27
12-17-80
74C

240 N TIS

#1

R710

UCRL-S3075

The generation of tunable coherent far-infrared radiation using atomic Rydberg states

William Bookless

(Ph.D. Thesis)

MASTER

December 1980

**Lawrence
Livermore
Laboratory**

The generation of tunable coherent far-infrared radiation using atomic Rydberg states

William Bookless

(Ph.D. Thesis)

Manuscript date: December 1980

LAWRENCE LIVERMORE LABORATORY
University of California • Livermore, California • 94550

Available from: National Technical Information Service • U.S. Department of Commerce
5285 Port Royal Road • Springfield, VA 22161 • \$11.00 per copy • (Microfiche \$3.50)

DISTRIBUTION

UNCLASSIFIED

Bookless, William A., The Generation of Tunable Coherent Far-Infrared Radiation using Atomic Rydberg States, PhD, Department of Physics and Astronomy, December, 1980

A source of tunable far-infrared radiation has been constructed. The system has been operated at 91.6 cm^{-1} with a demonstrated tunability of $.63 \text{ cm}^{-1}$. The system is based on a Rydberg state transition in optically pumped potassium vapor. The transition energy is tuned by the application of an electric field to the excited vapor. The transition wavelength and the shifted wavelength were detected and measured by the use of a Michelson interferometer and a liquid helium cooled Ga:Ge bolometer and the data was reduced using Fast Fourier transform techniques.

Extensive spectroscopy was done on the potassium vapor to elucidate the depopulation paths and rates of the excited levels. Both theoretical and experimental results are presented to support the conclusions of the research effort. Additionally, possible alternative approaches to the population of the excited state are explored and recommendations are made for the future development of this source as well as the potential uses of it in molecular spectroscopy.

THE GENERATION OF TUNABLE COHERENT FAR-INFRARED
RADIATION USING ATOMIC RYDBERG STATES

by

William Bookless

A Thesis

Submitted to the

Department of Physics and
and Astronomy

The Graduate School of the University
of Wyoming in Partial Fulfillment of Requirements
for the Degree of
Doctor of Philosophy

University of Wyoming

Laramie, Wyoming

December, 1980

ACKNOWLEDGMENTS

I would like to express my sincere thanks to everyone in the Chemistry Department at the Lawrence Livermore National Laboratory for making my research there both very rewarding and pleasant. In particular, I would like to thank Dr. L. Hrubesh and Dr. C. G. Stevens for their complete support and guidance throughout the course of the work and D. Johnson and L. L. Steinmetz for their help in many facets of the planning and building of the apparatus. I would also like to thank Helen Meredith for her excellent work and patience in typing the manuscript. Special thanks is due to Dr. E. A. Rinehart for his guidance and for his effort in making my stay at Livermore possible.

I would like to express my appreciation to the Associated Western Universities, Inc. for their financial support throughout my research.

I wish to express special gratitude to my wife, Stacy, for her complete support during my graduate career. Her encouragement has been greatly appreciated.

TABLE OF CONTENTS

I. INTRODUCTION.	1
II. THEORY.	8
Physics of the Atom	8
Stark Effect.	30
III. EXPERIMENTAL.	36
Heat Pipe	36
Configurations.	51
Experimental Procedure.	59
IV. RESULTS	100
V. CONCLUSIONS	132

LIST OF FIGURES

Figure	Page
1. Absorptivity versus frequency for diatomic rotational transitions.	4
2. The Grotrian diagram for atomic potassium with some transition wavelengths shown	5
3. An expanded diagram of potassium Rydberg states with the nearest-neighbor transition energies shown. All levels and transition energies are in cm^{-1}	6
4. A schematic diagram of a heat pipe oven	38
5. The temperature profile of a heat pipe containing lithium	41
6. A schematic diagram of the "can" type heat pipe oven with Stark plates.	43
7. A photograph of the "can" type heat pipe oven with Stark plates removed	44
8. Sketch of the "pancake" heat pipe oven.	47
9. Cross section view of the "pancake" heat pipe oven.	48
10. Power spectrum for R6G laser dye when used in a Molelectron DL-200 laser pumped by a Molelectron UV-1000 N_2 laser.	53
11. The experimental configuration used with the N_2 -Dye laser pump	54
12. The experimental configuration used with the Chromatix CMX-4 dye laser pump.	56
13. A typical etalon scan of the Spectra-Physics 375 dye laser showing the 4s-12p doublet.	63
14. The configuration used for C.W. spectroscopy using the Spectra-Physics 170 A^+ -375 dye laser pump.	65
15. The opto-galvanic signal associated with the group of lines near 580 nm	67
16. The configuration used for the C.W. Kr^+ -dye laser, CMX-4 pulsed laser experiments.	69
17. FIR output from the detectors as the pump laser is scanned. a) the golay cell, b) the Ga:Ge bolometer with $<2\mu\text{A}$ current, c) the Ga:Ge bolometer with $8.2\mu\text{A}$ current.	80

18.	The window function used to "taper" the data. The 50% function is a true "Hanning" function.	85
19.	The bolometer response to a broadband source (hot wire) showing the water spectrum due to atmospheric water	.88
20.	The schematic of the high voltage pulse generator	.90
21.	A schematic diagram of the total FIR experimental apparatus	.93
22.	The absorption spectrum of the U.V. highly reflecting mirror R >99% at 300 nm)	.95
23.	The absorption of the pump laser for the 4s-13p transition with an exponential curve fitted.	.98
24.	The fluorescence from the K vapor when pumped at 767 nm, a) on the 4s-4p transition, b) .04 nm off of the 4s-4p transition.	101,103
25.	The diagram of the molecular potassium system showing the proposed dissociative ground state of the broad molecular feature	105
26.	A scan of the fluorescence from the potassium vapor from 400-600 nm pumped to the 13p level. No temporal filtering has been used	108
27.	The same scan as Figure 26, but with the addition of temporal filtering using a boxcar integrater, a) hot K vapor, b) cold (for background signal)	110,111
28.	The same as 27, but from 600-800 nm. The line at 674 nm is the second grating order of scattered 337 nm N ₂ laser light, a) hot K vapor, b) cold.	113,114
29.	The same as 27, but from 400-800 nm. The broad background about 700 nm is due to a fluorescing filter. Not the partial absorption of the broad fluorescence about the sharp peaks near 690 nm, a) hot K vapor, b) cold.	115,116
30.	Photographs of the temporal character of the groups of lines shown in Figure 27a, a) 475.4 nm, b) 480.2 nm, c) 486.5 nm, d) 495.3 nm, e) 510.8 nm, f) 533.2 nm, g) 579.8 nm, h) 585.5 nm, i) same as (h), but on a 20 nsec/cm scale	118
31.	Typical output for a low resolution scan of the Michelson interferometer.	120

32.	Fourier transform spectrum of the output of the heat pipe using the Ga:Ge bolometer, a) total spectrum, b) expanded view of (a).	121,122
33.	High resolution scan of the heat pipe output using the golay cell.	124
34.	High resolution scan of the "pancake" heat pipe output with no field using the bolometer.	126
35.	Output of the "pancake" heat pipe with the field on (~ 2000 V/cm) and off, a) from $0-100\text{ cm}^{-1}$, b) from $88-100\text{ cm}^{-1}$	128-129
36.	Expanded view of Figure 19 showing more clearly the water vapor absorptions	130

LIST OF TABLES

Table	Page
1. Potassium energy levels measured and calculated. All energies are in cm^{-1}	9
2. Transitions from $13p \rightarrow ns$ and $13p \rightarrow nd$ with the A coefficients, lifetimes, and doppler linewidths as well as the total A coefficient	12
3. The quantum defects for potassium ($9 < n < 15$).	18
4. Transitions from $11d-np$ and $11d-nf$ with the A coefficients and lifetimes as well as the total A coefficient.	28
5. Results from the calculations by Zimmerman on the potassium Stark shift.	33
6. Potassium shifts expected for 2000 V/cm	34
7. The data for the absorption measurements on potassium vapor for the $4s-13p$ transition	97

CONTENTS OF APPENDICES

Appendix	Page
A. The computer program to take the interferometric data and do the Fast Fourier Transform (FFT) and plot the results on an X-Y plotter with axis.	142
B. The computer program to take and plot the fluorescence spectra.	149

1. INTRODUCTION

The generation of tunable Far-infrared (FIR) radiation has long been the object of experimental study. In the past, the most common sources in this region of the spectrum ($100\mu - 1000\mu$) have been black-body radiators. The appearance of the maser and later the laser stimulated interest in the FIR region of the electromagnetic spectrum. This region is only now becoming accessible to the techniques of laser spectroscopy.

The earliest attempts to generate tunable radiation were through the use of mixing techniques whereby two suitable infrared laser beams separated in frequency by the appropriate amount would be mixed in a crystal with a high non-linear index. The difference frequency produced is in the FIR region.(1-9) All of these systems have suffered from either a very limited tuning range or a very large linewidth which has prevented their general acceptance as spectroscopic sources.

The most popular FIR laser source has been the optically pumped molecular laser which generally consists of a CO_2 laser pumping molecules such as CH_3OH to produce very strong, narrow laser lines in the FIR region. These systems are, however, only line tunable; attempts to Stark tune them to produce a continuously tunable source have produced tuning ranges of only about 10 MHz (10). An excellent review of the progress in source development is given in (1').

The method proposed here is based on the very small separation between the Rydberg states of atomic alkali metals. When one reaches states with the principle quantum number n greater than 11

the energy separation between adjacent states with allowed dipole transitions is in the FIR region. These levels have also shown enormous Stark polarizabilities (12) which would make the transitions potentially tunable over a wide range. The linewidths of these transitions are limited fundamentally by the decay rates of the states which in the absence of collisions would be only 180 KHz, but practically are limited to about 10 MHz. The large number of FIR transitions available in the alkali metals coupled with the large tunabilities available make this method of generation a very attractive and potentially quite useful source.

The upper states of the alkali metals have long been known to display stimulated emission and have been studied using laser excitation for over 13 years (13). More recently, lasing has been shown in alkali metal systems (14,15). These methods were extended in a proposal type paper by Lau et al. (16) to include the possibility of Stark tuning the transitions. In (17), optically pump stimulated emission was shown to occur in the FIR region from the highly excited Rydberg states in potassium. This work has been the experimental basis for the work presented here. A very interesting and informative article on the experimental work on the Rydberg states of atoms has been recently published by Latimer (18) and gives an excellent background for our work.

The tunability of this system as mentioned above, comes from the large Stark polarizabilities of the atoms and thus requires an applied electric field for its implementation. This field can be in the form of a periodic field giving the dynamic Stark effect,

or in the form of a d.c. field for the d.c. Stark effect. The former has been demonstrated using a pulsed CO_2 laser, but the equipment requirements are prohibitive for many applications. The latter method has been demonstrated in an atomic beam apparatus (19). This method was limited in output power by the extremely small number of atoms available in an atomic beam.

This research grew out of the need for a highly sensitive monitor for gases. In the past the microwave region of the spectrum has been the most selective area to do the work, but has suffered from a lack of sensitivity. This problem could be alleviated by the use of FIR radiation to do pure rotational spectroscopy on the molecular systems of interest. As shown in Figure (1) this region of the spectrum offers an increase in absorptivity of up to 10^3 over similar processes in the microwave region. The author has been interested in the application of lasers to atomic and molecular physics and this project provided a vehicle by which all of these interests could be satisfied.

To begin, the particular system of interest is described. A source of FIR radiation based on the $13p-11d$ transition in atomic potassium has been modeled and constructed. The Grotrian diagram in Figure (2) shows the energy levels of the potassium atom and the transition energies between the ground state and the excited states connected by dipole transitions. The transitions of interest are shown in more detail in Figure (3).

In this thesis the theoretical and experimental work done to demonstrate the generation of tunable FIR radiation in the region

Figure 1. Absorptivity versus frequency for diatomic rotational transitions. (58)

of 92 cm^{-1} ($109\mu\text{ M}$) will be outlined. The next chapter will concentrate on the theory of highly excited atoms and their depopulation from upper excited states by radiative and non-radiative (collisional) processes. A discussion of the Stark effect will be included. The third chapter will discuss the experimental arrangements used and the configurations considered. The fourth chapter will examine the results of these experiments and our conclusions will be presented in the final chapter.

II. THEORY

The method used here for the generation of tunable sub-millimeter radiation can be addressed theoretically in a very straightforward fashion. We can break the theoretical treatment up into two general areas and investigate each independently.

Physics of the Atom

The first area to be considered is that of the atomic physics of a highly excited atom. In particular, since an alkali metal is being considered, many simplifying approximations can be made for the quantum mechanics of the atom. Core interactions are going to become decreasingly important the further from the core and in fact can be ignored when high n states are dealt with. Table 1 shows that at high n values and large l values, a simple Rydberg formula using effective quantum numbers supplied by experiment, describes the energy levels of the atom quite well. The measured values for some of these levels is also given for comparison at lower values.

Similarly, at high values of $n + l$ the semi-empirical methods of Bates and Damgaard (20) provide accurate results for the transition probabilities. A semi-empirical method used by Anderson and Zilitis (21) to calculate the oscillator strengths of potassium for all of the transitions of interest will be discussed. From these oscillator strengths the transition probabilities and natural lifetimes of states will then be calculated. Using these lifetimes and probabilities the absorption cross sections as well as the stimulated emission and spontaneous cross sections for the

TABLE 1. Potassium energy levels measured and calculated. All energies are in cm^{-1}

Level	Moore (J)	Calculated	Level	Moore (J)	Calculated
4s	0		8f		33290.80
4p	13042.89 (3/2)	14046.80	8g		33294.66
5s	21016.80	21229.72	10p	33411.54 (3/2)	33412.19
3d	21536.75 (3/2)	20317.62	9d	33572.11	33570.86
5p	24720.20 (3/2)	24858.96	11s	33598.17	33599.74
4d	27398.11 (3/2)	27134.81	9f	33652.0	33651.95
6s	27450.65	27497.30	9g		33654.66
4f	28127.70	28116.61	11p	33737.44 (3/2)	33737.68
6p	29007.70 (3/2)	29041.41	10d	33851.76	33851.34
5d	30185.69 (3/2)	30110.95	12s	33869.7	33872.24
7s	30274.26	30290.12	10f		33910.18
5f	30605.60	30602.57	10g		33912.16
5g	30619.80	30618.42	12p	34148.63 (3/2)	33972.96
7p	31074.46 (3/2)	31085.30	11d	34056.7	34057.15
6d	31695.75 (3/2)	31670.90	13s	34069.3	34072.76
8s	31764.95	31772.20	11f		34101.18
6f	31953.00	31951.26	11g		34102.67
6g	31960.8	31960.43	13p	34148.63 (3/2)	34148.53
8p	32230.12 (3/2)	32234.29	12d		34212.60
7d	32598.46	32589.04	14s		34224.58
9s	32648.17	32651.79	12f		34246.42
7f	32764.52	32763.78	14p	34283.15 (3/2)	34283.00
7g		32769.55	13d		4332.92
9p	32942.08 (3/2)	32943.69	15s		34342.28
8d	33178.36	33174.64	15p	34388.46 (3/2)	34388.27
10s	33214.39	33216.16			

FIR transitions of interest. We will also examine the competing processes to the stimulated emission and compare these rates to those of the desired transitions. Finally we will look at the conditions for lasing in the K vapor and discuss the threshold conditions for super fluorescence as well as true lasing.

We begin by looking at the methods used for the calculation of the oscillator strengths of potassium. If we designate $P(r)$ as the radial part of the wave function of the outer or valence electron then we can write

$$\psi(r, \theta, \phi) = P(r)/r Y(\theta, \phi)$$

where $Y(\theta, \phi)$ are the well known spherical harmonic functions. The oscillator strengths can be calculated from the matrix elements

$$(r_{ik}) = \int_0^\infty P_k r P_i dr. \quad (1)$$

If we are interested only in the average oscillator strength, then we can write

$$f_{ik} = \frac{\max(l_i, l_k)}{2l_i + 1} \frac{2}{3} (E_k - E_i) (r_{ik})^2 \quad (2)$$

where $\max(l_i, l_k)$ is the maximum azimuthal quantum number l_i or l_k of the initial and final states. E_i and E_k are the initial and final energies in atomic units (21). This calculation does not take into consideration the sign of f_{ik} . The functions $P(r)$ have been calculated by the semi-empirical system proposed in (22). Equation (3) is solved for $P(r)$, where E_{exp} is the

$$\ddot{P}(r) + 2 E_{\text{exp}} + \frac{2z}{r} - 2V(r) - \frac{l(l+1)}{r^2} P(r) = 0 \quad (3)$$

experimental value of the ionization potential for potassium and $V(r)$ is the screening potential taken from the self-consistent field method of (23).

These calculations have been carried out by Anderson and Zilitis (21) and they are used without further improvement. As the high n -value states of the potassium atom have a very large radius, r goes as n^2 , we expect the approximations of Anderson and Zilitis to be quite good in the region of $n = 13$.

Using these oscillator strengths we can calculate the spontaneous transition probabilities by using

$$A_{ik} = 2\alpha^3 (E_k - E_i)^2 f_{ik} \quad (4)$$

or in units of sec^{-1}

$$A_{ik} = 0.6669 (E_k - E_i)^2 f_{ik} \quad (5)$$

where E is measured in cm^{-1} .

This is the probability of a transition from level i to level k in sec^{-1} by spontaneous decay. The spontaneous lifetime of a level is then

$$\tau_i = \frac{1}{A_{i\tau}} = \left[\sum_k A_{ik} \right]^{-1} \quad (6)$$

An example of this is in Table (2). This illustrates the case for spontaneous decay from the $13P$ state of potassium in the absence of collisions.

TABLE 2. Transitions from 13p→ns and 13p→nd with the A coefficients, lifetime, and doppler linewidths as well as the total A coefficient.

