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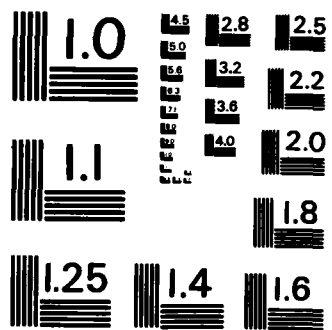
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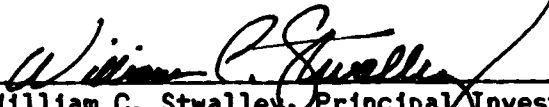
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Annual Technical Report
for
Air Force Office of Scientific Research grant
AFOSR-84-0178, "Alkali Metal Diffuse Band Lasers"
for
June 1, 1984-May 31, 1985


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1. Abstract

Progress is described on study of a new class of potential excimer lasers based on diffuse bands of the alkali metal vapors. In particular, it has been established that the violet diffuse bands of sodium vapor and the yellow diffuse bands of potassium vapor are each composed of overlapping singlet and triplet "excimer" emission continua of the diatomic molecule. The violet gain and low absorption loss previously found with laser optical pumping of sodium vapor have been confirmed and extended to yellow gain and low absorption loss in potassium vapor. Prospects for laser oscillation in the near future appear to be very good.

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2. Research Objectives

As stated in our original proposal, the basic research objectives of this grant are as follows:

- a. Attempted demonstration of violet diffuse band laser action in sodium vapor using laser optical pumping.
- b. Investigation of alternate pumping schemes.
- c. Investigation of other alkali metal diffuse band laser possibilities.
- d. Assistance to Physical Sciences, Inc. (Andover, Massachusetts) in their attempted demonstration of a near infrared solar-pumped sodium vapor laser.

The progress in each of these areas is described below in Sections 3. a., b., c. and d., respectively.

3. Research Progress

- a. Attempted demonstration of violet diffuse band laser action in sodium vapor using optical pumping

The major item of progress in this area was the establishment of the correct electronic assignment of the violet bands of sodium vapor (publication 4. a.). In particular, by exciting single vibrational-rotational levels of the $2^1\Sigma_u^+$ state of Na_2 at Iowa and the $2^3\Pi_g$ state of Na_2 at MIT, we have established that the violet bands of sodium vapor are in fact overlapping singlet and triplet bands of Na_2 . The detailed assignments are $2^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+$ (peaking near 452 nm) and $2^3\Pi_g \rightarrow 1^3\Sigma_u^+$ (peaking near 436 nm). Thus the variability of the ratio between the 436 nm and 452 nm emission peaks (for both of which we have previously shown there is significant gain) is now understood for the first time.

Another significant advance was measurement of the absorption cross section (measured at 2 points in Figure B. 1.-2. of our original proposal) at many other points in regions of high gain. As expected from our preliminary results, the absorption cross sections are acceptably small.

Pulsed laser (XeF excimer (353 nm) and tripled Nd:YAG (355 nm)) experiments were disappointing in that (unlike the cw experiments) significant gain was not found. However, strong ionization was later detected in the pulsed experiments, so it is thought that much lower peak power conditions are needed for pulsed gain. Hence we are emphasizing cw experiments in current work.

To obtain laser action in sodium vapor, two new sodium vapor containers have been built:

- i. a "magnum" heat pipe oven, with roughly double the 30 cm gain length of our standard heat pipe ovens and with careful baffling to prevent particulate formation (and consequent optical loss) at the sodium vapor-buffer gas interface.

- ii. a "recirculating supersonic molecular beam oven" (RSMBO), to provide direct optical access to a thin "sheet" of sodium vapor.

The former oven is operational and preliminary experiments are discussed below. The latter oven, funded in part by the Iowa High Technology Council, is now being modified (improved insulation and heating) to allow for high pressure (> 1 torr) operation, and will not be discussed further in this year's report. The heat pipe trough (developed at Physical Sciences; see d.) may also be used in the coming year.

