} - (\omega_{6S\rightarrow 5P} + \omega_{5P\rightarrow 3D(5P\rightarrow 5S)} + \omega_{laser})$$

amplified spontaneous emission (ASE)

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

The ASE emissions are characterized by approximately equal intensities in the forward and backward directions, while the wave-mixing processes result only in forward emission due to phase-matching requirements. These various processes have either been observed previously in potassium vapor,[1-2] or can be understood by analogy to similar observations made in other alkalis.[3-5]

We also observe strong forward and backward emission at 1170 and 1180 nm (potassium 3D → 4P) when pumping the vapor over a broad range between 725 and 760 nm. The excitation spectrum for the 1170 nm line is shown in Fig. 2. The forward to backward ratio is approximately 55 for a pump wavelength of 752 nm and an argon pressure of 1.8 Torr. As can be seen, the excitation spectrum consists of 3 broad peaks: a very strong one centered at 752 nm, a somewhat weaker one at 741 nm, and a very weak one at 730 nm. The most intense peak has a series of sharp dips superimposed.

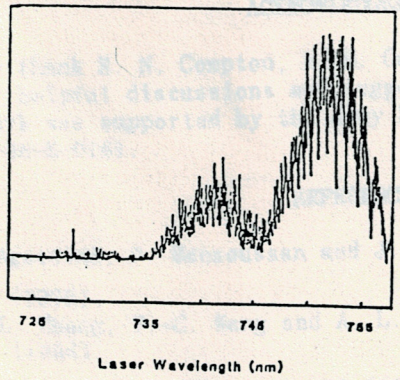

Fig. 2 - 3D → 4P excitation spectrum

Measurements of the 1170 nm forward and backward intensity as a function of argon pressure and laser power were carried out. As the pressure increased from 2 to 25 Torr, the forward signal strength

decreased by about a factor of 2 for a pump wavelength of 752 nm. Over the same range the 742 nm peak almost completely disappeared. Similar but not identical observations were made for the backward emission. Laser intensity dependences showed log-log slopes of ~.7 and 1.3 for the backward and forward 1170 nm emission, respectively. These observations support the idea that the forward and backward emission are caused by different mechanisms.

An investigation of the positions of the sharp dips in the 752 nm peak of the excitation spectrum show that they coincide with 3D → nF transitions with n ranging from 21 to 28. Thus at these pump wavelengths the laser depopulates the 3D state by inducing transitions to high-lying Rydberg states, and thereby reduces the 1170 nm emission. Similar excitation spectra for the 1180 nm line, which probe the $3D_{5/2}$ fine-structure level, show dips shifted by the expected 2 cm^{-1}.

It is worth noting that the 3D → nF dips are significantly more pronounced in the backward direction. This reflects the greater sensitivity of the 3D - 4P population inversion required for ASE compared to total 3D population responsible for forward wave mixing. We now believe the 3D → 4P backward emission can be explained as ASE probably following a two-photon molecular photodissociation process. The forward emission is most probably due primarily to 6-wave mixing involving 2 laser photons and one photon at 1170 or 1180 nm. Also involved in the process are 4P → 4S resonance photons (which are highly trapped and therefore unobservable), and two IR photons which lie beyond our detector response.

We must emphasize that these explanations are still somewhat speculative at present, and additional data is needed before final conclusions can be reached.

ACKNOWLEDGEMENTS

We thank R. N. Compton, W. R. Garrett and M. A. Moore for several helpful discussions and suggestions concerning this work. This work was supported by the Army Research Office under grant DAAL03-86-K-0161.

REFERENCES

1. P. Agostini, P. Bensoussan and J. C. Boulassier, Op. Commun. 5 293 (1972).
2. P.-L. Zhang, Y.-C. Wang and A. L. Schawlow, J. Opt. Soc. Am. B 1 9 (1984).
3. W. Hartig, Appl. Phys. 15 427 (1978).
4. Z. G. Wang, H. Schmidt and B. Wellegehausen, Appl. Phys. B 44 41 (1987).
5. S. M. Hamadani, J. A. D. Stockdale, R. N. Compton and M. S. Pindzola, Phys. Rev. A 34 1938 (1986).