Transition		f_{ik}	$(E_k - E_i) \text{ cm}^{-1}$	$A_{ik} \text{ sec}^{-1}$	$t_{ik} \text{ sec}$	$\nu \text{ sec}^{-1}$	$\Delta\nu_d \text{ sec}^{-1}$
13p	13s	$1/3 \times 4.87$	7.933×10^1	6.81×10^3	1.47×10^{-4}	2.38×10^{12}	6.73×10^6
13p	12s	$1/3 \times .170$	2.79×10^2	2.94×10^3	3.40×10^{-4}	8.36×10^{12}	2.37×10^7
13p	11s	$1/3 \times .0341$	5.50×10^2	2.30×10^3	4.35×10^{-4}	1.65×10^{13}	4.67×10^7
13p	10s	$1/3 \times .0115$	9.34×10^2	2.23×10^3	4.48×10^{-4}	2.80×10^{13}	7.94×10^7
13p	9s	$1/3 \times .00487$	1.50×10^3	2.44×10^3	4.10×10^{-4}	4.50×10^{13}	1.27×10^8
13p	8s	$1/3 \times .00225$	2.38×10^3	2.84×10^3	3.52×10^{-4}	7.18×10^{13}	2.03×10^8
13p	7s	$1/3 \times .00101$	3.87×10^3	3.37×10^3	2.97×10^{-4}	1.16×10^{14}	3.29×10^8
13p	6s	$1/3 \times .000354$	6.70×10^3	3.53×10^3	2.83×10^{-4}	2.01×10^{14}	5.69×10^8
13p	5s	$1/3 \times .000170$	1.31×10^4	6.51×10^3	1.54×10^{-4}	3.94×10^{14}	1.11×10^9
13p	4s	$1/3 \times .0000560$	3.41×10^4	1.45×10^4	6.89×10^{-5}	1.02×10^{15}	2.90×10^9
13p	11d	2.29	9.16×10^1	1.28×10^4	7.78×10^{-5}	2.75×10^{12}	7.78×10^6
13p	10d	.163	2.97×10^2	9.58×10^3	1.04×10^{-4}	8.90×10^{12}	1.52×10^7
13p	9d	.0451	5.76×10^2	1.00×10^4	1.00×10^{-4}	1.73×10^{13}	4.90×10^7
13p	8d	.0181	9.70×10^2	1.14×10^4	8.80×10^{-5}	2.91×10^{13}	8.24×10^7
13p	7d	.00849	1.55×10^3	1.36×10^4	7.35×10^{-5}	4.65×10^{13}	1.32×10^8
13p	6d	.00417	2.45×10^3	1.67×10^4	5.98×10^{-5}	7.36×10^{13}	2.08×10^8
13p	5d	.00196	3.96×10^3	2.05×10^4	4.87×10^{-5}	1.19×10^{14}	3.37×10^8
13p	4d	.000763	6.75×10^3	2.32×10^4	4.31×10^{-5}	2.02×10^{14}	5.74×10^8
13p	3d	.000160	1.26×10^4	1.70×10^4	5.89×10^{-5}	3.78×10^{14}	1.07×10^9

$$\text{Total } A_{13} = 1.82 \times 10^5 \text{ sec}^{-1}$$

$$\tau_{13} = 5.49 \times 10^{-6} \text{ sec}$$

From this we can calculate the B coefficient for stimulated absorption and emission.

The stimulated emission or absorption rate W, and the A and B coefficients are related by the well known relation

$$W = I_{\omega} B = A \frac{4\pi^3 c^2}{\hbar \omega^3} I_{\omega} \quad (7)$$

where ω is the radial frequency of stimulated transition and I is the intensity of the radiation field per unit frequency in units of energy $\times \text{dis.}^{-2}$. Relating the variable ω to ν by the relation,

$$\nu = \frac{\omega}{2\pi} \quad (8)$$

we have,

$$B = A \frac{4\pi^3 c^2}{\hbar (2\pi \nu)^3} \quad (9)$$

or

$$B = A \frac{c^2}{2\hbar \nu^3} \quad (10)$$

for $\hbar = 1.0547 \times 10^{-34} \text{ J-sec}$

$c = 2.9979 \times 10^{10} \text{ cm sec}^{-1}$

$$\Rightarrow B = 4.2606 \times 10^{54} \frac{A}{\nu^3} \text{ cm}^2 \text{ J}^{-1} \text{ sec}^{-1} \quad (11)$$

We can find the cross section for stimulated absorption from the relation

$$\sigma_{\omega} a = \frac{1}{4} \lambda^2 \frac{g'}{g} a_{\omega} \quad (12)$$

where g and g' are the multiplicities of the beginning and final states and a is the transition probability between ω and $\omega + d\omega$ defined by,

$$A = \int a_{\omega} d\omega \quad (13)$$

From this we can calculate an integrated cross section by,

$$\sigma^a \Delta\nu = \frac{1}{4} \lambda^2 \frac{g'}{g} A \quad (14)$$

To find the average stimulated absorption cross section we must then divide by the linewidth of the spectral line, $\Delta\nu$.

To get σ^{ST} , the cross section for stimulated emission, we must multiply by the ratio of the multiplicities of the levels involved. From this we get,

$$\sigma^{ST} = \frac{g'}{g} \sigma^a \quad (15)$$

or more directly in terms of A ,

$$\sigma^{ST} = \frac{1}{4 \Delta\nu} \lambda^2 A \quad (16)$$

The three things affecting the linewidth $\Delta\nu$ of the transition are; the natural linewidth of the transition, the doppler broadening due to the thermal energy of the system, and the collision broadening due to the collision between excited atoms. As will be seen, the collisions between excited atoms are the only collisions needed because of the enormous difference in physical size between excited and ground state atoms.

From Table (2) it is seen that the natural lifetime of the potassium 13P state is approximately 5.5μ sec. By using the relationship,

$$\Delta\nu = \Delta\omega/2\pi = \frac{1}{2\pi\tau} \quad (17)$$

the natural linewidth for this state is,

$$\Delta\nu \approx 2.9 \times 10^4 \text{ Hz} \quad (18)$$

This is exceedingly narrow and as will be seen, is overwhelmed by the other broadening mechanisms.

Next we look at the doppler broadening of the transition due to the thermal energy of the atoms. The linewidth due to this mechanism may be written as,

$$\begin{aligned} \Delta\nu_d &= 2\nu_0 \sqrt{\frac{2kT}{Mc^2} \ln 2} \\ &= 7.162 \times 10^{-7} \left(\frac{T}{M}\right)^{1/2} \nu_0 \end{aligned} \quad (19)$$

where M is the atomic weight of the atom, T is the temperature of the system in $^{\circ}\text{K}$, and ν_0 is the center frequency of the transition. For potassium with $M = 39.1$ we have,

$$\Delta\nu_d = 1.14 \times 10^{-7} T^{1/2} \nu_0 \quad (20)$$

If we again look at the 13p state as the one of interest and take ν_0 for the 4s-13p absorption transition we have,

$$\nu_0 = 1.02 \times 10^{15} \text{ Hz} \quad (21)$$

Also, using a pressure of 1 Torr for the potassium vapor the

temperature of the vapor from ref. (24), is

$$T = \frac{4430}{7.1830 - \log P} \quad (22)$$

or $T = 617^{\circ}\text{K}$

Using this result and equation (21) in (20) we get,

$$\Delta\nu_d = 2.89 \times 10^9 \text{ Hz} \quad (23)$$

We should note here that since this is a function of $T^{1/2}$, it will not vary greatly with temperature (or pressure).

The third broadening method to be discussed is that due to collisions. We can write the linewidth contribution due to collisions as,

$$\Delta\nu_c = \omega_c/2\pi \quad (24)$$

where ω_c is the collision frequency which in turn can be written as

$$\omega_c = \sigma_c n v \quad (25)$$

In this equation σ_c is the collision cross section, n is the number density of colliding atoms, and v is the average velocity of the atoms. The geometric cross section of the atom may be written as,

$$\sigma_c = \pi r_{av}^2 \quad (26)$$

where from Ref. (25),

$$r_{av} = 1/2 \left[3n^{*2} - 1(1+1) \right] a_0 \quad (27)$$

and n^* is the effective quantum number defined by

$$n^* = n - \mu \quad (28)$$

where μ is the quantum defect, and a_0 is the Bohr radius. The quantum defects for potassium are shown in Table (3). Again using the 13p state as an example,

$$\sigma_c = 3.46 \times 10^{-13} \text{ cm}^2 \quad (29)$$

The number density of excited atoms is, in the limit of saturation of the excited state, just one half of the total number density of atoms. This number density may be written,

$$\begin{aligned} n_t &= \frac{p}{kT} \\ &= \frac{1}{1.38 \times 10^{-16} \text{ erg } ^\circ\text{K}^{-1}} \frac{p}{T} \\ &= 7.24 \times 10^{15} \frac{p}{T} \quad (p = \text{dynes cm}^{-2}, T = ^\circ\text{K}) \end{aligned}$$

But we can write,

$$\begin{aligned} p \text{ (Torr)} &= 1.33 \times 10^3 p \text{ (dynes cm}^{-2}\text{)} \\ \Rightarrow n_t &= 7.24 \times 10^{15} \times 1.33 \times 10^3 \frac{p}{T} \text{ Atoms cm}^{-3} \\ &= 9.63 \times 10^{18} \frac{p}{T} \text{ Atoms/cm}^{-3} \quad (p = \text{Torr}, T = ^\circ\text{K}) \end{aligned} \quad (30)$$

Finally we can calculate the rms velocity of the atoms from,

$$\begin{aligned} v &= \sqrt{\frac{3kT}{m}} \\ &= 2.52 \times 10^3 T^{1/2} \text{ cm/sec } (T = ^\circ\text{K}) \end{aligned} \quad (31)$$

for atoms of 39.1 gm/mole.

We, therefore, have combining (29), (30), and (31),

TABLE 3. The quantum defects for potassium ($9 < n < 15$)

l	μ	Reference
0	2.178 (6)	(9)
1	1.712 (2)	(9)
2	0.267 (1)	(9)
3	0.010 (1)	(9)

$$\begin{aligned}\omega_c &= \sigma_c \frac{nt}{2} v \\ &= 4.20 \times 10^9 \frac{p}{T^{1/2}} \text{ radians sec}^{-1}\end{aligned}$$

or from eq. (24),

$$\Delta\nu_c = 6.68 \times 10^8 \frac{p}{T^{1/2}} \text{ Hz} \quad (32)$$

For our standard case of $p = 1$ torr and $T = 617^0\text{K}$ we have,

$$\Delta\nu_c = 2.69 \times 10^7 \text{ sec}^{-1} \quad (33)$$

We see from this result, eq. (18), and eq. (23), that line broadening for the $n = 13$ state in potassium at 1 Torr and 617^0K is essentially all due to the doppler effect. We will use only this term in calculating the average cross section from eq. (14). Using eq. (14) and the result in eq. (23) we have,

$$\sigma^a = \frac{1}{4} \lambda^2 \frac{g}{g^r} A \frac{1}{2.89 \times 10^9} \quad (34)$$

If we rewrite eq. (5) as,

$$A_{ik} = 0.6669 \frac{1}{\lambda_{ik}^2} f_{ik} \quad (35)$$

and combine this with the oscillator strength f_{ik} from ref (21) while noting that the multiplicities have been included in the calculations of ref (21), then we have for eq. (34),

$$\begin{aligned}\sigma^a &= \frac{1}{4} \times 0.6669 \times 5.60 \times 10^{-5} \times \frac{1}{2.89 \times 10^9} \\ &= 3.23 \times 10^{-15} \text{ cm}^2 \quad (36)\end{aligned}$$

or using eg. (15) and the multiplicities g and g' for the 13p and 4s states respectively, we have for the stimulated emission cross section for the 13p $\rightarrow$ 4s transition,

$$\begin{aligned}\sigma^{ST} &= \frac{6}{2} \times 3.23 \times 10^{-15} \text{ cm}^2 \\ &= 9.69 \times 10^{-15} \text{ cm}^2\end{aligned}\quad (37)$$

We would now like to examine the cross sections of the other transitions out of the 13p excited state. To do this we must calculate the linewidths of these lines. We note here that the collisional linewidths for these lines are unchanged and will still be much larger than the very narrow natural linewidths. The doppler linewidths, however, are directly proportional to the frequency of the transition ν_0 and will, therefore, decrease significantly as we reduce the frequency of the transitions. The linewidths of the transitions from the 13p-nd and 13p-ns states are shown in Table (2) and have been calculated using eq. (20) at a temperature of 617⁰K. We see from this that for the 13p-11d and the 13p-13s transitions, we can consider the collision linewidth as the dominant factor in the cross-section calculations. Therefore, for the FIR transition at 91.6 μ m between the 13p and 11d states, we have,

$$\Delta\nu = \Delta\nu_c = 2.69 \times 10^7 \text{ sec.}^{-1}\quad (38)$$

To be more correct we can get a first order estimate of the real linewidth by adding the collisional and doppler contributions together to get an upper limit. We then would have,

$$\Delta\nu = \Delta\nu_c + \Delta\nu_d = 3.47 \times 10^7 \text{ Hz} \quad (39)$$

We will use this in all further calculations concerning the 13p-11d transition.

Using eq. (39) and eq. (16) we can write,

$$\sigma^{ST} = \frac{1}{4 \times 3.47 \times 10^7} \lambda^2 A$$

and using eq. (35) we have,

$$\begin{aligned} \sigma^{ST} &= \frac{1}{1.39 \times 10^8} \times .6669 f_{ik} \\ &= 4.81 \times 10^{-9} f_{ik} \end{aligned}$$

from Table (2) we have,

$$\begin{aligned} f_{ik} (13p-11d) &= 2.29 \\ \Rightarrow \sigma^{ST} &= 1.10 \times 10^{-8} \end{aligned} \quad (40)$$

and using (11) we calculate the Einstein B coefficient for this transition,

$$\begin{aligned} B &= 4.26 \times 10^{54} \frac{A}{\nu_{ik}^3} \\ &= 4.26 \times 10^{54} \lambda_{ik}^3 A \frac{1}{c^3} \\ &= 4.26 \times 10^{54} \times .6669 \times \lambda_{ik} f_{ik} \frac{1}{c^3} \\ &= 2.63 \times 10^{21} \text{ cm}^2 \text{ J}^{-1} \text{ sec}^{-1} \end{aligned} \quad (41)$$

If we assume some intensity for the blackbody background such as,

$$I = \rho(\nu) \Delta\nu \quad C = \frac{8\pi h\nu^3}{c^2} \frac{1}{\exp(h\nu/kT)-1} \Delta\nu \quad (42)$$

$$= \frac{8\pi \times 6.63 \times 10^{-34} \times (2.75 \times 10^{12})^3}{(3 \times 10^{10})^2} \left[\frac{1}{\exp(6.63 \times 10^{-34} \times 2.75 \times 10^{12} / 1.38 \times 10^{-23} \times 617) - 1} \right] \\ \times 3.47 \times 10^7 \\ = 5.60 \times 10^{-8} \text{ J cm}^2 \text{ sec}^{-1} \quad (43)$$

We can also calculate the intensity from spontaneous decay. From the A coefficient given in (5) and the information in Table (2) we have for the 13p-11d transition,

$$A = 0.6669 \times (91.6)^2 \times 2.29 \\ = 1.28 \times 10^4 \text{ sec}^{-1} \quad (44)$$

The density of excited states, in the limit of saturation is,

$$n_0 = n_t/2 = 1.56 \times 10^{16} \text{ cm}^{-3} \quad (45)$$

After 10 nanoseconds we see that the number of atoms/cm³ left in the excited state is,

$$n(t) = n_0 e^{-At} \\ = (1.56 \times 10^{16}) \exp(-1.28 \times 10^4 \times 10^{-8}) \text{ cm}^{-3} \\ = 1.5598 \times 10^{16} \text{ cm}^{-3}$$

or the number of decays is

$$n_d = n_0 - n(t) = 2.00 \times 10^{12} \text{ cm}^{-3} \quad (46)$$

Each of these photons has the energy,

$$E = h\nu = 1.80 \times 10^{-21} \text{ J} \quad (47)$$

So we have a total energy flux out of the system of

$$\begin{aligned} F &= n_d E c \\ &= 1.08 \times 10^2 \text{ J cm}^{-2}\text{-sec} \end{aligned} \quad (48)$$

We see that this is much larger than (43a) and will dominate.

This energy is going into 4π steradians. We would like to look at the energy flux going into the system to stimulate other atoms to emit. To calculate the percentage of F stimulating further emissions, we first note that from (40), (45), and

$$l = \frac{1}{\sigma s \Gamma_n} \quad (49)$$

for the characteristic length before the stimulated emission occurs, we have,

$$l = 8.66 \times 10^{-9} \text{ cm} \quad (50)$$

This is exceedingly short, since the internuclear distance at this pressure is $\sim 5 \times 10^{-6} \text{ cm}$. We may consider all 4π steradians of the flux in a calculation for the total stimulated rate. So, from (41), (39), and (48),

$$\begin{aligned} W &= B I \omega \\ &= B F / \Delta \nu \\ &= 8.19 \times 10^{15} / \text{sec.} \end{aligned} \quad (51)$$

where we have used the linewidth $\Delta\nu$ in (39) because it is the characteristic linewidth for the spontaneous emission involved in I_ω .

This is the initial stimulated emission rate and will grow as the total energy emitted from this transition grows. We see that this rate is very fast and we will in practice be limited by the pump rate and duration of the laser used to pump the 13p upper excited state.

We should also note here that we are de-exciting the upper state much faster by stimulated emission than by any other method (i.e. spontaneous emission or collisions.) Therefore, any rate equations would be dominated completely by the stimulated emission rate.

To get a better feeling for the process, we should compare the time t when the stimulated rate exceeds the collisional rate. We, therefore, want to solve for t in the equation,

$$\begin{aligned}
 \Delta\nu_c &= W(t) \\
 &= B I_\omega(t) \\
 &= B F(t)/\Delta\nu \\
 &= B n_d(t) EC/\Delta\nu \\
 &= B EC/\Delta\nu (n_0 - n_0 e^{-At})
 \end{aligned} \tag{52}$$

Using the values of these variables given in eqs. (38), (39), (41), (44), and (45), we have,

$$\begin{aligned}
 2.69 \times 10^7/\text{sec} &= 4.09 \times 10^3 (1.56 \times 10^{16} - 1.56 \times 10^{16} \\
 &\quad e^{-1.28 \times 10^4 t})
 \end{aligned}$$

or

$$4.21 \times 10^{-13} = 1 - e^{-1.28 \times 10^4 t}$$

which yields

$$t \ll 1 \times 10^{-14} \text{ sec} \quad (53)$$

To more accurately describe this system, one must write the complete set of rate equations and solve them for the photon population as a function of time. This method is outlined extensively in Refs. (26,27) and will not be explored here. This method would give an accurate picture of the temporal behavior of the system, but because of the extremely large stimulated emission probability for our system, we would learn very little from the exercise.

One useful result from this type of calculation is the population density required in the upper state to get above threshold. As shown in Ref. (26), we can write this threshold population density as,

$$n_{th} = \frac{8\pi}{3} \frac{\Delta\nu}{\nu} \frac{\tau_{rad}}{\tau_c} \left(\frac{1}{\lambda}\right)^3 \quad (54)$$

Where τ_{rad} is the spontaneous lifetime of the state and τ_c is the cavity lifetime. In our case the cavity lifetime is the time light takes to travel one length of our excited state column. This is,

$$\begin{aligned} \tau_c &= \frac{15 \text{ cm}}{3 \times 10^{10} \text{ cm/sec}} \\ &= 5 \times 10^{-10} \text{ sec} \end{aligned} \quad (55)$$

Solving (54) for the 13p-11d transition we obtain,

$$n_{th} = 1.17 \times 10^7 / \text{cm}^3 \quad (56)$$

Comparing this with (45) we see that we are quite far above threshold under the conditions stated. This equation is derived for a steady state inversion and we must, therefore, approximate our laser as a square wave with a steady state level for approximately 8 nsec. This is actually a reasonable approximation and yields, for a pump rate to be above threshold,

$$\begin{aligned} R_{th} &= \frac{n_{th}}{t} = \frac{1.27 \times 10^7}{8 \times 10^{-9}} \text{ Atoms/cm}^3 \text{-sec} \\ &= 1.59 \times 10^{15} \text{ Atoms/cm}^3 \text{-sec} \end{aligned} \quad (57)$$

If we use the energy per photon to excite the upper state.

$$\begin{aligned} h\nu &= 6.63 \times 10^{-34} \times 1.02 \times 10^{15} \\ &= 6.76 \times 10^{-19} \text{ Joules/photon} \end{aligned} \quad (58)$$

and typical operating parameters of

$$\begin{aligned} d &= \text{beam diameter} = .2 \text{ cm} \\ l &= \text{column length} = 15 \text{ cm} \\ E_p &= \text{Pump energy} = 6 \times 10^{-6} \text{ Joules/pulse,} \end{aligned} \quad (59)$$

our available pump rate is,

$$R_p = \frac{E_p}{h\nu} \cdot \frac{1}{\pi \frac{d^2}{4} l} \cdot \frac{1}{t_p}$$

where t_p is the pulse duration of 8 nsec. This yields

$$R_p = 2.35 \times 10^{21} / \text{cm}^3\text{-sec} \quad (60)$$

which is much larger than the threshold pump rate in (57). We should, therefore, expect the system to operate in the stimulated emission mode when pumped as stated above.