The "magnum" heat pipe oven has been used in two attempts (thusfar unsuccessful) at optically pumped violet band laser oscillation using a ring cavity. In the first attempt, the Brewster's angle windows quickly developed brown spots (presumably because of the intense UV irradiation), while in the second attempt, spots were discovered on the dichroic optics. Both these optics problems are now supposedly solved and we hope to obtain laser oscillation soon. Note that no such optics problems are anticipated for the potassium yellow bands (see c.), so we are planning to attempt laser oscillation on those bands also.

- b. Investigation of alternate pumping schemes

Among the many possible alternatives to laser optical pumping (e^- beam, flashlamp, discharge, sodium lamp, solar, nuclear, etc.), it now appears that lamp pumping is the most desirable in the short term (either using the recirculating supersonic molecular beam oven or the Physical Sciences Inc. heat pipe trough (see d.) as the active medium). We will have available beginning in August a 4.5 kW 10 cm long mercury lamp and power supply (primary use: pumping a 60 watt cw alexandrite laser, the second of its kind, custom built by Allied Corporation). We plan to attempt lamp optical pumping with this equipment once laser optical pumping is successful.

An additional interesting new possibility is chemical pumping; J. L. Gole

(Georgia Tech) has informed us he has seen strong violet and near ultraviolet chemiluminescence in sodium-halogen reactions which appears to correlate well with our calculations of $\text{Na}_2 2^1\Sigma_U^+ \rightarrow X^1\Sigma_G^+$ "violet band" emission (the ultraviolet features are bound-bound transitions). A proposal for chemical pumping schemes is being considered at Los Alamos National Laboratory (contact: Randy Jones).

c. Investigation of other alkali metal diffuse band laser possibilities

Partly because of the optics problems in strong UV pumping, we have spent more time on the heavier alkali metals (especially potassium) than originally anticipated. In particular, we have been able to establish that, as in the violet bands of sodium, the yellow bands of potassium include overlapping singlet and triplet emissions ($2^1\Sigma_U^+ \rightarrow X^1\Sigma_G^+$, peak at 570 nm; $2^3\Pi_G \rightarrow 1^3\Sigma_U^+$, peak at 575 nm). Significant gain has been measured at 565 nm (paper 4. b.); further experiments are in progress.

In addition, the triplet portion of the alkali vapor diffuse bands (in Na, K, Rb and Cs) have been directly excited for the first time (free \rightarrow bound absorption followed by bound \rightarrow free emission). A paper (4. c.) describing this new technique for isolating the triplet emission is in preparation; the Ph.D. thesis of W. T. Luh presents most of the basic results. Some new previously unobserved lithium diffuse bands in the violet have also been found (paper 4. e.).

d. Assistance to Physical Sciences, Inc. (Andover, Massachusetts) in their attempted demonstration of a near infrared solar-pumped sodium vapor laser

We have consulted extensively with Physical Sciences, Inc. concerning their solar-pumped near infrared sodium vapor concept, both by phone and complementary visits (further visits are planned for July and August 1985). We have assisted in the modified design of their heat pipe trough, in their consideration of the kinetics of the active medium, and by providing values

for the absolute bound-bound radiative transition probabilities for all vibrational levels in the $B^1\Pi_u - X^1\Sigma_g^+$ bands of Na_2 (these bands are critical to the production of a high concentration of Na^* , which is in turn critical to their laser concept). Bound-free (photodissociation) radiative transition probabilities are now being calculated.

4. List of Written Publications

- a. "Electronic Assignments of the Violet Bands of Sodium", G. Pichler, J. T. Bahns, K. M. Sando, W. C. Stwalley, D. D. Konowalow, L. Li and R. W. Field, submitted to Phys. Rev. Letters and currently being revised to incorporate the referee's comments and new spectroscopic data from R. F. Barrow, Oxford.
- b. "Observation of Gain in the Yellow Bands of Potassium Vapor", S. P. Heneghan, K. P. Chakravorty and W. C. Stwalley, manuscript in preparation to be submitted to Appl. Phys. Letters or JOSA B.
- c. "Direct Excitation and Simulation of $2^3\Pi_g - 1^3\Sigma_u^+$ Diffuse Bands in Sodium, Potassium, Rubidium and Cesium Vapor", W. T. Luh, J. T. Bahns, K. M. Sando and W. C. Stwalley, manuscript in preparation to be submitted to J. Chem. Phys.
- d. "Electronic Assignments of the Yellow Bands of Potassium", W. T. Luh, K. M. Sando, K. P. Chakravorty, W. C. Stwalley, G. Pichler and D. D. Konowalow, manuscript in preparation to be submitted to Chem. Phys. Letters.
- e. "Diffuse Bands of the Lithium Dimer", J. T. Bahns, W. C. Stwalley and G. Pichler, to be submitted to J. Chem. Phys.