So far we have discussed only the pump transition 4s-13p and the desired FIR emission transition 13p-11d. We have assumed a population inversion in the upper state (quite reasonable since our thermal energy is $\sim 432 \text{ cm}^{-1}$ compared to the 34000 cm^{-1} lower lasing level) but have not examined the depopulation processes for the lower level. These would include spontaneous emission, stimulated emission, and level changing collisions. We see from Table (4) that the natural lifetime of the 11d state is $4.5\mu \text{ sec}$, and therefore, will probably not be important in the total lifetime. In fact the arguments put forth for the 13p-11d transition can be invoked with virtually no change for the 11d-12p and 11d-10f transitions so we expect a very rapid depopulation of this level by stimulated emission with very little effect from collisions since the time between collisions is long (factor of 10) compared to the stimulated emission rate.

Therefore, the temporal behavior of the laser is expected to be completely controlled by the laser pump with the actual atomic processes happening at a rate that is so fast as to be unobservable.

To get an idea of the power out of the system in the FIR transition, we can make a very simple calculation based on the

TABLE 4. Transitions from 11d-np and 11d-nf with the A coefficients and lifetimes as well as the total A coefficient.

Transition	f_{ik}	$(E_k - E_i) \text{ eV}^{-1}$	$A_{ik} \text{ sec}^{-1}$
11d - 12p	1.95	83.96	9167.27
11d - 11p	.0352	319.46	2393.00
11d - 10p	.00437	645.36	1.215×10^3
11d - 9p	.000714	1114.82	5.918×10^2
11d - 8p	.0000672	1826.78	1.500×10^2
11d - 7p	.00000744	2982.44	4.413×10^1
11d - 6p	.000103	5049.20	1.744×10^3
11d - 5p	.000247	9336.70	1.434×10^4
11d - 4p	.000397	21014.01	1.160×10^5
11d - 10f	1.00*	146.7	1.435×10^4
11d - 9f	.113	404.9	1.235×10^4
11d - 8f	.0319	765.86	1.248×10^4
11d - 7f	.0118	1292.38	1.314×10^4
11d - 6f	.0047	2103.9	1.387×10^4
11d - 5f	.00175	3451.3	1.390×10^4
11d - 4f	.000476	5929.2	1.116×10^4

$$A_i (\text{TOT}) = 2.369 \times 10^5 \text{ sec}^{-1}$$

$$\tau_i = 4.22 \times 10^{-6} \text{ sec}$$

*extrapolation from reference (2)

Manley-Rowe limit. This is, for every photon absorbed one photon is emitted in the FIR.

Assuming now $1\mu\text{J}$ of incident pump power per pulse, if we are at the Manley-Rowe limit, we would expect a signal corresponding to,

$$\begin{aligned} E_{\text{FIR}} &= E_p \frac{\nu_{\text{FIR}}}{\nu_p} \\ &= 1 \times 10^{-6} \frac{2.75 \times 10^{12}}{1.02 \times 10^{15}} \\ &= 2.70 \times 10^{-9} \text{ J pulse}^{-1} \end{aligned} \quad (61)$$

which corresponds, in the limit of a square wave, to

$$\begin{aligned} P_{\text{FIR}} &= \frac{E_{\text{FIR}}}{t} \\ &= \frac{2.70 \times 10^{-9}}{8 \times 10^{-9}} \text{ J sec}^{-1} \\ &= 337 \text{ m Watts} \end{aligned} \quad (62)$$

for the peak power in the system.

The system is not controlled by a cavity geometry so we would expect less than the power indicated in (62) to exit the device through the output end. The fraction exiting this way will be determined by both the aspect ratio of the amplifying medium and by the rate of stimulated emission, as this will determine how much effect neighboring atoms have on each other. From a purely geometric consideration, we can argue that only that portion of

the FIR traveling down the column will be amplified, and therefore, we should expect,

$$a = \frac{f}{d} = 75$$

to be the ratio of the power out along the column to that which goes out in other directions. This means that 98.7% of the radiation will exit colinear to the column with a divergence associated with this aspect ratio of $\frac{1}{a} = 0.013$ radians. One can argue here that because of the extremely fast stimulated emission rate, we would only expect the FIR to exit in one direction, the direction the pump beam is traveling, because the stimulated emission will happen almost instantaneously after the medium has been excited.

Stark Effect

The second major theoretical area to be explored is the response of a highly excited atomic system to an applied electric field. Recently a detailed study of the Stark structure in the alkalis has been done with emphasis on the $n = 15$ levels (28). This work outlines the theoretical methods used to do detailed calculations on the Rydberg states of the alkalis and gives results for the $n = 15$ manifold of potassium. Here we will review the results and apply them to the system levels of interest.

We begin by writing Schrodinger's equation for the atom in a static electric field, neglecting spin,

$$\left[-\frac{1}{2} \Delta^2 + V(r) + F_z \right] \psi(\vec{r}) = W \psi(\vec{r}) \quad (63)$$

where W is the energy and F is the magnitude of the electric field which is taken to be along the z -axis. $V(r)$ is not known and the Hamiltonian is not separable so no analytical solution is possible. Because all levels are changing drastically with time a perturbation method is not suited to the problem. Constructing the wave functions by the use of Matrix diagonalization is, therefore, the most appropriate path to follow. Two approximations which prove very helpful and provide good results are; outside of the electron core, the potential $V(r)$ is essentially Coulombic and the off-diagonal matrix elements decrease very quickly as the energy difference between the states these represent increases. Therefore, only those elements that are near the level of interest need be included in the evaluation of the matrix.

Since the unperturbed energies for the alkalis are diagonal in the spherical representation and the Stark effect is not diagonal in the parabolic basis, it is more straightforward to work in the spherical representation.

The matrix elements of the Stark interaction may be written

$$\langle \omega, l, m / Fz / W', l', m' \rangle = \quad (64)$$

$$\delta_{l, l'} \delta_{m, m'} F \langle l, m / \cos \theta / l', m' \rangle \langle \omega, l / r / \omega', l' \rangle$$

We have the usual result for the angular portion using spherical harmonics,

$$\langle l, m/\cos \theta/l-1, m \rangle = \left[\frac{l^2 - m^2}{(2l+1)(2l-1)} \right]^{1/2} \quad (65)$$

$$\langle l, m/\cos \theta/l+1, m \rangle = \left[\frac{(l+1)^2 - m^2}{(2l+3)(2l+1)} \right]^{1/2}$$

The radial part of the wavefunction is all that remains to be evaluated and this has been done in Ref. (28) using numerical integration methods.

As mentioned in (28) the fine structure does not become important until the heavier alkalis. In the case of potassium we do not need to consider it, as it is much less than the $.3 \text{ cm}^{-1}$ resolution of our experimental apparatus.

The theoretical methods in (28) then provide us with a method for calculating the energy level shifts to an accuracy of $< .05 \text{ cm}^{-1}$. Recently this method has been extended to the $n^* = 11$ manifold of potassium (29) and the results of these calculations are shown in Table (5). We have shown here only those states that are of interest in our application although calculations for all values of l have been done. In Table (6) we see the shifts expected at 2000 volts/cm for the three transitions observed. We can write

$$E = E_0 + \alpha F^2 \quad (66)$$

where α is the polarizability of the level in $\text{cm}^{-1}/(\text{KV}/\text{cm})^2$, F is the field strength in KV/cm and E_0 is the unperturbed energy of the level. Using the convention for energy levels in (29) we have,

TABLE 5. Results from the calculations by Zimmermar, on the potassium Stark shift.

$M = 0$				
<u>State</u>	<u>Field KV/cm</u>	<u>0</u>	<u>1.5</u>	<u>3</u>
14s		785.45	785.82	786.88
13p		860.93	861.22	862.11
13s		937.35	937.55	938.16
11d		953.14	953.67	955.27
12p		1036.39	1036.55	2037.05

$M = 1$				
13p		860.93	861.18	861.93
11p		953.14	953.62	955.09
12p		1036.39	1036.53	1036.94

TABLE 6. Potassium shifts for 2000 V/cm.

<u>Transition</u>	<u>Predicted</u>	<u>Observed</u>
14s - 13p	$-.11 \text{ cm}^{-1}$	$-.59 \pm .15 \text{ cm}^{-1}$
13p - 11d	$+.42 \text{ cm}^{-1}$	$.43 \pm .15 \text{ cm}^{-1}$
11d - 12p	$-.66 \text{ cm}^{-1}$	$-.24 \pm .15 \text{ cm}^{-1}$

$$\begin{aligned}
\alpha(13p) &= 0.131 \text{ cm}^{-1}/(\text{KV}/\text{cm})^2 \\
\alpha(11d) &= 0.237 \text{ cm}^{-1}/(\text{KV}/\text{cm})^2 \\
\alpha(1f_s) &= 0.159 \text{ cm}^{-1}/(\text{KV}/\text{cm})^2 \\
\alpha(12p) &= 0.073 \text{ cm}^{-1}/(\text{KV}/\text{cm})^2
\end{aligned} \tag{67}$$

but

$$\Delta E = E - E_0 = \alpha F^2 \tag{68}$$

$$\Delta E_s = \Delta E_k - \Delta E_i = (\alpha_k - \alpha_i) F^2$$

so for the 13p $\rightarrow$ 11d transition

$$\Delta E_s(13p-11d) = 0.106 F^2 \text{ cm}^{-1} \tag{69}$$

similarly

$$\Delta E_s(14s-13p) = -0.028 F^2 \text{ cm}^{-1} \tag{70}$$

and

$$\Delta E_s(11d-12p) = -0.164 F^2 \text{ cm}^{-1} \tag{71}$$

Equations (69), (70), and (71) then give us the direction and magnitude of the shifts expected in the energies of these transitions. These shifts will be the observables in the experiment as well as the absolute shift of the upper pumped level as determined by the wavelength change of the pump laser.

III. EXPERIMENTAL

In the process of developing this FIR source, many different configurations for pumping the upper level of the atom were considered. Some of these will be described here with special emphasis on those that show some potential for further development. We will also describe a device known as a heat pipe oven, that has made possible the confinement of hot potassium vapor while still keeping optical access to the vapor. Finally the series of experiments demonstrating the generation of the FIR and then the tuning of the FIR will be described in detail.

As mentioned in the introduction, this work was prompted by results of experiments done at the IBM labs by Wynne & Sorokin (30) and by a proposal-type paper written by Lau et al. (31). In these papers C. W. operation of a FIR laser was postulated and pulsed operation demonstrated.

Early in the experimental phase of the work, it was decided that the stimulated emission results in (30) should be reproduced. This was done for two reasons. We wanted to get a measure of familiarity with the hardware involved in such experiments and a baseline was needed from which we could compare the results of later modified systems.

Heat Pipe

The first problem with the experimental work was the confinement of the alkali metal vapor at temperatures of up to 450°C. To address this problem we will examine the methods used to contain hot metals previous to 1970.

The most common system was one of a hot furnace with some combination of heated windows, baffles, or inert gas jets to keep the windows clear. These methods, however, all suffer from their own set of problems and one generally would end up with a poorly characterized vapor region that was not stable spatially or temporally. Sophisticated solutions to these problems have been used but with limited success.

Another common technique for the generation of pure metal vapor is the use of an atomic beam. This technique gives a collision free vapor on which to do spectroscopy, but is limited by the techniques needed to characterize the vapor and by the very low density of the vapor (typically $< 10^{12}$ atoms/cc). In a system where we expect to need a large number of atoms for the active medium, a beam would seem unsuited to the task.

Fortunately an alternative to these methods was developed by Vidal and Cooper (32) in the form of the heat pipe oven. This device is based on the heat pipe developed by Grover et al. (33) for the transfer of heat. The system uses the heat of vaporization of the metal as the heat transport mechanism and relies on the surface tension of the liquid metal to return the liquid to the heat source for recycling. A schematic of a simple heat pipe oven is shown in Figure (4). We see that it consists of a heater, cooling coils, stainless steel screen which acts as a wick, and inert gas. The potassium metal is placed in the middle of the heat pipe and a vacuum is pumped on the pipe. This is done at an elevated temperature but below the melting point of the metal

HEAT PIPE OVEN

Figure 4. A schematic diagram of a heat pipe oven.

(63.7°C) to remove any solvent used to clean the metal. We used cyclohexane for this purpose. When this step is complete, the pipe is filled with an inert gas, (in this case argon,) to the pressure at which the pipe will be operating. It is important to note here that the working pressure of the vapor will be determined solely by the pressure of the buffer gas. As more power is put into the heaters, the column length will increase. If the buffer gas reservoir is large the overall pressure will remain essentially unchanged. The hot metal atoms will collide with cold buffer gas atoms and will condense on the wick and flow back to the heated region because of surface tension. We can, therefore, control the pressure of the vapor very accurately by controlling the pressure of the buffer gas which means controlling the temperature of the buffer gas. For a system with a large buffer gas reservoir, we need control only the room temperature well to have satisfactory control over the vapor pressure. As shown in (34) one need only control the temperature of the reservoir to 2% to maintain the oven temperature to 0.1%. Therefore, in our laser lab where the temperature is controlled to $22^{\circ}\text{C} \pm 1^{\circ}\text{C}$ or 4.5%, we expect the oven to be controlled to better than .3%. As mentioned above, by adding heat to the system we do not increase the pressure of the system but rather increase the length of the vapor column. We can, therefore, change the density and size of the vapor accessed independently. This can be of great advantage when optimizing the experimental configuration. The boundary layers between the vapor and the buffer gas are extremely narrow

and constitute a small fraction of the total length of the vapor column Figure (5).

Our heat pipes have been operated with pressures of 0.1 to 10 Torr (270°C to 450°C) for up to 500 hours without window problems. The changing of windows was initially done in an argon filled glove box but later it was found sufficient, by observation of the potassium oxidation, to continuously flush the heat pipe with argon gas as the window was changed. Window materials included sapphire, quartz, polyethylene, and pyrex. All made suitable windows for spectral areas of interest.

The heat pipe illustrated in Figure (4) consisted of a 3/4" stainless steel (316 stainless) pipe 50 cm in length. The windows were attached by way of conflat varian flanges with copper gaskets and the wick material was 100 mesh stainless steel screen. The cooling coils were 3 turns of 1/8" copper tubing and the heater was a 200 watt heat tape controlled by a Variac variable transformer. All pressure measurements were done with an MK3 capacitive monometer with a 1000 Torr head. Temperature measurements were done with an iron-constantan (copper-nickel) thermocouple.

This heat pipe was used for most of the experiments described here but did not have the capability for the most important experiment, that of tuning the FIR output of the device. For this job a heat pipe with electric field plates was necessary and construction of two models were undertaken. The two models were drastically different in appearance, but quite similar in

Figure 5. The temperature profile of a heat pipe containing lithium. (32)

potential usefulness. The first model illustrated in Figure (6) was designed to have heated Stark plates which would be held approximately 5°C above the potassium reservoir temperature to keep the vapor from condensing on the plates. The plates would be totally inside the vapor region, being supported from the top by stainless-steel tubes which allow the heater and thermocouple wires to exit. The thermocouples were chromel-alumel (nickel-chromium, nickel-aluminum) and the heaters were controlled by a variac with isolation transformers to prevent an electrical feedback of the applied d.c. voltage into the a.c. line. A photograph of the completed heat pipe is shown in Figure (7). There are two small ports in the top of the plate support cover. One of these was used to view fluorescence in the pipe and the other was used to place a thin (1/8" diameter) stainless-steel electrode near the top of the plates to be used to detect ions formed between the electrodes. In the case where the Stark plates were not in use, one could use this electrode to probe for any ionized atoms formed by ionizing collisions of the highly excited atoms. A small voltage applied to this electrode would then induce a current that could be measured with a picoammeter. When the Stark plates were in use the current flowing in that system would be an indication of any ionizing processes taking place. This heat pipe was designed with a 0.5 cm spacing between the plates and did not have a stainless-steel screen for picking. The potassium liquid was to be returned by gravity feed. The windows of the heat pipe were attached to flexible stainless steel bellows

Figure 6. A schematic diagram of the "can" type heat pipe oven with Stark plates.

Figure 7. A photorraph of the "can" type heat pipe oven with Stark plates removed.

to allow the windows and hole coupled intra-pipe mirrors to be adjusted. The entire pipe assembly was cradled in a base consisting of aluminum brackets supported by invar steel rods 1" in diameter. The heater for the main pipe consisted of a mantle heater under the pipe and a 200 watt heat tape wrapped around the upper portion of the pipe and around the inner portion of the arms. The cooling consisted of 1/4" i.d. copper tubing wrapped twice around each arm about three inches from the main tube and around the main tube about four inches from the top. The main tube is six inch diameter 316 stainless steel and the arms are one and one-half inch 316 stainless steel inserted into the main tube at a 10° slope to allow the liquid potassium to flow back to the main reservoir area. A plexiglass hood was built to cover the exposed electrical connections to the field plates and all electrical connections were made through a single cable to the hood. A chassis was constructed to house the four thermocouple readouts (one for each cartridge heater in the plate supporting arms), the variac to control the cartridge heater, and the isolation transformer.

A potential problem with this design is that not all of the alkali metal vapor in the column defined by the pump laser is between the plates. At each end of the plates there is vapor that would be in either a very inhomogeneous field or zero field when voltage is applied to the plates. Fortunately this is not a significant problem because of the Stark response of the potassium vapor. The upper state is an $l = 1$ state and as seen in Chapter

II is shifted strongly in an electric field of moderate strength. Therefore, to continue to pump this upper level, we have to tune the pump laser back into resonance with the now-shifted upper state. This means that only that portion of the metal vapor that has been shifted will absorb the pump laser light and we have effectively circumvented the problem.

The second model of heat pipe designed for use with electric fields is quite different in design from the one just described. It consists of two large plates separated by an insulating element as illustrated in Figure (8). This device is very similar in design to one described in Ref. (35). It consists of two 316 stainless-steel plates 17" x 17" x 5/8". These are separated by a stainless-steel ring and are both insulated by 1/4" O.D. O-ring gaskets. There is also an epoxy composite insulating standoff between the plate and the ring on the non-load bearing surface Figure (9). One can think of this device as simply a normal heat pipe oven with 360° symmetry about an axis normal to the plane of the plates. Therefore, instead of having a working vapor in the shape of a column we have the vapor in a disk shape. Windows were placed conveniently around the pipe so a variety of measurements could be made with no need for changing windows. A sapphire input window was used to allow access to the pipe by both visible and U.V. radiation. Output windows used were Z-cut quartz and polyethylene to allow visible, U.V., and FIR radiation to be monitored. Both output windows were attached to the same side of the pipe and were separated by approximately 5 cm. The output

Figure 8. Sketch of the "pancake" heat pipe oven.

Figure 9. Cross section view of the "pancake" heat pipe oven.

could be monitored through either window by simply rotating the pipe. The very useful added feature of this heat pipe was the window which ran along the length of one of the walls adjacent to input and output windows. This window was approximately 1.5 cm high and 33 cm long and gave a clear side view of the excited alkali metal vapor when the pump laser operated. This aided alignment enormously and helped identify resonances quickly. Two heaters were used, one on each plate. They were 750 watt Kame?rok model 63102, 5 inch diameter ring type heaters and were insulated electrically (and unfortunately partially thermally) from the plates with a .004 inch thick mica sheet. A bolt was welded to the plate and a 1/2 inch thick aluminum plate was used to hold the heater to the stainless-steel heat pipe plate. This was also insulated from the heater by a mica sheet. All surfaces were coated with nonconductive heat sink compound to aid thermal transfer. Both heaters were controlled independently by variacs and the temperature of the stainless-steel plates were monitored by iron-constantan thermocouples. Cooling coils were made of 1/4 i.d. copper tubing and were formed into 30 cm diameter circles of two windings and were soldered onto the stainless-steel plates. Heat sink compound was also used on them to insure good thermal contact. These were connected to the Low Conductivity Water (LCW) supply by plastic hose. The LCW supply is a closed laboratory wide source of cooled deionized water which has, as its name implies, very low electrical conductivity. The two plates were held together with approximately 20 C-clamps which were insulated

from the plates by 1/8" thick epoxy resin board. The separating ring was insulated from both plates as stated above and was insulated from the vacuum system by the insertion of a glass transition in the vacuum line. The wicking of the upper plate consisted of a single layer of 100 mesh stainless-steel spot welded directly to the plate. The lower wick consisted of 100 mesh stainless-steel spot welded to a 1/8 inch thick stainless steel plate which was then bolted to the bottom plate. This was done to allow access to the potassium metal reservoir which consisted of a 3/8 inch deep area 2 inches square machined into the bottom plate. When loading the pipe, the potassium was set in the machined out area and the thinner plate bolted on top of it. This allowed us to keep all high melting point contaminants out of the working area. Several holes were drilled in the 1/8 inch plate above the reservoir area to allow the potassium vapor to enter the working area. In addition, to prevent thermal doming in the 1/8 inch plate, a circle of approximately 30 cm diameter was cut and the cut out piece was replaced and held into place by the screen. This left a 1/16 inch gap around the entire section which allowed the disk to expand without buckling when heated. The plate spacing was approximately 1.3 cm before heating but the doming effect of the large plates reduced this to less than 1 cm. The exact distance was not measured when hot. The uniformity of the field was only good to 15% due to this effect. To prevent thermal transfer through the plate outwards, a thermal groove was cut in a square pattern about 20 cm on each side centered about

the center of the plate and approximately 3/8 inch deep. This groove and the cooling coils kept the edges of the heat pipe below 60°C. At no time was fogging of the windows observed. The electric field was applied directly to the plates and was used in both a c.w. and pulsed mode. The lower plate was attached to ground and supported on the laser table by four lab jacks. The upper plate held the high voltage and the middle ring was floated to provide more uniform fields. Care was taken to pump a good vacuum on the system (approximately 10^{-6} Torr) before loading and whenever the heat pipe was cold. This eliminated any oxide build-up on the potassium. Loading was accomplished by placing thin (1/4 inch) potassium metal slices in the reservoir which was filled with cyclohexane to prevent oxidation of the potassium. A vacuum was then pumped on the system to evaporate the cyclohexane and heat was applied to increase the rate of evaporation. When the system held the vacuum satisfactorily, argon gas was put in at a suitable pressure (~1 Torr) and the device was heated to a temperature appropriate to the pressure (332°C at 1 Torr).

Configurations

In the course of the theoretical work, it became obvious that there were several ways to experimentally achieve the desired population inversion in the potassium vapor. Many different pumping schemes were explored on paper and some were tried experimentally in the laboratory. Here we will simply explain the experimental configurations tried and will leave the discussion of

the experiments done with these configurations to the last section of this chapter.

The first configuration tried is the same as in Ref. (30). We used a Molectron Model U.V. 1000 nitrogen laser to pump a Molectron D. L. 400 dye laser. The only element in the cavity to reduce the dye laser linewidth was a telescope to expand the beam on the grating and thereby improve the number of grating lines illuminated which narrows the dye laser output. This gives us a dye output bandwidth of $.5 \text{ cm}^{-1}$ (15 GHz). Pulses from the dye laser are typically 8 nsec in duration with an output energy of 500μ Joules per pulse at a repetition rate of 10 Hz using Rodamine 6G (R6G) as the active lasing dye. The power spectrum of this dye is shown in Figure (10). We see that the peak output is at 580 nm. Our measurements were all done at 585 nm. To excite the 13p state of potassium, we need the second harmonic of the dye laser output. This wavelength is $\sim 292.5 \text{ nm}$. To obtain the second harmonic one must focus the laser beam down onto a crystal with a high non-linear index which effectively mixes the beam with itself and generates photons of twice the energy. For the wavelengths in which we are interested, the suitable crystal for the job is KDP (potassium dihydrogen phosphate) in an angle tuned configuration. Typical doubling efficiency was 3% which gave an output energy of $10\text{-}15 \mu$ joules per pulse in the U.V. with a pulse duration of approximately 4 nsec. The output beam was then re-collimated before entering the heat pipe. This configuration is illustrated in Figure (11).

Figure 10. Power spectrum for R6G laser dye when used in a Molectron DL-200 laser pumped by a Molectron UV-1000 N_2 laser.

Figure 11. The experimental configuration used with the N_2 -Dye laser pump.

The second configuration attempted was similar in that we attempted to pump the upper level with a single U.V. photon. In this configuration we used a Chromatix Model CMX-4 flasnlamp pumped dye laser. The overall energy from this system was substantially higher than in the Molelectron system. Typical visible output was 1 m Joule at 585 nm, but the pulse was now approximately 1μ sec long. This system was much more efficient, however, in its production of U.V. light. The doubling crystal was placed inside the cavity and the output mirror of the laser was changed to a dichroic mirror which is very highly reflective in the visible, thereby keeping a very high finesse cavity for visible lasing, but also transmits greater than 80% of the ultraviolet light at the wavelength of interest. We, therefore, have the very high circulating power in the dye cavity to double with instead of the usual 20% dye output. This system was routinely operated with a U.V. output of 200μ joules per pulse at a 10 Hz repetition rate with $.5\text{ cm}^{-1}$ linewidth and 1μ second pulse duration. Because of the long pulse length, the peak power of this pulse is only 200 watts compared to 2500 watts for the Molelectron system. So the advantage is total energy emitted and the disadvantage is the peak power of the pulse. The Chromatix also had the minor advantage of a more compact unit. A schematic of this pump scheme is illustrated in Figure (12).

The next pump scheme uses a quite different approach to accessing the upper state of the atom. Instead of using a single photon of highly energetic light, which is quite costly to obtain

Figure 12. The experimental configuration used with the Chromatix CMX-4 dye laser pump.

(in terms of system efficiency), we could also use two photons of suitable wavelengths to sequentially pump a lower excited state and from there pump up to the excited state of interest. We should note here that the final state in the case of potassium would then be an $l = 0$ or $l = 2$ state, since the first excited state pumped would be an $l = 1$ state. This would not be a problem, however, in the operation of the device. Since the final upper state in potassium would lie in the range of $34,000 \text{ cm}^{-1}$ above the ground state, we would like to access a lower excited state very nearly half-way above the ground state. The first excited state in potassium lies at approximately $13,000 \text{ cm}^{-1}$ above the ground state so we would, therefore, need a second photon of about $21,000 \text{ cm}^{-1}$ or 475 nm wavelength to reach the upper state. This second wavelength is quite easily accessible with a flashlamp pumped dye laser. We could also use a krypton pumped dye laser in the c.w. mode. Because we only had one Kr^+ laser available, and the pulsed system would not operate at $13,000 \text{ cm}^{-1}$ (767 nm) we could not operate in a completely c.w. or pulsed mode. An attempt was made to operate in a hybrid mode where we would pump the first excited state with a krypton ion dye laser using an oxazine dye followed by a pulsed flashlamp pumped dye laser operating at 475 nm using the Exiton laser dye L.D. 473.

During this period of pump choice, many other systems were proposed. We will mention here one of those choices and why it was not tried. Since this work began, another large effort in the

group here has been to develop a synchronously pumped dye laser system Ref. (36, 37, 38, 39). We, therefore, explored the possibility of using these extremely short (10p sec) pulses with high peak power (1 K watt when cavity dumped) to efficiently generate tunable U.V. light. This at first would seem like a very attractive system because the pulse rate is very high (80 MHz without cavity dumping and 4 MHz with cavity dumping) and we could do quasi-C.W. spectroscopy with relatively high efficiency in our doubling crystal due to the high peak power. The very fast repetition rate was actually the problem with the system when we look at our method for detecting the FIR output of the device. As will be discussed in the next section, all of the FIR detectors available to us are of the thermal type, and therefore, have associated with them long (1 msec) time constants which make them unsuitable for a fast repetition rate system. They are in fact much less than ideal for the extremely fast pulses used in all of the configurations so far discussed. We would not gain any signal to noise advantage because we could not use any temporal filtering. The best mode for the detectors used would have been a C.W. pumping system chopped at a low rate (~ 10 -15 Hz). As mentioned above, the appropriate pump lasers were not available.

Only a few of the many possible ways of exciting the upper state have been mentioned here, but for the purpose of the experiment the ones mentioned were most appropriate in terms of results and equipment available.

Experimental Procedure

The experimental work began with the construction of a working heat pipe oven. As mentioned in the first section of this chapter, we began with a simple device approximately 50 cm long and 1.9 cm in diameter. With this apparatus, we had three distinct goals in mind. The first was to characterize the operation of the oven itself. The second was to do spectroscopy on the potassium vapor using white light sources as well as the laser systems mentioned above. The last goal for this device was to observe FIR stimulated emission and observe the Stark shifted emission.

We, therefore, began by operating the heat pipe under conditions ranging in pressure from .1 Torr to 10 Torr and in temperature from 270°C to 405°C. A simple but effective diagnostic for the presence of potassium vapor was the use of a flashlight. When the pipe is operating properly the transmitted light takes on a greenish tint when shined through the vapor. This is the result of very strong absorption in the red part of the spectrum due to the presence of molecular potassium at about the 1% level. Another straightforward method for observing the presence and position of the K vapor was a logical extension of the above experiment. A small HeNe laser was used to probe the vapor and once again was absorbed by the 1% of molecular species which in turn fluoresced in the red and appeared as a bright red column the diameter of the HeNe laser beam and the length of the potassium vapor region. The laser beam was not absorbed completely and the column in the heat pipe appeared uniform in intensity at all

pressures tested. At the higher pressures it was much brighter than at the lower pressures due to the higher total vapor pressure of potassium in the pipe and because at higher temperatures the percentage of molecular potassium increases. In all experimental arrangements, these two simple diagnostics proved invaluable in diagnosing problems associated with the heat pipe itself.

The next step in the characterization of the system was the absorption spectroscopy of the potassium vapor. A variety of white light sources were used to observe the absorption in the vapor, but the broadest source with the fewest annoying features (such as spikes) turned out to be a quartz halogen lamp. The output of the lamp was put directly into the heat pipe and the output of the heat pipe was then focussed into either a Spex 3/4 meter monochrometer Model 1802 or a Spex Doublemate Model 1672 double monochrometer. The 1802 has higher resolution but the 1672 was generally more available and had enough resolution for most applications. The detectors used were generally either an RCA 1P28 or RCA 8850 photomultipliers. Both tubes have very little response past 700 nm so our spectra were generally limited to the 250 nm to 700 nm range. Due to the low brightness of the lamp and the broadness of the detector-monochrometer system, only the strongest lines were expected to be seen. Even though our detection system was very poor above 700 nm the features at 760 nm were expected because they are the strongest lines in potassium. This feature and two others were observed. They appeared at 590

nm and 404 nm. The 404 nm line is attributed to the 4s-5p transition, the 766 nm line to the 4s-4p transition and the 590 nm line to the 4p-5d or 4p-7s transitions. When the lamp output was run through the monochrometer first and then into the heat pipe, only the 4s-4p and 4s-5p transitions were observed.

These observations assured us that the vapor in the heat pipe was indeed potassium and that the heat pipe was working properly. Since the detection bandwidth ($\sim 20 \text{ cm}^{-1}$) was much greater than the linewidth of the transitions involved ($\sim .03 \text{ cm}^{-1}$), we were assured that even a small detected absorption was actually very substantial.

The next step was to attempt to observe U.V. absorption in the 4s-13p transition. Using a very narrow ($.002 \text{ nm}$ or $.15 \text{ cm}^{-1}$ linewidth) tunable dye laser we attempted to pump this transition for the first time. The system used consisted of a Spectra-Physics Model 171-09 argon ion laser pumping a Spectra-Physics Model 375 tunable dye laser with an optional intracavity etalon to reduce the operating linewidth. The output of this laser was then focussed with a 10 cm focal length lens into a KDP doubling crystal and the output of the crystal was recollimated and passed through a Corning 7-54 glass filter which passes the U.V. light but not the visible. This beam was then directed into the heat pipe by suitable mirrors and detected as it exited the heat pipe.

A set of Melles Griot ultraviolet grade neutral density filters was used to attenuate the laser beam so it would not saturate the detector. The detector used was an EG&G model UV-444B photodiode. The output of this photodiode was put directly onto a

chart recorder or amplified by an operational amplifier and then put out to the chart recorder. When operating at 292 nm a sharp resonance was observed. Since the etalon had to be hand tuned and only had a tuning range of 1 nm the discovery of the absorption took several hours to accomplish. However, once tuned to the absorption, the system showed excellent stability and could be shut-off and restarted the following day and remain on the desired absorption line. Figure (13) shows a typical hand scan of the etalon. We see the laser output powers vary over the range of the scan and a sharp absorption near the middle of this range. The total scan magnitude is approximately 1 nm. The ultraviolet power entering the system was too small to accurately measure with the power meters available. We can see from Fig. 13 that when tuned onto the absorption, approximately 40% of the laser light was absorbed.

The neutral density filters were then removed and fluorescence from the excited states was looked for. None was observed from the output end of the pipe. The total ultraviolet power was still very low because of the poor doubling efficiency of a c.w. system. It was decided that the best possibility of observing the fluorescence was to look back at the excited atoms from the input and of the pipe. This would put the observer as close as possible to the excited vapor. A front surfaced mirror was constructed with a small (2 mm) hole in the middle to let the U.V. into the pipe. It was placed at 45° to the laser beam and an appropriate lens was used to focus the output into a 1/4 meter Jobin Yvon

Figure 13. A typical etalon scan of the Spectra-Physics 375 dye laser showing the 4s-12p doublet.

Model H20V monochrometer. This was selected because of its very high throughput. The detector was once again an RCA 1P28 photomultiplier tube. The output of the tube was put into a Kiethley Model 417K chromatograph electrometer and the output of this was put into a strip chart recorder.

No fluorescence was observed with this configuration either, so it was decided to pump a lower transition which would absorb more efficiently and possibly have a corresponding higher fluorescence output. The 4s-8p level was chosen because of its strength (the Einstein A coefficient for 4s-8p is ~ 10 times greater than that for 4s-13p) and the U.V. output of the pumping system was about the same. No visible fluorescence was observed.

It was then decided that we should attempt to observe the FIR region of the spectrum since the first transitions out of the upper states are in this region. The apparatus was set up as shown in Figure (14). The U.V. input was chopped at 10 Hz and the output of the Golay detector was put into a Princeton Applied Research (PAR) Model HR-8 lock-in amplifier with a type D differential pre-amplifier plug in. The output of the heat pipe was focused into the Golay cell by a 30 cm focal length polyethylene lens 5 cm in diameter. A black polyethylene filter was used to block all wavelengths shorter than $50\mu\text{m}$ from entering the detector. No signal could be observed with the lock-in amplifier when a time constant of 10 seconds was used. This confirmed the early suspicion that we would have to go to a pulsed system.

Figure 14. The configuration used for C.W. spectroscopy using the Spectra-Physics 170 A⁺ -375 dye laser pump.

At about this time, it was suggested that a useful method for finding the absorption lines in potassium might be obtained by using the opto-galvanic effect (40). In this configuration the laser is directed into a hollow cathode discharge tube and excites the atom to an upper state at which time it is ionized due to the high effective electron temperature. The signal is then detected as a voltage change on the electrodes of the hollow cathode tube. This is a potentially very sensitive device for locating resonances in any atomic species that can be operated in a hollow cathode tube or a discharge. In our experiments, a commercial hollow cathode tube, Westinghouse Model WL-22863A, with potassium in a neon atmosphere was used with a constant current source running at 15.5 mAmps. We had approximately 350 mW of visible power chopped at a 1.8 KHz rate. The voltage across the electrodes was monitored through a 5 pF capacitor by the PAR lock-in amplifier with a 100 M Ω input impedance. The output of the PAR was put onto a strip chart recorder. The output is shown in Figure (15). This scan is centered at about 580 nm, but was not well calibrated due to the need for hand tuning of the dye laser etalon. We see that there are at least three features in the scan. These would correspond quite well to the 4p-5d and 4p-7s transitions in potassium. After this initial success, we decided to try to use the same effect to locate the U.V. transitions. Unfortunately the commercial hollow cathode tube was equipped with a pyrex glass window which was unsuitable for ultraviolet. An attempt to use a demountable hollow cathode tube loaded with potassium and operated with argon

Figure 15. The opto-galvanic signal associated with the group of lines near 580 nm.

as the fill gas was unsuccessful. The tube, designed in the Chemistry Department at Lawrence Livermore Laboratory (41) was a much different configuration than the commercial model, so the operating parameters could not be compared directly.

We now began once again to look at the heat pipe oven for an absorption. The molelectron laser system described above and shown in Figure (11) was used to look at the 4s-13p transition. Using the lock-in amplifier, we found the absorptions at both the 4s-13p transition and the 4s-12p transition. The ultraviolet detector was replaced with the Golay cell. The output of the heat pipe was focused into the Golay with the polyethylene lens and filtered with black polyethylene. A signal was observed which depended critically on the tuning of the dye laser and the temperature of the heat pipe. The pressure in the heat pipe was raised from 2 Torr to 7 Torr to more closely reproduce the results of Ref. (30). With the lock-in amplifier time constant raised to 100 sec., we achieved a signal to noise ratio of approximately 10. It was at this point that our nitrogen laser failed.

The next step in our procedure was to try another available pumping configuration. At this point we had a krypton ion laser available as well as a Chromatex CMX-4 flashlamp pumped dye laser. It was decided to try to excite the K vapor to the first excited state using the ion laser to pump a near infrared dye and then use the flashlamp pumped laser to excite these excited atoms to the desired upper state. The experiment was set up as in Figure (16). The krypton ion laser, a Spectra-physics Model

Figure 16. The configuration used for the C.W. Kr^+ -dye laser, CMX-4 pulsed laser experiments.