5. Professional Personnel* (University of Iowa)
- a. William C. Stwalley, Principal Investigator
 - b. Paul D. Kleiber, Assistant Research Scientist
 - c. Shawn P. Heneghan, Postdoctoral Research Associate
 - d. Kuo-Kwang Wang, Postdoctoral Research Associate
 - e. A. Marjatta Lyyra, Postdoctoral Research Associate
 - f. Wei-Tzou Luh, Graduate Student (scheduled to receive Ph.D. in August 1985 in Physical Chemistry; passed final exam on thesis entitled "Laser Spectroscopic Studies of Alkali Metal Dimers"[†] on July 5, 1985).
 - g. Warren T. Zemke, Visiting Professor (Summer only)

* Other University of Iowa researchers collaborating on this research include Dr. John T. Bahns (Iowa High Technology Council support), Dr. Krishna P. Chakravorty (Visiting Research Scientist with support from the Government of India), and Professor Kenneth M. Sando (unsupported).

[†] Copy available on request.

6. Interactions - William C. Stwalley et al.

a. Papers and Seminars

- i. "Diffuse and Interference Bands in Alkali Metal Dimers", Paper ME3 at the 39th Symposium on Molecular Spectroscopy (Columbus, Ohio, June 1984).
- ii. "Photophysics and Photochemistry of Alkali Metal Vapors", Department of Physics, University of Missouri (St. Louis, July 1984).
- iii. "Spectra and Potential Energy Curves of Alkali Metal Diatomic Molecules: Experimental Results Compared with Theory", invited paper at American Chemical Society meeting (Philadelphia, August 1984).
- iv. "Photochemistry and Photophysics of Alkali Metal Vapors", Laser Seminar, University of Toronto (Toronto, October 1984).
- v. "Photochemistry and Photophysics of Alkali Metal Vapors", Department of Chemistry, Pennsylvania State University (State College, October 1984).
- vi. "Photophysics and Photochemistry of Alkali Metal Vapors", invited paper, International Conference on Applications of Lasers and Electro-Optics (Boston, November 1984).
- vii. "Diffuse Bands in Alkali Metal Vapors" and "Ionization and Energy Transfer in Alkali Metal Vapors", invited papers at Lasers '84 (San Francisco, December 1984).
- viii. "Photochemistry and Photophysics of Alkali Metal Vapors", Department of Chemistry, University of Michigan (Ann Arbor, January 1985).
- ix. "Photophysics and Photochemistry of Alkali Metal Vapors", Combustion Research Facility, Sandia National Laboratories (Livermore, February 1985).

b. Consultations

- i. Visited Physical Sciences, Inc. (D. Ham, host) regarding collaboration on solar-pumped near infrared sodium vapor laser (Andover, Massachusetts, July 1984).
- ii. Visited Air Force Weapons Laboratory (S. Davis, host) regarding optically pumped lasers (Albuquerque, New Mexico, October 1984).
- iii. Hosted David Ham, Physical Sciences, Inc., regarding collaboration on solar-pumped near infrared sodium vapor laser (November 1984).

7. New Discoveries

- a. Discovery of gain at ~565 nm in the yellow bands of potassium vapor.
- b. Discovery that sodium and potassium visible continua represented overlapping singlet and triplet transitions ($2^1\Sigma_u^+$ \rightarrow $X^1\Sigma_g^+$ and $2^3\Pi_g$ \rightarrow $1^3\Sigma_u^+$, respectively).
- c. Discovery of new violet diffuse bands in lithium vapor.

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