171-09, was used to pump a Spectra-Physics Model 375 dye laser with Oxazine 725 as the working dye. The first objective was to find the 4s-4p absorption at 757 nm and 759 nm. With the heat pipe operative at 340°C and 300 mW of dye laser output, we saw total absorption of the near infrared laser light by the dye. Using our hole coupled input mirror at 45°, we saw very bright fluorescence in the tube in the yellow-green region of the visible spectrum. If all of the processes were single photon, we would not expect to see any visible light. It was also observed at this point that the spectrum changed as we tuned the dye laser on or off of the very strong atom lines. The fluorescence did not disappear completely when the dye laser was tuned off. It was also observed that the red K_2 band was very prominent when the dye laser was tuned off of the atomic lines and the intensity of this band dropped sharply when the dye laser was tuned to the atomic line.

The yellow-green features of this part of the experiment were investigated more carefully with the Spex 1672 double monochromator. Using .25 mm slits on the monochromator, we had 5 Å resolution which is enough to resolve any of the atomic doublets in this region. Scans were made under various pumping and heat pipe conditions and the results will be presented in the next chapter with a possible explanation for these observations.

After the short detour into the apparent multiphoton processes in the potassium vapor, it was clear that any energy lost here would not effect the FIR performance. In the process of

recalculating the parameters for the two photon pumping scheme, however, it became obvious that the duty cycle of the blue second photon laser would be mismatched to the first photon pump. We could saturate the first transition with the ion pumped dye laser, but the pump rate of this system could not match the pump rate of the pulsed system and we would, therefore, waste most of the second laser pulse. It was calculated that less than .1% of the pulse would be absorbed. A brief experiment confirmed our calculations. We could see little or no absorption of the blue in the once excited vapor.

The only method available then was a pulsed single photon method. The Chromatix CMX-4 used for the pulsed part of the above experiment also has the ability to be intra-cavity doubled using various KDP crystals cut at the appropriate angles for phase matching at several wavelength ranges. It also has a choice of several intra-cavity etalons available to narrow the linewidth of the system (at the expense of total output energy). The system was set up with R6G as the lasing dye, the low finesse ($.5 \text{ cm}^{-1}$ linewidth) etalon, and the appropriate optics to allow intracavity doubling. We obtained .2 mJ per pulse output energy with a pulse duration of approximately $1\mu \text{ sec}$.

The heat pipe was operated at 1 Torr and the system was set up as in Figure (12). The laser was scanned electronically which keeps the etalon and the Lyot filter scanning at the same rate. The dye laser was scanned in the region of 292 nm and the desired absorption was found. Actually two absorptions were found

approximately 10 cm^{-1} apart, but this was found to be a problem with the scanning dye laser. The resolution of the system was too low to permit the observation of the doublet $13p\ 1/2$ and $13p\ 3/2$ states. The Golay cell was then positioned at the output of the heat pipe and a black polyethylene filter was used to filter out all radiation with wavelengths shorter than $50\mu\text{ M}$. Using the PAR lock-in amplifier a signal was observed out of the Golay cell. The signal depended on the tuning of the dye laser and the temperature of the heat pipe, but was very weak. Several focusing arrangements were tried, but the expected output in the FIR was never achieved. The pressure was raised to 5 Torr and the experiments with focusing and tuning repeated with negative results. The energy was being absorbed, but was not coming out in stimulated emission along the axis of the pipe.

It appeared as if the cavity needed to be defined better to eliminate the possibility of stimulated emission off-axis. Mirrors were installed in the heat pipe on flexible stainless-steel bellows. They were hole-coupled polished stainless steel ($\lambda/4$ at optical wavelengths) mirrors with a 1 mm input hole and 1.5 mm output hole. The input mirror had a 1.5 M radius. These were aligned optically using a He Ne laser. The absorption spectroscopy was done with a PAR Model 162 boxcar integrator, using two channels and a ratio function. Both detectors were again EG&G UV-444B photodiodes. The beam was split at the input with a piece of z-cut quartz and directed into one of the photodiodes as a reference. The main beam entered the heat pipe through a sapphire

window and exited through a polyethylene window. Because polyethylene does not pass ultraviolet light well, the detector actually looked at the fluorescence of the polyethylene due to the U.V. light. Care was taken to eliminate any detector saturation problems. Absorption was found quite easily, but the FIR was not observed.

During the period several series of experiments were run to try to optimize the FIR output. The system was run from .7 Torr to 8 Torr with no FIR output observed. Focusing was also varied from 25 cm to an essentially collimated beam, again with negative results. It appeared now that a fundamental problem was showing itself. Since all other variables had been changed without success, we decided to return to the short (5 nsec) pulse system and try to recover the FIR signal. Beam time on a Molelectron nitrogen pumped dye laser system was arranged and the apparatus was set up. The dye output of the laser was 36KW peak which converts to about 290μ J per pulse. This was then doubled with the characteristic 4% to give approximately 11.5μ J per pulse in the U.V. Using the boxcar to detect the absorption, we scanned the dye laser. The absorption was found to be equivalent to an optical density of .23 and very narrow. This optical density may be related to the intensities by,

$$\text{O.D.} = \log_{10} \left(\frac{I_i}{I_t} \right) \quad (1)$$

where I_i is the intensity of the incident light and I_t is the intensity of the transmitted light.

Once again the Golay cell was set up and the appropriate filters were used. The lock-in amplifier was connected and used with a 100 sec time, constant. A strong signal was observed. With a time constant of 30 seconds on the lock-in, a signal to noise of 6:1 was observed. The laser was then tuned off of the resonance by .04 nm and the signal went away completely. When the laser was brought back to the same place, the signal returned. This was done several times on both sides (higher and lower frequency) of the atomic resonance. The Z-cut quartz beam splitter was then removed and the signal increased greater than 10%. With a 10 second time constant the signal to noise was 3:1. Being able to reduce the time constant was very important because the laser parameters could be adjusted in a reasonable amount of time. The Corning 7-54 filter was removed to let all of the visible into the heat pipe as well as remove any attenuation of the U.V. There was no apparent effect on the FIR power measured. The insertion and removal of the beam splitter appeared to make a 30% difference in FIR power measured at this point. The difference between this measurement and the earlier one, could have been due to different insertion angles. The earlier ones were done with the disk inserted closer to Brewster's angle. At this time, several things were done to try to improve the output of the device. The position of the device was changed with respect to the focal point of the beam and no change was apparent. The output was focused with several different polyethylene lenses, but again no improvement was seen. An estimate of the power out in

the FIR was made. The lock-in indicated a signal of 1×10^{-4} volts out of the Golay. The responsivity of the detector is 1.5×10^6 volts/watt so we apparently have 6.67×10^{-11} Watts average or 1.3×10^{-3} watts peak for a pulse duration of 5 nsec and a repetition rate of 10 Hz. This means about 6.5 picojoules per pulse were being generated.

The dye laser, as mentioned earlier has a linewidth of $.5 \text{ cm}^{-1}$. In the last chapter we saw that the absorption linewidth for the potassium should be about $.033 \text{ cm}^{-1}$. We, therefore, tried to narrow the dye laser linewidth to $.1 \text{ cm}^{-1}$ using an intracavity etalon. The insertion of this dropped the output intensity of the dye by 70% and the U.V. output even further (due to the I^2 dependent efficiency of the doubling crystal). The FIR output was not changed much although the apparent optical density of the vapor increased dramatically. The stability of the laser suffered markedly from the insertion of the etalon so we removed it and did all subsequent experiments with the laser operating with a $.5 \text{ cm}^{-1}$ linewidth.

With some degree of confidence in our pump method, we decided to continue on to the next step of the experiment. Testing the first Stark cell heat pipe oven began. The configuration of the pipe was quite different from that of the normal pipe and the operating characteristics needed to be checked. The mirrors were installed on the pipe at the outset of the experiments because of the added difficulty of working on this system in a glove box. This addition made broad band laser absorption spectroscopy

difficult due to the beam movement during scanning. Vapor was successfully generated in the pipe and the U.V. laser absorption was located. The FIR output was found, but the potassium reservoir ran dry quite rapidly. This proved to be a fundamental problem with the design of the pipe. The ratio of the diameter of the central "can" to its height was too large to allow sufficient cooling near the base of the can. This in turn allowed much of the potassium to reach the cold region where it solidified. The lack of wicking also proved to be a major problem for the return of liquid potassium to the reservoir area. Several modifications of this device were tried, but it was concluded that nothing short of a complete redesign would be suitable for the experiments envisioned. This was not undertaken because the second design was already in the process of being built.

At about this time it also became obvious that a dedicated pump laser system to provide the tunable U.V. laser light would be needed. A suitable system was found in the Laser Isotope Separation program at LLNL. It was obtained to use the system in our laboratory for an extended period of time. The system was a Molelectron U.V. 1000 nitrogen laser, D L-400 Dye Laser system identical to the one previously used except for the welcome addition of a stepper motor scan option which allowed .001 nm scan resolution and direct wavelength readout. This was very useful in scanning the dye laser output over a small range and then being able to return to the exact spot of the desired feature. Since the doubling crystal is manually tuned for phase matching, the

scan limit for the U.V. is about 1 nm before the crystal has to be retuned. This did not prove to be a problem. The output of this dye laser was found to be calibrated extremely well with the wavelength dial on the chassis. This proved to be quite convenient when looking at different levels in the potassium. The output power of the nitrogen laser also appeared to be quite good which allowed us to run it at a lower voltage to maintain stability.

The first set of experiments with this laser system were to find the FIR with our conventional heat pipe oven. All mirrors were removed as they had shown no advantage on the system previously and had made absorption spectroscopy difficult. They were not used again. The heat pipe was set up as in Figure (11) with the boxcar to detect the absorption. The 4s-13p absorption was found very easily in the heat pipe running at 1 Torr. The FIR signal was found immediately with an increase in signal level over previous measurements of a factor of 5. The output of the Golay cell was again put into the lock-in with a 1 second time constant and a signal to noise ratio of 5 was achieved. This allowed us to adjust optics directly as we watched the lock-in to peak up the signal. It was clear, however, that better signal to noise ratio would be needed if we were to go through a wavelength measuring device and still be able to be detected. To solve this problem, two potentially useful bolometers which are suitable for the submillimeter part of the spectrum were located. The first is a Molectron Model SB15-202(SL) Silicon Bolometer Cryostat System,

which consists of a Model DB2-1000 silicon bolometer, a Model DW20 liquid helium dewer and a Model AB-10 pre-amplifier. This system was cooled several times, but was never working satisfactorily. The pre-amplifier seemed to be the problem area for the system, but the exact cause of the problem was never fully elucidated. The second bolometer system used was a Texas Instruments Model 170866 Bolometer system. This system consisted of a gallium doped germanium (Ga:Ge) bolometer, a Texas Instrument Model CLF 3 cryoflask, and a preamplifier. This system was particularly well suited to the task because it covered the region of the submillimeter spectrum of interest ($20\text{--}200\text{ cm}^{-1}$) and had very little response outside this region. This is partly due to the response of the detector itself and partly because of the cooled sapphire and black polyethylene filters which cut off all higher frequencies.

The bolometer was cooled to 4.2°K with liquid helium and the output of the pre-amplifier was put into the lock-in amplifier. Although the output level on the lock-in was about the same as that of the Golay cell, the signal to noise was much better. The pre-amplifier could be used in two modes. One was with a variable bias control and the second was with a fixed bias set according to factory supplied calibration charts. In the first mode the signal to noise was 10:1 with the current reading $1.5\mu\text{ A}$ on the pre-amplifier. This is to be compared to the 5:1 signal to noise output of the Golay cell under identical conditions. In the second mode, with the internal bias control, the

current read 8.2μ A and the observed signal to noise was 40:1 with a 1 sec. time constant.

To illustrate the relative performance of these detectors several scans were made of the U.V. pump laser while monitoring the FIR output of the heat pipe. These are shown in Figure (17). We can see from this the dramatic decrease in noise obtained by using the bolometer. This advantage is somewhat offset by the increased difficulty of using the bolometer and by the limited bandwidth of the device for some measurements in other wavelength regions.

At this point it was clear the signal to noise ratio was good enough to attempt a wavelength measurement, but the beam characteristics were uncertain. If we hoped to see the radiation at a. on the other side of a wavelength measuring device the beam's divergence would have to be controlled. To do this several different polyethylene lens were tried and it was found that the beam could be controlled quite easily by simple optical techniques. For instance we found that a 30 cm focal length lens focused the beam sufficiently that there was little or no signal loss when the bolometer was placed 31 cm from the lens. The focusing of the pump beam had very little effect on the FIR output so the pump beam was collimated to obtain the best possible output beam quality. The FIR output was very well defined along the axis of the U.V. pump beam as evidenced by the sharp dependency of the FIR signal to the spatial position of the detector.

Figure 17. FIR output from the detectors as the pump laser is scanned.

- a) the golay cell
- b) the Ga:Ge bolometer with $<2\mu$ A current,
- c) the Ga:Ge bolometer with 8.2μ A current.

Our first attempt to measure the wavelength of the FIR signal was simulated by the use of microwaves. A Fabry-Perot interferometer was constructed using one 6 inch diameter front surfaced flat mirror and one 6 inch diameter one meter focal length front surfaced mirror. The microwave power was injected with a beam splitter and the circulating power was monitored near the edge of the curved mirror. The microwave frequencies could be measured to about 10 megahertz. Using a modulated microwave source (1 μ sec pulses at 10 Hz) we simulated the FIR system used the Golay as our detector. Using the lock-in amplifier we detected the microwave signal at the one milliwatt peak level. This was, however, a 1 μ second pulse, and therefore, much more energy than would be present in the FIR. The FIR was then injected into the cavity with negative results. It is postulated that cavity losses are greater for the FIR and as mentioned above, energy available is considerably less.

It was therefore decided to go to a high throughput system such as a Michelson interferometer. The advantages of this system are low losses, since it is a single pass system, high finesse, and ease of construction. The interferometer was constructed from two flat, front surface, 6 inch mirrors mounted on standard NRC mirror mounts and moved by Ardel-Kinematic translation stages with fine translation controls. Translation resolution was 1 μ m and total travel was 2.5 cm. The beam splitter was the most critical component of the system and many materials were considered. Mylar was the material of choice and 12.5 μ M mylar was selected as the

correct thickness for this wavelength range (42). One mirror remained stationary and the other was scanned by either a synchronous motor or a stepper motor. Both scan methods provided satisfactory results. The system was aligned by removing the heat pipe from the dye laser beam path and allowing the visible dye output go through the interferometer. The mirrors were aligned by observing the interference pattern of the laser on the output polyethylene lens. The system was then scanned and checked to insure good alignment throughout the scan. The heat pipe was then placed back into the beam path and the throughput of the interferometer was measured. The system routinely passed 10% of the total input power when a polyethylene lens is used to focus the FIR into the interferometer. The beamsplitter was enlarged and the mylar stretched over a polystyrene frame to give a good optical surface and prevent any aperturing due to the beamsplitter frame. Initial spectra were then run using the Golay cell as the detector. The initial results were very encouraging. A definite interferometric pattern was seen. Another problem that developed rather suddenly was the damage done to the instrument by an earthquake. The detector which had been sitting on the laser table, fell off and damaged the helium fill tube. The Molelectron dewar was available however and were able to move the Ga:Ge bolometer to this dewar and still use it. A good signal to noise was obtained with this system but we still had the problem of data acquisition and reduction.

At this point we decided to incorporate a digital computer into the apparatus to take, analyze, and display the data. In this respect we were fortunate to have available a Tektronix 4051 graphic computer system for a dedicated application. Although the system had been used in conjunction with experiments before, there was no applicable software available. The project became, therefore, to interface this system to our existing instrument. The system was quite straightforward to work with and the necessary program was written quickly to take the readings off of a digital voltmeter attached to the output of the lock-in amplifier. The data was then normalized to eliminate a d.c. offset due to poor modulation in the interferometer. This was a minor problem stemming from the less than perfect mirror mounting system used which created misalignment during a scan. The data were now ready for reduction by Fourier transform techniques.

The Tektronix computer system proved quite valuable at this point by having available a hard wired ROM (read only memory) pack with a Fast Fourier Transform (FFT) program, a Taper program to provide a Hanning function, and a rectangular to polar conversion to go from the real and imaginary output of the FFT program to an amplitude and phase output which was much more useful for our purposes (43). The Taper program was the first used in our data reduction process. This function compensated for what is sometimes termed "leakage." Leakage occurs when we acquire periodic data which is not an integral number of cycles and transform this data. This generates broadening and attenuation of the spectral

lines due to the violation of some assumptions that are made in the FFT program. To alleviate this problem we multiply the time domain waveform by a window function to remove the discontinuities in the data. The function used here is the Hanning function and is given by

$$x(t) = \frac{1}{2} - \frac{1}{2} \cos \frac{2\pi t}{T_C}, \quad 0 \leq t \leq T_C \quad (2)$$

where T_C is the total scan time. This is the form of the function for 50% taper or a true Hanning function. We also have available, with this program, the option of reducing the taper to as little as 10% or eliminating it all together. Figure (18) shows the taper function for 10%, 25%, and 50% tapering. In our data reduction, a taper function of 20% was found to reduce leakage quite well. This amount of "windowing" reduced the artificially induced frequency components sufficiently so they were in the real noise level of the system.

The Fourier transform is done using a fast Fourier transform algorithm. Since the data is discrete we can replace the usual Fourier integral with a discrete sum. It will be sufficient here to say that the method used is the Sandy-Tukey decimation-in-frequency FFT algorithm with a quarter-period negative sine table for generating the necessary complex exponentials. The sine wave is sampled densely enough to perform a discrete Fourier transform on a real data array of any power of 2 between 16 and 1024. In our case the upper limit of this program was always used and

Figure 18. The window function used to "taper" the data.
The 50% function is a true "Hanning" function.(43)

represented the limit of the system based on the scan distance of the mirrors.

The next step in the data reduction process was to do a rectangular to polar coordinate transformation to display the data in a more useful fashion. This, again was done by the ROM pack using a simple access command. The resulting data arrays represented the amplitude of the data and the phase of the data. For our purposes only the amplitude information was of interest. The rest of the program written was devoted to storing this information and plotting it out in a useful calibrated fashion. A copy of the complete program listing is given in Appendix A.

The data acquisition and reduction system were now complete and we proceeded once again with the problem of measuring the wavelength of the FIR output. The lock-in amplifier time constant was set at 3 sec. to duplicate the time between data points. This has been found to be the best method for digital interferometric work using a continuously scanning interferometer (p.49 of Ref. 42). The results of these scans will be discussed in detail in the next section, but it will be sufficient to say that they were quite impressive in their ease of generation and in the consistent agreement with the calculated energy levels of the excited potassium atoms.

During the process of taking these spectra it became necessary to know the true response curve of our detector. To check this a very simple experiment was done. A hot wire source with a black body temperature of about 2000⁰C was setup. It was

assumed that in the region of interest it would be a good approximation of a flat source of radiation. This was then chopped at a 10 Hz rate and put through a .5 meter aluminum tube whose output was focused into the interferometer. The output of a scan is shown in Figure (19). At first it appeared that we had excessive noise in our system so it was run again with identical results. After some reflection we decided it must be water vapor so the absorption peaks were compared to the well documented water vapor spectrum in this region. All of the absorption found could be identified as water vapor lines and none of the expected features were missing. From these results it was clear that the whole interferometer had to be enclosed in a water free environment. This was done by placing the device in a polyethylene glove bag and using a small positive flow of argon gas as well as bottles of desiccant. The experiment to verify the absence of water vapor features was not completed. The response of the Ga:Ge bolometer and filter system has been approximated by the curve drawn over the water vapor spectrum in Figure (19).

The next major step in the experimental process was to generate the FIR in an environment which would allow Stark tuning. As mentioned earlier the first design for a heat pipe with Stark plates was unsuccessful in producing FIR output. The second design, described in detail earlier, was now ready to be tested. Before loading the device, it was tested for field breakdown with only argon gas present. The breakdown was observed through the side window while a d.c. voltage was increased. This was done at

Figure 19. The bolometer response to a broadband source (hot wire) showing the water spectrum due to atmospheric water.

pressures ranging from .1 Torr to 10 Torr and it became clear that the minimum of the Paschen curve for this system was at about 1 Torr. This meant that the breakdown would occur most easily (with the smallest applied voltage) at 1 Torr. This was quite unfortunate because all work to date had indicated that 1 Torr was a very convenient pressure and produced very good quality FIR emissions. It was, however, found that the breakdown could be controlled quite well at a pressure of .3 Torr. Even here we could only obtain a c.w. field of 800 V/cm. This was not going to be enough to shift the levels so their relative difference would be measurable. At this point it was suggested that a pulsed field would be ideal in this application. If the stimulated emission occurs in a very short time after the initial exciting pulse, then we need only have the field on a short time to shift the levels of interest. Because the rate of arc propagation in gases at these pressures is about 1μ sec/cm, we could reduce or eliminate the problem by keeping the field on for 1μ sec or less.(44) To do this, a pulse generator was constructed which could deliver pulses of up to 2500 V with widths variable between 900 nsec and 3μ sec. This design is shown in Figure (20). It was found that a pressure of .3 Torr was necessary to be able to hold off the 2000 volts to be used. The field could be held off with no sign of arcing until the pulse width was increased to 2μ sec at which time the onset of ionization became apparent in the heat pipe. The pulser was triggered by the laser trigger and the pulse turn on and turn off were separately variable so we could observe the laser pulse with

Figure 20. The schematic of the high voltage pulse generator.

a fast detector (Molelectron P305 pyroelectric) and position the electric field pulse to start 100 nsec before the laser light pulse arrived.

The heat pipe was then loaded with 3 grams of potassium metal and a vacuum pumped on it for 24 hours to get rid of all residual solvents. The pipe was backfilled with 1 Torr of argon and the pipe heated to 100°C to release any trapped gases in the metal sticks. A vacuum was again pumped on the system and the rate of rise of the pressure in the system was checked. This procedure was repeated until the system held a vacuum ($<1\mu$ of Hg) well. The system was then filled to the desired working pressure with argon and the heaters were turned up to the appropriate temperature. As mentioned earlier, this heat pipe was particularly simple to align optically because of the long window in one side. The visible light from the laser was allowed to enter the heat pipe which was aligned to let the light exit through the Z-cut quartz window. The Michelson interferometer was aligned with this output and the detector put into place.

The pump laser was already tuned to the correct wavelength from the last set of experiments with the old heat pipe, so we just had to place the visible-blocking filter in and turn it on to test the new heat pipe. Because of the long window in the side, the fluorescent output of the device could be observed quite easily. The fluorescence was clearly visible with no laser tuning necessary and the pump parameters such as doubling crystal angle and laser cavity adjustment could be made while monitoring the

visible fluorescence to find the optimum conditions. The FIR was immediately found and the heat pipe was rotated to allow the FIR to exit through the polyethylene window which is better suited to this wavelength range. It became obvious at this point, however, that we would have only limited working time available on this device due to the formation of condensed potassium metal droplets on both the top and bottom screens at the interface of the cold and hot regions in the pipe.

The detection system was set up and spectra were taken of the FIR with the field on at a level of 2000 volts and with the field off. Three spectra of each were taken in an alternating fashion before the beam path of the laser was completely blocked by the growth of the droplets. The data was reduced and the three spectra with the field were averaged as were the three without the field. The results will be discussed in the next section. The complete system is shown in schematic form in Figure (21).

With this phase of the experimental work done, the only remaining experiments were those to clarify the deexcitation paths from the excited states. All transitions in the FIR region could be observed by the Michelson system, but we had no time resolution due to the very long (1 msec) time constants associated with the FIR detectors. The only detectors available with the speed necessary to resolve the times expected were in the visible region of the spectrum. This was not a problem because as mentioned earlier we would use the visible fluorescence to tune up the system.

SCHEMATIC OF THE FIR EXPERIMENTAL APPARATUS

Figure 21. A schematic diagram of the total FIR experimental apparatus.

To accomplish this part of the project we once again set up the standard heat pipe oven and set the operating parameters at 1 Torr and 330°C. The u.v. pump light was collimated and the visible pump laser light was filtered out by the Corning 7-54 filter. A sapphire output window was used on the heat pipe to let all visible and u.v. light exit. The Spex 1672 monochromator was used with micrometer adjustable slits to provide resolution to 1 Å for .05 mm slit width. The output of the monochromator was monitored by a cooled RCA 31034 GaAs photomultiplier tube. This gives relatively flat response from 300 nm to 800 nm. The monochromator was aligned along the axis of the heat pipe. The u.v. pump laser light was filtered out by two techniques. First a mirror coated for >99% reflectivity at 300 nm $\pm$ 10 nm was inserted at a 45° angle to deflect most of the u.v.. The rest was absorbed by the 10 cm focal length lense used to focus the visible fluorescence into the monochromator. The visible transmittance of the mirror is shown in Figure (22). The lense was standard glass and transmitted none of the u.v., but did transmit well above 350 nm. The output of the photomultiplier tube was put into the boxcar integrater and the narrow window and variable delay features of the boxcar were used to temporally select only those components of the spectrum that were shortlived ($t < 50$ nsec). The resulting spectra will be discussed in the next chapter. Scans were made over the region of 400 nm to 600 nm and from 400 nm to 800 nm. Once again the volume of data being obtained suggested the use of the computer to store and manipulate this data so it could be

Figure 22. The absorption spectrum of the U.V. highly reflecting mirror $R > 99\%$ at 300 nm.

evaluated effectively. A program was written to take this data and store it on magnetic tape so it could later be compared to other scans. The program is listed in Appendix B. This allowed us to run scans with the heat pipe on and then subtract the background with the heat pipe turned off. We effectively were doing a two beam experiment with a single beam system. From these experiments we had good spectral information on the system, but still no temporal information. To achieve good temporal resolution we took the output of the photomultiplier tube and terminated it into a $1k\Omega$ load. The time response of the system was then less than 10 nsec which allowed viewing all visible fluorescence in a pump pulse limited situation. Because of the low responsivity of the photomultiplier with a $1k\Omega$ load, we had to reduce the resolution of the monochromator to increase its throughput. The results of these measurements will be discussed in the next section.

The last experiments done on the system were to quantify the energy absorbed in the vapor as a function of pressure. A scientech Model 362 power meter was used to calibrate a Molectron Model J305 joulemeter which was used to make all energy measurements. The J305 was found to have a responsivity of 3259 volts/Joule when terminated into a $1M\Omega$ load. The data listed in Table (7) shows the results of these measurements at various pressures. In Figure (23), we have the data displayed and a least squares fit to these data using the function

TABLE 7

Pressure (Torr)	Intensity I/I_0
0.295	0.86
0.32	0.83
0.51	0.82
0.71	0.82
1.00	0.81
3.00	0.61
5.00	0.53

Figure 23. The absorption of the pump laser for the 4s-13p transition with an exponential curve fitted.

$$Y = A \exp(BX)$$

The R-square error is 0.980, the residual error is 0.000, and the maximum residual is 0.029.

It is difficult to determine from these results what the limit on the system is. Because of the mismatch in linewidths between the laser pump and the potassium absorption, we can say little more about the system. Needless to say, we could improve the energy coupling efficiency by using a narrower pump laser, but as mentioned earlier, this generally leads to lower energy output of the pump laser and is, not necessarily advantageous.

IV. RESULTS

This chapter will deal with the results of the experiments discussed in the last chapter. These results will be presented in two major sections: results from visible spectroscopy and results from FIR spectroscopy. Included in the visible results will be the optogalvanic spectroscopy done using a potassium hollow-cathode lamp.

We begin this chapter by discussing the results of some spectroscopy done on the potassium vapor when it was excited with a high average power continuous wave, tunable dye laser. The pumping system consisted of Spectra-Physics krypton ion laser operating at 3 watts on the 647 nm line pumping a Spectra-Physics dye laser using Oxizine 725 as the dye. This system was capable of producing 1 watt of tunable laser power in the region of the 767 nm 4s-4p fundamental line in potassium. The fluorescence in the vapor was observed by a front surface, hole coupled mirror set at the input end of the heat pipe at a 45° angle. This allowed us to view the input boundary layer of the potassium as it was being excited. Visual inspection revealed components of fluorescence in the green-yellow portion of the spectrum. These were studied more carefully and the spectra taken are shown in Figure (24). Figure (24a) was taken with the dye laser tuned on the 4s-4p absorption. We see five distinct features here: four sharp lines and one broad one. The four sharp lines can be identified as atomic transitions. The fifth broad line can be identified as a molecular transition. Note that the fluorescence occurs at higher

Figure 24a. The fluorescence from the K vapor when pumped at 767 nm, a) on the 4s-4p transition, b) .04 nm off of the 4s-rp transition.

energy than the pump. In Figure (24b) the dye laser has been tuned off of the atomic absorption line by approximately .04 nm. We see that all of the atomic features have disappeared and the molecular feature has increased in magnitude. Both scans were done under identical conditions and the vertical scales are the same.

These features can be explained if we look at the two sets of features, the atomic and the molecular, separately. Similar atomic features have been observed previously (45), (46) in sodium under quite similar conditions. In (46) it was argued that the production of high-lying states can result from energy-pooling collisions where formation of states higher than $2h\nu$ requires conversion of kinetic energy. In our work the atomic features can be identified as $5d \rightarrow 4p$ and $7s \rightarrow 4p$ fluorescent transitions as shown in Figure (2). The energy of these upper states is 30185 cm^{-1} and 30274 cm^{-1} respectively which is greater (by more than 4000 cm^{-1}) than twice the laser pump energy of 13023 cm^{-1} . This means that 4000 cm^{-1} (.5 eV) of energy must come from collisions. In (45) similar energies were required to achieve the observed Na transitions. One difference between our experimental conditions and those of both (45) and (46) is the significantly higher pressures used. We worked at an atomic density of about $10^{16} \text{ atoms/cm}^3$ while both (45) and (46) worked at pressures of about $10^{13} \text{ atoms/cm}^3$. We would, therefore, expect more collisions and also collisions of higher energy transfer due to the higher temperatures involved (620°K v.s. 500°K .) We would, however, only

Figure 24b. The fluorescence from the K vapor when pumped at 767 nm, a) on the 4s-4p transition, b) .04 nm off of the 4s-4p transition.

expect .025% of the atoms to have kinetic energy in excess of 4000 cm^{-1} .

The molecular feature in Figure (24) appears structureless in the limit of our experimental resolution (2 Å). This feature was observable both with the laser tuned onto the atom resonance line and with the laser tuned off by .04 nm. The intensity decreased as the pump laser was tuned on the line which could be the effect of less laser energy being available for the molecular species to absorb. This molecular feature was first observed by Rebbeck and Vaughn (47) in a low power discharge operating at about $10^{16} \text{ atoms/cm}^3$. The main band in their work was centered at 572.5 nm and was found to be structureless even at high dispersion. They reported a linewidth (full width at half-maximum) of 3 nm and a symmetrical line shape. We can see from Figure (24) that this is a very good description of the feature we have observed. The proposed mechanism for this feature is a Σ_u^+ upper state decaying to a $^3\Sigma_u$ dissociative lower state which results in two ground state atoms. The upper K_2 level must be excited by the energy-pooling in the collision between two K_2 molecules excited to the $^1\Sigma_u^+$ state by the laser. A tentative diagram for this process is shown in Figure (25). This upper excited state could also be created by the collision of an excited molecule with an atom or through multiphoton absorption. No attempt was made here to determine which of these processes were dominant.

Figure 25. The diagram of the molecular potassium system showing the proposed dissociative ground state of the broad molecular feature.

The molecular features are very interesting because if the lower state is a dissociating molecular state, we could pump the upper state more efficiently and, as in the eximer lasers, create a population inversion which could be used as a laser itself. More work, however, will need to be done on the excitation mechanism before one could estimate the potential efficiency of such a device.

Some preliminary studies were done on the effect of pressure and pump intensity on the atomic transitions, but were discontinued because of priorities of other work.

The next experimental results discussed are those from the brief experiments with the opto-galvanic effect. This effect has been observed experimentally as long ago as 1925 (48), but was first applied as we used it in Reference (49). The method used was outlined in the last chapter. We see in Figure (15) the results of a scan of the dye laser which excited $4p-7s$ and $4p-5d$ transitions in the potassium vapor of the hollow-cathode tube. We clearly see the components of these transitions. This technique appears ideally suited for atomic absorption spectroscopy as described in (50) and to lock a c.w. dye laser (51). This last possibility holds great promise for locking a laser to the desired u.v. absorption line. This would create a very stable, perfectly tuned pump system for work involving atomic excitation. This technique should be easily extendable to use with pulsed laser system as well as the demonstrated c.w. systems. This technique

was intended to be used to tune our laser to the correct transition but was never used for this purpose.

The last visible spectroscopy results are crucial to the understanding of deexcitation dynamics in the potassium vapor. These spectra give us an idea of the path of cascading as the upper excited state is depopulated by stimulated emission. The experiment was set up as described in the last chapter. The first spectra were taken without the aid of the boxcar integrator. The output of the photomultiplier tube was put into a picoammeter and used the internal time constant of the picoammeter to average the pulses of the pump laser. The monochrometer was scanned over a 200 nm range from 400 nm to 600 nm. The recorded output is shown in Figure (26). We see several sharp features as well as a few broader lines and a general offset of the whole spectra. We would expect to see many if not all of the visible transitions in the potassium because as can be seen from Figure (2), all of the transitions in the 400 nm to 600 nm region are the result of ns-4p or nd-4p transitions. Since the 13p level is being populated, we should see a series of lines beginning at about 475 nm and continuing up to 584 nm. These features can be seen in Figure (26), but the issue is quite confused due to other spectral features. For instance the broad featureless molecular line discussed earlier at 572.5 nm can also be seen. We also see a very broad band centered in the 440 nm region where there is a well known molecular potassium band (52). Since molecular features should have characteristic lifetimes much longer than atomic features that are

Figure 26. A scan of the fluorescence from the potassium vapor from 400-600 nm pumped to the 13p level. No temporal filtering has been used.

collisionally controlled, we expected to be able to filter out these features in the time domain by using the boxcar integrater with the sampling gate delayed very little from the laser exciting pulse. Figure (27a) shows a similar scan to Figure (26), but now using the time discrimination of the boxcar. A dramatic drop in the background interference and the almost total absence of the molecular features such as the 572.5 line is shown. Figure (27b) shows the same scan with the heaters on the heat pipe turned off. The large line at 585 nm is residual fundamental dye laser light getting through the Corning filter and provides an excellent wavelength marker for calibration of the spectra. In the scan: Figure (27a), this line is the farthest to the long wavelength end of the scan. We begin to lose the ns-4p lines as we move to higher n states. This is an indication that we have no population of these states and supports our calculations that the 11d level will be populated in favor of the 13s level in the initial super-fluorescent transition out of the upper excited state. As we move to lower n values the ns-4p and nd-4p transitions seem to become comparable in magnitude, with the 7s-4p actually larger than the 5d-4p transitions. We also see a hint of some transitions originating in a state higher than 13s. As we will see later, we have some excitation, presumably from collisions, to the 14s state and would expect a comparable population of the 12d level. The separation between the 4p $1/2$ and 4p $3/2$ states naturally remains the same for all transitions, but since we are displaying the data in wavelength and not energy, the splitting appears to decrease as

Figure 27a. The same scan as Figure 26, but with the addition of temporal filtering using a boxcar integrater, a) hot K vapor, b) cold (for background signal).

Figure 27b. The same scan as Figure 26, but with the addition of temporal filtering using a boxcar integrator, a) hot K vapor, b) cold (for background signal).

we approach the high energy or short wavelength end of the spectrum. Figures (28a) and (28b) show a scan for 600 nm to 800 nm. Once again the second of these scans was done with a cold heat pipe for reference. The large feature at 674 nm is the second order of the 337 nm N_2 laser off of the monochrometer grating. This is due to scatter in the dye laser cell. Once again this provides a convenient wavelength marker. We see here a large featureless line and the two sharp atomic lines due to the 6s-4p and 4d-4p potassium transitions. The extremely wide line is in both of the scans and is due to fluorescence of the Corning 7-54 filter. The next set of figures, Figures (29a) and (29b) show a complete scan at a resolution of .5nm from 400 nm to 700 nm with the addition of a filter to remove the N_2 laser scatter. We can see clearly the pattern of fluorescence from the excited states. One point should be noted. If Figure (29b) is carefully compared to Figure (29a) in the region of 695 nm, we see that some of the fluorescent energy of the filter is absorbed by the atomic system or the molecular system associated with this atomic line. This indicates the very large cross section for these transitions.

It is now clear that virtually all levels below the pumped level (and possibly even the ones directly above it) are involved in the depopulation process. The information needed is the rate of the depopulation so we can predict the rate of depopulation of the upper lasing level. To achieve this we proceeded as was described in the last chapter. The output of the photomultiplier tube was viewed directly on the oscilloscope (Tektronix Model

Figure 28a. The same as 27, but from 600-800 nm. The line at 674 nm is the second grating order of scattered 337 nm N_2 laser light, a) hot K vapor, b) cold.

Figure 28b. The same as 27, but from 600-800 nm. The line at 674 nm is the second grating order of scattered 337 nm N_2 laser light, a) hot K vapor, b) cold.

Figure 29a. The same as 27, but from 400-300 nm. The broad background about 700 nm is due to a fluorescing filter. Note the partial absorption of the broad fluorescence about the sharp peaks near 690 nm, a) hot K vapor, b) cold.

Figure 29b. The same as 27, but from 400-800 nm. The broad background about 700 nm is due to a fluorescing filter. Note the partial absorption of the broad fluorescence about the sharp peaks near 690 nm, a) hot K vapor, b) cold.

7844) and pictures were taken of the trace. These pictures are shown in Figures (30a-30j) for all of the wavelengths shown in Figure (27a). Because the monochromator slit widths had to be increased to let more light in to compensate for the lower response that is inherent in a fast measurement, the wavelength resolution was sacrificed and the individual transitions could not be identified in the ns-4p and (n-2)d-4p bunches. The wavelengths shown in Figures (30) are the monochromometer reading at the peak of the photomultiplier response. It is clear from Figures (30h) and (30j) which are pictures of the dye laser output, that the detection system has the capability to resolve the 10 nsec pulses. In Figures (30a) through (30g) we see that as we get to longer wavelengths or lower lying transitions, the pulse length becomes longer and the peak of the pulse is delayed longer. The depopulation rate in the higher lying transition indicates that they are being depopulated by superfluorescent transitions. This observation agrees quite well with those in reference (53). This work was done on the excited s ($n = 12$) and d ($n = 10$) states of rubidium with a system quite similar to ours. The main difference is the working densities of the vapor (10^{12} atoms/cm³ vs. 10^{16} atoms/cm³.) The results presented here are the first, time resolved, fluorescent results available for the excited states of potassium as far as is known.

The next main area of spectroscopic results for us to discuss are the results in the FIR region of the spectrum. By using the techniques discussed in the last chapter, we were able to develop

Figure 30. Photographs of the temporal character of the groups of lines shown in Figure 27a, a) 475.4 nm, b) 480.2 nm, c) 486.5 nm, d) 495.3 nm, e) 510.8 nm, f) 533.2 nm, g) 579.8 nm, h) 585.5 nm, i) same as (h), but on a 20 nsec/cm scale.

a spectroscopic method which could resolve shifts or splittings in the FIR of less than $.3 \text{ cm}^{-1}$ after several improvements. Here we will look at results using both of our FIR detectors and also at the FIR spectrum of water vapor.

As described in Chapter III, the output of the FIR detector was monitored by the lock-in amplifier whose output was sampled and digitized by a digital voltmeter and recorded by the computer. Figure (31) illustrates a typical interferometric output when the instrument was run at low resolution. We see here that we have a definite periodic signal which is being modulated by at least one other signal beating against it. If we had only one frequency, we would expect a pure sine wave whereas if we had a very broadband signal we would expect to see only one spike corresponding to the zero difference position of the two arms of the interferometer. In Figure (31) we see we have something between these two extremes, and therefore, expect several distinct features. We also see here that we have a large d.c. offset and a shifting of the average offset. These are due to less than perfect alignment and will show up as a large first Fourier component and low frequency noise respectively.

When the output of interferometer is processed by the computer to obtain the fast Fourier transform, we obtain a spectra such as that shown in Figure (32a). We see here several very sharp features as well as the predicted low frequency noise. The sharp features are more easily located in the expanded view of Figure (32b). These features can be identified by reference to the

Figure 31. Typical output for a low resolution scan of the Michelson interferometer.

Figure 32a. Fourier transform spectrum of the output of the heat pipe using the Ga:Ge bolometer, a) total spectrum, b) expanded view of (a).

Figure 32b. Fourier transform spectrum of the output of the heat pipe using the Ga-Ge bolometer, a) total spectrum, b) expanded view of (a).

expanded diagram of the upper excited states of potassium shown in Figure (3). Several identifications can be made in this spectrum, but there is still a problem of unresolved peaks in the region about 90 cm^{-1} . The resolution was increased by taking our data points at longer time intervals which allowed us to improve the resolution by a factor over three. This comes from the fact that we are scanning over more fringes and the resolution is directly proportional to that number. A scan at this higher resolution is shown in Figure (33). This spectra was taken with the Golay cell as the detector and the heat pipe pressure was 1.0 Torr. The time constant was 10 seconds and the total running time for the scan was 171 minutes. The peaks have been clearly resolved and the appropriate identifications can be made. The largest peak is measured to be 91.5 cm^{-1} and the calculated 13p-11d transition energy is 91.6 cm^{-1} . The two large peaks below the 91.5 cm^{-1} feature are measured to be 76.4 cm^{-1} and 84.2 cm^{-1} . The 84.2 cm^{-1} peak is identified as the 11d-12p transition predicted at 83.9 cm^{-1} . The 76.4 cm^{-1} peak, however, can only be identified as the 14s-13p transition calculated to be 76.1 cm^{-1} . This apparent anomaly in the spectrum can be understood when it is remembered that the thermal energy of the atoms is about 400 cm^{-1} so some collisional population of this state would be expected. The emission might appear as stimulated emission then, because the 13p state very rapidly deexcites and we are left with a population inversion in the 14s state. The curiosity is, therefore, not that we see emission from the 14s state, but that we do

Figure 33. High resolution scan of the heat pipe output using the golay cell.

not see any emission from the 12d state. Neither of the nearest neighbor transitions appear.

Two other features that can be identified are those at 114.1 cm^{-1} and 121.2 cm^{-1} . These correspond exactly to the 114.1 cm^{-1} and 121.2 cm^{-1} 10d-11p and 12p-10d transitions respectively. There are two significant absences from our spectra that should also be discussed. At no point do we see the cascading process include the upper excited ns states. We have the ability here to see any transition populating the 11s or higher states, but no significant signal at 79.2 cm^{-1} or 103.3 cm^{-1} is seen. The other absence noted here is the 146.7 cm^{-1} 11d-10f peak observed in the lower resolution scan. This can easily be accounted for by the fact that the crystal quartz window on the Golay detector has a broad band absorption which apparently absorbs this output peak. Scans done with the bolometer such as Figure (34) show this peak quite distinctly. The scan in Figure (34) was done at .3 Torr in the Stark cell heat pipe oven described in Chapter III. This is with the flat or "pancake" design. The dramatic improvement in signal to noise can be seen which is made more impressive when it is noted that the time constant for this scan was 3 seconds as opposed to the 10 second time constant of Figure (33). This allowed us to complete the scan in less than 52 minutes, one-third of the time for the Golay cell. The relative peak heights of these two scans have changed because of the change in pressure. This indicates a strong collisional effect on the 11d-12p and 14s-13p transitions, but very little

Figure 34. High resolution scan of the "pancake" heat pipe output with no field using the bolometer.

effect on the cascading further down in the system. This goes along with what one would expect. The lower the level, the smaller its effective cross section, and therefore, the less effect pressure changes should have on its population.

Using the same conditions as those used in Figure (34) the pulsed electric field was applied. After tuning the dye laser to the transition once again, because the 13p level had shifted about $.53 \text{ cm}^{-1}$, the interferometer was scanned. Three scans were taken alternately with scans of the heat pipe output with no field applied and the results of the scans with the field on were averaged as were those with the field off. The linewidths of the emission lines are seen to be similar, consisting of about four experimental points. The averaged results are shown in Figure (35a). A shift in the spectral lines can be seen quite clearly. The region from 38 cm^{-1} is shown on an expanded scale in Figure (35b) and illustrates both the excellent signal to noise of the measurements and the clear shift of $.3 \text{ cm}^{-1} \pm .15$ to the higher frequency when the field is applied.

The last spectroscopic results discussed are the measurements done to characterize the response of the Ga:Ge bolometer. This was accomplished as described in Chapter III. In Figure (19), we see the spectra taken with the hot wire source and in Figure (36) this is shown expanded. This scan has a resolution of 1 cm^{-1} . Because the work was done at atmospheric pressure, the water absorption lines which are very broad, and therefore, overlap over much of the spectral range covered. It is seen from this figure

Figure 35a. Output of the "pancake" heat pipe with the field on (~ 2000 V/cm) and off, a) from $0-100$ cm^{-1} , b) from $83-100$ cm^{-1} .

Figure 35b. Output of the "pancake" heat pipe with the field on (~ 2000 V/cm) and off, a) from 0-100 cm^{-1} , b) from 88-100 cm^{-1} .

Figure 36. Expanded view of Figure 19 showing more clearly the water vapor absorptions.

that the detector, whose response is smooth in this region, has its peak response in just the right region for our purposes and does not significantly distort the relative peak heights of the important lines whose energy lies between 75 cm^{-1} and 150 cm^{-1} .

V. DISCUSSION AND CONCLUSIONS

In this chapter the conclusions based upon the experimental results will be presented, problems concerning the experiment will be discussed, and recommendations for future work will be made.

By comparing the results of our experiments to the theory, an insight into the validity of the approximation methods used in the theory is obtained and the potential shortcomings of the experimental procedure are shown. The results of these comparisons will then point the way for improvements in our understanding of the problem.

We begin by comparing the results of the FIR spectroscopy to the predicted levels and shifts. The dominant transition in the experimental work is the $13p-11d$ transition which accounts for as much as $2/3$ of total FIR power out of the system as measured by the Michelson interferometer. As shown in Chapters II and IV the magnitude and direction of the shift agree well within the experimental limits outlined. This is a major result because it shows that substantial amounts of FIR radiation which is tunable over a potentially wide region of the spectrum can be generated by using several transitions and more than one atomic system. This can help extend the laboratory measurements of the rotational levels of molecules which would then help with the development of this branch of physics. Diatomics such as HCl and CO have spectral lines throughout this part of the spectrum.

We have, however, observed one level shift which does not agree with the predictions of Chapter II. In particular, we see

that the predicted shift for the $11d-12p$ transition is $-.66 \text{ cm}^{-1}$ yet our measurement shows a shift of $-.24 \pm .15 \text{ cm}^{-1}$. This discrepancy cannot be accounted for in the theory. The theoretical results for this shift seem reasonable. We would, therefore, hold the experimental results suspect. This, however, is not satisfying either because of the good agreement at other wavelengths and because of the extreme care used to insure the accuracy of the measurements. We, therefore, have a result which we are not comfortable with and which needs more experimental verification.

The unshifted FIR transition energies are well within the experimental errors of those energies predicted by the Rydberg formula. As mentioned earlier these predicted values will be much better as the value of $n + 1$ gets larger.

The next areas to look at are the results of the visible spectroscopy. It was seen that as we come down in n value of the upper fluorescent state, the time constant associated with that level increases. This also is quite consistent with the physical picture of the system we have described. The stimulated processes become decreasingly important as we move to lower transition energies and the system becomes much less organized in terms of the excited state location of the outer electron. The interesting, but not surprising absence of the resonant transition at 767 nm is explained quite simply as self-absorption by the potassium vapor. The decrease in energy emitted as we move to higher emission frequencies is explained by looking at the relative

Einstein B coefficients for the upper level transitions. Once again because of their higher frequency, the transitions to low lying states are less likely, and therefore, the energy of the upper states goes mainly into nearby states. It was also seen how upper states can attain a significant population by energy transfer processes such as energy-pooling collisions. These processes are not well understood and have had very little experimental attention. The similar molecular process that has been observed is also not well understood. Both of these areas show great promise for helping unravel the physics of the collisions in excited states of atoms and molecules.

The last spectroscopic area is that of the opto-galvanic effect. This method of spectroscopy, while not explored extensively here, shows great promise as a tool in experimental work. Its very complex physical nature would seem to be a basic problem with its use as a theoretically assailable system. We have realized very large signal to noise ratios with very good reproducibility from a simple system. These advantages have made this method of spectroscopy quite attractive and will continue to make it a useful adjunct to standard absorption spectroscopy for the location of transitions in many atomic systems.

The problems we have run into have been due to three main sources. The first is the difficulty in generating large amounts of narrow linewidth ultra-violet radiation in the 292 nm region of the spectrum. This has been recently overcome by the introduction of several commercial laser systems. These are the Nd:Yag pumped

dye laser systems which are capable of delivering 10^3 times the energy of our system in pulses of about the same temporal profile. Some of these systems offer excellent linewidth matches to our absorption linewidth and would prove nearly ideal for this application. Because of the linewidth match which could couple three times the energy in and the greater energy available, we would expect 3×10^3 greater output energy if there are enough atoms available in the vapor. This would require a longer device or higher pressure.

The second problem area has been the detectors used in the FIR. The detectors used in this work were long time constant thermal detectors which are inherently unsuited to the short pulse system we have developed. Currently there seem to be two alternative methods of detection which would be suitable for our use. These are a Schottky diode detector using a FIR laser as a local oscillator. An atomic beam of potassium pumped to the energy level desired and then ionized if radiation is absorbed. The latter method has been used extensively in the microwave and FIR regions and provides very low level detection with narrow linewidth resolution (54-56). A similar method has been used in (57) to detect infrared radiation by observing the visible output of the excited atomic system. These systems are very promising, but all have the disadvantages of size and complexity. No simple solution to the detection of the FIR at low levels seems to be at hand. There is one possibility for detection of the FIR which we have not yet mentioned. This is the use of pyroelectric

detectors. These devices are capable of subnanosecond response times, but suffer from very low responsivities. This, however, would be somewhat alleviated with the higher signal levels available from a Nd:Yag system.

The last problem source has been the heat pipe oven used to Stark shift the atomic levels. The design which proved successful had basic problems in the response of the structure to the thermal stresses applied at high temperature. A more complicated design based on the original Stark cell heat pipe appears to have the best chance of overcoming the problems. However, the design used, if built in a sturdier fashion could be modified to give the experimenter an excellent test bed for the study of metals at high temperatures under widely varying conditions with excellent optical access to the experimental region.

The areas of further study then, are the atomic and molecular energy pooling processes and the experimental apparatus development necessary to achieve high resolution, time resolved FIR spectra. These areas hold great promise for the experimentalist interested in atomic and molecular processes.

We have seen that the theory of highly excited atoms, while not complete, gives very good results in its predictions for the position and shifts of the energy levels. The improvements made are chiefly due to the availability of fast digital computers to do the calculations such as the numerical integration of the radial wave function. As the theoretical calculation techniques have improved, so have the experimental procedures and apparatus.

In particular, the development of the tunable laser has made possible the study of atoms in a single highly excited state. We have shown that we can in turn make other regions of the spectra available for study. The FIR source developed here will hopefully help bridge the gap in tunable sources that has previously existed.

REFERENCES

1. D. C. Laine, *Nature* 191, 795 (1961).
2. J. Gelbwachs, R. H. Pantell, H. E. Puthoff, and J. M. Yarborough, *Appl. Phys. Lett.*, 14, 258 (1969).
3. J. M. Yarborough, S. S. Sussman, H. E. Puthoff, R. H. Pantell, and B. C. Johnson, *Appl. Phys. Lett.*, 15, 102 (1969).
4. B. C. Johnson, H. E. Puthoff, J. Soohoo, and S. S. Sussman, *Appl. Phys. Lett.*, 18, 181 (1971).
5. M. A. Piestrup, R. N. Fleming, and R. H. Pantell, *Appl. Phys. Lett.*, 26, 418 (1975).
6. K. H. Yang, J. R. Morris, P. L. Richards, and Y. R. Shen, *Appl. Phys. Lett.*, 23, 669 (1973).
7. Y. R. Shen, *Appl. Phys. Lett.*, 23, 516 (1973).
8. T. I. Brown and P. A. Wolff, *Phys. Rev. Lett.*, 29, 362 (1972).
9. R. L. Aggarwal, B. Lax, H. R. Fetterman, P. E. Tannerwald and B. J. Clifton, *J. Appl. Phys.* 45, 3972 (1974).
10. K. P. Koo and P. C. Clasper, *Appl. Opt.*, 18, 1314 (1979).
11. D. H. Martin and K. Mizuno, *Adv. in Phys.* 25, 211, 1976.
12. C. Fabre and S. Haroche, *Opt. Comm.*, 15, 254 (1975).
13. M. E. Movesyan, N. N. Badalyan, and V. A. Irotyan, *ZLETF Pis'ma* 6, 631 (1967) (Russian).
14. P. P. Sorokin and J. R. Lankard, *Jour. Chem. Phys.*, 54, 2184 (1971).
15. D. R. Griscnowsky, J. R. Lankard, and P. P. Sorokin, *IEEE J. Quan. Elect.*, QE-13, 391 (1977).
16. A.M.F. Lau, W. K. Bischel, C. K. Rhodes, and R. M. Hill, *Appl. Phys. Lett.* 29, 245 (1976).
17. J. J. Wyne and P. P. Sorokin, *J. Phys. B*, 8, 137 (1975).
18. C. J. Latimer, *Contemp. Phys.* 20, 631 (1979).
19. M. Gross, P. Gray, C. Fabre, S. Haroche, and J. M. Raimond, *Phys. Rev. Lett.*, 43, 343 (1979).
20. D. R. Bates and A. Damgaard *Phil. Trans. R. Soc. London* 242, 101 (1949).

21. E. M. Anderson and V. A. Zilitis Optics and Spectroscopy (USSR) 16, 99-101 (1964).
22. M. I. Petroshev and I. V. Aborenkov, Vestn. Lenigr. Univ. No. 5 141 (1954).
23. D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) A 166, 450 (1933).
24. A. N. Nesmeyanov, Vapor Pressure of the Elements, N.Y. Academic 1963.
25. C. E. Theodosiov, Minima in the emission oscillator strengths of alkali Rydberg states, J. Phys. B. 13 (1980) L1-L5.
26. A. E. Siegman, An Introduction to Lasers and Masers, McGraw-Hill, 1971.
27. M. Sargent, Laser Physics, Reading, Mass., Addison Wesley, 1974.
28. M. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev., 20, (1979).
29. Myron Zimmerman, private communication
30. J. J. Wynne and P. P. Sorokin, J. Phys. B. 6 L37-L41 (1975).
31. A. M. F. Lau, W. K. Bischel, C. K. Rhodes, and R. M. Hill, Ap. Phys. Lett. 29 p 245 (1976).
32. C. R. Vidal and T. Cooper, J. Appl. Phys. 40 p. 3370 (1969).
33. G. M. Grover, T. P. Cotter, and G. F. Erickson, J. Appl. Phys. 35, 1990 (1964).
34. C. R. Vidal and F. B. Haller, Rev. of Sci. Ins. 42, 1779 (1971).
35. R. W. Boyd, J. G. Dodd, J. Krasinsky, and C. R. Stroud, Ja, Optics Letters, 5, 117 (1980).
36. L. L. Steinmetz, J. H. Richardson, B. W. Wallin, and W. A. Bookless, presented at a Topical meeting on Picosecond Phenomena, May 24-26, 1978, Hilton Head, S.C..
37. J. H. Richardson, L. L. Steinmetz, S. B. Deutscher, W. A. Bookless, and W. L. Schmelzinger, Z. Naturforsch 33a 1592 (1978).
38. J. H. Richardson, L. L. Steinmetz, S. B. Deutscher, W. A. Bookless, W. L. Schmelzinger, Anal. Biochem. 97, 17 (1979).
39. L. L. Steinmetz, W. A. Bookless, and J. H. Richardson, Apl. Optics, to be published (1980).

40. R. B. Green, R. A. Keller, G. G. Luther, P. K. Schenck, and J. C. Travis, *Appl. Phys. Lett.*, 29, 727 (1976).
41. R. Bystroff, LLNL, Gen. Chem. Div., Private Communication.
42. D. H. Martin, *Spectroscopic Techniques*, N. Y., Wiley, 1967.
43. Tektronics 4050 Series R08 Signal Processing ROM Pack. 2 Instructions.
44. L. L. Steinmetz, Private Communications
45. M. Allegrimi, G. Alzetta, A. Kopystoynska, L. Moi, and G. Orriols, *Opt. Comm.*, 22, 329 (1977).
46. G. H. Bearman and J. J. Leventhal, *Phys. Rev. Lett.*, 41, 1227 (1978).
47. M. M. Rebbeck and J. M. Vaughan, *J. Phys. B*, 4, 258 (1971).
48. Paul D. Foote and F. L. Mohler, *Phys. Rev.* 26, 195 (1925).
49. R. B. Green, R. A. Keller, G. G. Luther, P. K. Schenck, and J. C. Travis, *Appl. Phys. Lett.*, 29, 727 (1976).
50. W. B. Bridges, *J. Opt. Soc. Am.*, 68, 352 (1978).
51. R. B. Green, R. A. Keller, G. G. Luther, P. K. Schenck, and J. C. Travis, *IEEE J. Quant. Elec.*, QE-13, 63 (1977).
52. S. P. Sinha, *Proc. Phys. Soc. Ion.*, 5 436 (1947).
53. F. Gounard, M. Hugu, P. R. Fournier, and J. Berlande, *J. Phys. B.*, 12, 547 (1979).
54. T. F. Gallagher and W. E. Cooke, *Appl. Phys. Lett.*, 34, 369 (1979).
55. T. W. Ducas, W. P. Spencer, A. G. Vaidyanathan, W. H. Hamilton and D. Kleppner, *Appl. Phys. Lett.*, 35, 382 (1979).
56. M. Gross, P. Gay, C. Fabre, S. Haroche, and J. M. Rainard, *Phys. Rev. Lett.* 43, 343 (1979).
57. J. A. Gelbwachs, C. F. Klein, and J. E. Wessel, *IEEE Jour. Quant. Elec.* QE-16, 137 (1980).
58. L. Hrubesh, LLNL, Private Communication, 1980.
59. Atomic Energy-Level & Grotrian Diagrams Vol. 2, by Stanley Bashkin and John O. Stoner, Jr., North-Holland Pub. Co., Amsterdam, 1979.

APPENDICES

APPENDIX A

```

180 INIT
118 ON SRQ THEN 2280
120 PAGE
130 PRINT "THIS PROGRAM TAKES DATA FROM THE HP 3455A DIGITAL"
140 PRINT "VOLT METER AND DOES A FAST FOURIER TRANSFORM ON"
150 PRINT "THE DATA. TO USE THIS PROGRAM WITHOUT PROBLEM YOU"
160 PRINT "WILL NEED THE 'SIGNAL PROCESSING NO.1& NO. 2'"
170 PRINT "ROM'S IN THE 4051. IF THESE ARE NOT IN THE MACHINE"
180 PRINT "NOW THEN TURN OFF THE MACHINE, INSTALL THE ROM'S"
190 PRINT "AND BEGIN AGAIN."
200 PRINT "WHEN YOU HAVE READ THIS PRESS THE 'RETURN' BUTTON";
210 PRINT " TO BEGIN.";
220 INPUT A$
230 PAGE
240 INIT
250 ON SRQ THEN 2280
260 PRINT @16:"F1 R7 T2 M3 A1 H1"
270 PRINT "WHAT IS THE PULSE RATE OF THE TIME MARK GENERATOR?(SECS.) ";
280 INPUT K2
290 PRINT "HOW MANY SECONDS BETWEEN DATA POINTS? (THIS NUMBER MUST BE "
300 PRINT "AN INTEGER MULTIPLE OF THE TIME MARK GENERATOR RATE.)"
310 PRINT "T=";
320 INPUT T
330 PRINT "HOW MANY MINUTES DO YOU WANT TO RUN? M=";
340 INPUT K
350 G=60*K/T
360 S=T/K2
370 IF K2=T THEN 390
380 DIM A(S-1)
390 DIM B(G)
400 PRINT "PRESS THE RETURN BUTTON TO BEGIN TAKING DATA";
410 INPUT A$
420 PAGE
430 PRINT "NOW TAKING DATA. I'LL CALL YOU WHEN I NEED YOU!"

```

```

440 FOR I=1 TO C
450 INPUT @16:B(I)
460 IF K2=T THEN 480
470 INPUT @16:A
480 NEXT I
490 ON EOF (0) THEN 500
500 PRINT "GGGGGGG"
510 PRINT "DO YOU WANT THIS DATA ON TAPE? (Y OR N) ";
520 INPUT A$
530 IF A$="Y" THEN 580
540 IF A$="N" THEN 640
550 PRINT "Y OR N ONLY PLEASE! ";
560 INPUT A$
570 GO TO 530
580 PRINT "WHAT TAPE FILE WOULD YOU LIKE TO USE? F=";
590 INPUT F
600 PRINT "INSERT THE DATA TAPE AND PRESS THE 'RETURN' KEY.";
610 INPUT A$
620 FIND F
630 PRINT @33:B
640 PRINT "DO YOU WANT THE DATA DISPLAYED? (Y OR N) ";
650 INPUT A$
660 IF A$="Y" THEN 690
670 IF A$="N" THEN 760
680 GO TO 640
690 CALL "MIN",B,M1,I1
700 CALL "MAX",B,M2,I1
710 WINDOW 1,G,M1,M2
720 UIENPORT 20,100,20,80
730 PAGE
740 CALL "DISP",B
750 PRINT "1"
760 PRINT "DO YOU WANT TO DO A FAST FOURIER TRANSFORM ON?"
770 PRINT "THIS DATA? (Y OR N) ";
780 INPUT A$

```

```

790 IF A$="Y" THEN 930
800 IF A$="H" THEN 820
810 GO TO 760
820 REM
830 PRINT "DO YOU WANT TO RUN AGAIN?(Y OR N) ";
840 INPUT A$
850 IF A$="Y" THEN 230
860 IF A$="H" THEN 900
870 PRINT "Y OR N ONLY PLEASE! ";
880 INPUT A$
890 GO TO 850
900 PAGE
910 PRINT "GGGGGGG","FINISHED,GOODBYE"
920 END
930 PAGE
940 PRINT "DO YOU WANT TO USE THE TAPEP. FUNCTION ON THE DATA?(Y OR N) ";
950 INPUT A$
960 IF A$="H" THEN 1110
970 IF A$="Y" THEN 1010
980 PRINT "Y OR N ONLY PLEASE! ";
990 INPUT A$
1000 GO TO 960
1010 Y=1
1020 PRINT "HOW MUCH TAPEP WOULD YOU LIKE?(IN PERCENT FROM 10 TO 50) ";
1030 INPUT P1
1040 IF P1<10 OR P1>50 THEN 1060
1050 GO TO 1090
1060 PRINT "THIS NUMBER MUST BE BETWEEN 10 AND 50! ";
1070 INPUT P1
1080 GO TO 1040
1090 P1=P1/100
1100 GO TO 1120
1110 Y=0
1120 GO TO 1140
1130 PRINT "READ THE INSTRUCTION AND TRY AGAIN"

```

```

1140 PRINT "ENTER THE NUMBER OF POINTS"
1150 PRINT "THIS NUMBER MUST BE 128,256,512, OR 1024 ";
1160 INPUT R1
1170 R7=R1
1180 IF R1=128 OR R1=256 OR R1=512 OR R1=1024 THEN 1210
1190 GO TO 1130
1200 DELETE A
1210 DIM X(G)
1220 X=B
1230 DELETE B
1240 DIM B(R1)
1250 PRINT "HOW MANY MICRONS PER DATA POINT? ";
1260 INPUT S
1270 FOR I=1 TO R1
1280 B(I)=X(I)
1290 NEXT I
1300 DELETE X
1310 DIM L(R1/2+1),P(R1/2+1)
1320 CALL "MIN",B,N1,I2
1330 CALL "MAX",B,N2,I2
1340 FOR I=1 TO R1
1350 B(I)=B(I)-(N1+N2)/2
1360 NEXT I
1370 IF Y=1 THEN 1390
1380 IF Y=0 THEN 1420
1390 PAGE
1400 PRINT "GGG","TAPERING THE DATA, BE BACK IN A MINUTE"
1410 CALL "TAPER",B,P1
1420 PAGE
1430 PRINT "GGG","DOING THE FFT,IT WILL TAKE A FEW MINUTES"
1440 CALL "FFT",B
1450 CALL "POLAR",B,L,P
1460 CALL "MIN",L,M1,I1
1470 CALL "MAX",L,M2,I1
1480 Z=M2

```

```

1490 VIEWPORT 20,100,20,80
1500 R1=R1/2+1
1510 GOSUB 1940
1520 PRINT "1",R1;"HORIZONTAL POINTS ARE CURRENTLY DISPLAYED"
1530 PRINT "DO YOU WANT TO EXPAND THIS DISPLAY? (Y OR N) ";
1540 INPUT A$
1550 IF A$="N" THEN 1640
1560 IF A$="Y" THEN 1600
1570 PRINT "Y OR N ONLY PLEASE! ";
1580 INPUT A$
1590 GO TO 1550
1600 PRINT "HOW MANY POINTS WOULD YOU LIKE DISPLAYED? ";
1610 INPUT R4
1620 R1=R4
1630 GO TO 1510
1640 PRINT "2" VERTICAL POINTS ARE CURRENTLY DISPLAYED"
1650 PRINT "DO YOU WANT TO EXPAND THE DISPLAY?(Y OR N) ";
1660 INPUT C$
1670 IF C$="N" THEN 1760
1680 IF C$="Y" THEN 1720
1690 PRINT "Y OR N ONLY PLEASE! ";
1700 INPUT C$
1710 GO TO 1670
1720 PRINT "HOW MANY POINTS WOULD YOU LIKE DISPLAYED? ";
1730 INPUT D
1740 Z=D
1750 GO TO 1510
1760 PRINT " DO YOU WANT THIS ON THE PLOTTER? (Y OR N) ";
1770 INPUT B$
1780 IF B$="N" THEN 1840
1790 IF B$="Y" THEN 1830
1800 PRINT "Y OR N ONLY PLEASE! ";
1810 INPUT B$
1820 GO TO 1780
1830 GOSUB 2010

```

```

1840 PRINT "DO YOU WANT TO RUN AGAIN?(Y OR N) ";
1850 INPUT A$
1860 IF A$="N" THEN 1910
1870 IF A$="Y" THEN 230
1880 PRINT "Y OR N ONLY PLEASE! ";
1890 INPUT A$
1900 GO TO 1860
1910 GO TO 910
1920 END
1930 REM-- THIS BEGINS THE GS DISPLAY SECTION-----
1940 PAGE
1950 WINDOW 1,R1,M1,Z
1960 AXIS (R1-1)/10,(Z-M1)/10
1970 CALL "DISP",L
1980 PRINT "GGGGG"
1990 RETURN
2000 REM---THIS BEGINS THE PLOTTING SECTION-----
2010 PRINT "MAKE SURE THE PLOTTER IS TURNED ON AND THE 'LOAD'"
2020 PRINT "BUTTON IS IN THE 'OUT' POSITION. WHEN THIS IS DONE"
2030 PRINT "PRESS THE 'RETURN' BUTTON.";
2040 INPUT A$
2050 FOR I=1 TO R1
2060 DRAW @1:I,L(I)
2070 NEXT I
2080 AXIS @1:(R1-1)/10,(Z-M1)/10
2090 FOR I=0 TO 10
2100 PRINT @1,21:8*I+17,15
2110 R6=I*R1*19.4932*256/(R7*10)
2120 R6=R6/S
2130 R6=R6+0.05
2140 R6=INT(R6)
2150 PRINT @1:R6
2160 NEXT I
2170 FOR I=0 TO 10
2180 PRINT @1,21:10,6*I+19

```

```
2190 PRINT @1:I/10
2200 NEXT I
2210 PRINT @1,21:50,9
2220 PRINT @1:"FREQUENCY (1/CM)"
2230 PRINT @1,21:8,25
2240 PRINT @1,25:90
2250 PRINT @1:"INTENSITY (Arbitrary units)"
2260 PRINT @1,25:0
2270 RETURN
2280 POLL M,N;16
2290 RETURN
```

APPENDIX B

```

1 POLL H,H;16
100 INIT
110 ON SRQ THEN 2460
120 PAGE
130 PRINT "THIS PROGRAM TAKES DATA FROM THE HP 3455A DIGITAL"
140 PRINT "VOLT METER. TO USE THIS PROGRAM WITHOUT PROBLEM YOU"
150 PRINT "WILL NEED THE 'SIGNAL PROCESSING NO.1& NO. 2'"
160 PRINT "ROM'S IN THE 4051. IF THESE ARE NOT IN THE MACHINE"
170 PRINT "NOW THEN TURN OFF THE MACHINE,INSTALL THE ROM'S"
180 PRINT "AND BEGIN AGAIN."
190 PRINT "WHEN YOU HAVE READ THIS PRESS THE 'RETURN' BUTTON";
200 PRINT "TO BEGIN.";
210 INPUT A$
220 PAGE
230 INIT
240 DIM L$(40),M$(40)
250 X$="WAVELENGTH nM"
260 Y$="INTENSITY"
270 R3=0
280 K=0
290 H3=0
300 X1=10
310 A7=0
320 Y1=10
330 L$="ENTER SAMPLE NAME "
340 M$="ENTER DATE "
350 H$="ENTER STARTING WAVELENGTH (ie 400nm) "
360 O$="ENTER SCAN RATE (ie 40nm/MIN)"
370 S$="ENTER EXCITATION ENERGY IN MICROJOULES"
380 U$="ENTER NOTES "
390 B$="SAMPLE: "
400 C$="DATE: "
410 D$="STARTING WAVELENGTH: "
420 E$="SCAN RATE: "

```



```

430 I$="EXCITATION ENERGY: "
440 W$=" FILE # "
450 K$="NOTES: "
460 ON SRQ THEN 2460
470 PRINT "DO YOU WISH TO PLOT DATA FROM TAPE NOW ? (Y OR /RETURN/)"
480 PRINT ">";
490 INPUT A$
500 IF A$="Y" THEN 1320
510 PAGE
520 GOSUB 3680
530 PRINT @16:"F1 R7 T2 M3 A1 H0"
540 PRINT "HOW MANY SECONDS PER DATA POINT?GGGG ";
550 INPUT T
560 PRINT "WHAT IS THE TIME MARK GENERATOR SETTING?GGGG ";
570 INPUT K2
580 PRINT "HOW MANY POINTS WILL YOU BE USING? "; "GGGG"
590 INPUT G
600 PRINT "THIS PROGRAM REQUIRES ";G*T/60;" MIN.0";1/T;" /SEC TO TAKE ";
610 PRINT G;" POINTS"
620 REM INPUT K
630 G=G+1
640 R1=G
650 R2=1
660 PRINT "PRESS RETURN TO CONTINUE";
670 INPUT A$
680 DELETE B
690 PAGE
700 VIEWPORT 20,100,20,90
710 W1=-5
720 W2=1
730 WINDOW 1,G,W1,W2
740 AXIS (G-1)/10,(W2-W1)/10
750 DIM B(G)
760 HOME
770 PRINT "PRESS THE RETURN BUTTON TO BEGIN TAKING DATA";

```

```

780 INPUT A$
790 PRINT "I AM HOLDING"
800 PRINT "KK"
810 DELETE A
820 INPUT @16:A
830 IF A>0 THEN 790
840 PRINT
850 PRINT "NOW TAKING DATA. I'LL CALL YOU WHEN I NEED YOU!"
860 MOVE 0,0
870 FOR I=1 TO G
880 L=T
890 INPUT @16:B(I)
900 DRAW I,B(I)
910 IF K2=T THEN 960
920 INPUT @16:A
930 L=L-K2
940 IF K2=L THEN 960
950 GO TO 920
960 NEXT I
970 ON EOF (0) THEN 980
980 PRINT "GGGGGGG"
990 R1=G
1000 R2=1
1010 PRINT "HAS THIS DATA COME IN REVERSED?"
1020 PRINT "ie LONG WAVELLENGTH TO SHORT (Y OR 'RETURN') "
1030 INPUT A$
1040 IF A$="Y" THEN 1060
1050 GO TO 1130
1060 DELETE C
1070 DIM C(G)
1080 FOR I=1 TO G
1090 C(G-I+1)=B(I)
1100 NEXT I
1110 B=C
1120 DELETE C

```

```

1130 GO TO 1720
1140 PAGE
1150 IF A7=99 THEN 3530
1160 GO TO 2400
1170 PRINT "DO YOU WANT THIS DATA ON TAPE ? (N OR /RETURN/) ";
1180 INPUT A$
1190 IF A$="N" THEN 2200
1200 PRINT "GGGGGG INSERT YOUR DATA TAPEGGGGGG"
1210 PRINT "WHAT TAPE FILE WOULD YOU LIKE TO USE? F=";
1220 INPUT Z$
1230 IF Z$="" THEN 2200
1240 IF VAL(Z$)=1 THEN 1260
1250 GO TO 1200
1260 PRINT "THE PROGRAM IS IN FILE #1, CHOOSE AGAIN!"
1270 GO TO 1210
1280 FIND VAL(Z$)
1290 PRINT @33:X1,Y1,K2,?,K,G
1300 PRINT @33:B
1310 GO TO 2210
1320 PRINT "WHICH FILE TO BE INPUTTED ?";
1330 INPUT F
1340 PRINT "GGGGGG INSERT YOUR DATA TAPE GGGGGG THEN /RETURN/ ";
1350 INPUT A$
1360 FIND F
1370 GO TO 4090
1380 REM
1390 INPUT @33:X1,Y1,K2,T,K,G
1400 R1=G
1410 R2=I
1420 DELETE B
1430 DIM B(G)
1440 INPUT @33:B
1450 RETURN
1460 GOSUB 1480
1470 GO TO 1820

```

```

1480 REM *****$*****GS DISP
1490 PAGE
1500 CALL "MIN",B,M1,I1
1510 CALL "MAX",B,M2,I1
1520 VIEWPORT 14,100,10,65
1530 R3=R1-R2
1540 M3=M2-M1
1550 REM** TARGET
1560 WINDOW R2,R1,M1,M2
1570 AXIS R3/10,M3/10
1580 REM *****
1590 CALL "DISP",B
1600 PRINT "1"
1610 RETURN
1620 GO TO 1630
1630 GO TO 1720
1640 PRINT "DO YOU WANT TO RUN AGAIN ? (N OR /RETURN/) ";
1650 INPUT A$
1660 IF A$="N" THEN 1680
1670 GO TO 470
1680 PAGE
1690 PRINT "GGGGGGG","FINISHED,GOODBYE"
1700 END
1710 END
1720 REM*****EXPANSION SECTION
1730 GOSUB 1480
1740 PRINT "DO YOU WANT THIS ARRAY INVERTED? (Y OR /RETURN/) ";
1750 INPUT A$
1760 IF A$="Y" THEN 1780
1770 GO TO 1790
1780 B=B
1790 R5=VAL(D$)*16.6666666667/1000
1795 R5=R5*I
1800 M2=VAL(N$)+R5*(R2-1)
1810 M1=M2+R5*R3

```

```

1820 PRINT M2;" TO ";M1;" nN ARE DISPLAYED"
1830 PRINT "DO YOU WANT TO EXPAND THIS DATA ? (Y OR /RETURN/) ";
1840 INPUT A$
1850 IF A$="Y" THEN 1870
1860 GO TO 1970
1870 PRINT "WHAT IS THE NEW STARTING POINT ?";
1880 INPUT N2
1890 R2=(M2-VAL(M$))/R5
1900 R2=R2+1
1910 PRINT "WHAT IS THE NEW END POINT ?";
1920 INPUT M1
1930 R3=(M1-M2)/R5
1940 R1=R3+R2
1950 GOSUB 1480
1960 GO TO 1820
1970 PRINT M1;" TO ";M2;" VERTICAL POINTS ARE DISPLAYED"
1980 PRINT "DO YOU WANT TO OFFSET THIS ARRAY? (Y OR /RETURN') ";
1990 INPUT A$
2000 IF A$="Y" THEN 2020
2010 GO TO 2050
2020 PRINT "ENTER THE AMOUNT YOU WANT OFFSET.(1e -2) ";
2030 INPUT O$
2040 B=B+O$
2050 PRINT "DO YOU WANT TO EXPAND THE DISPLAY ?"
2060 PRINT "(Y OR H (HORIZONTAL EXPAND) OR /RETURN/) ";
2070 INPUT A$
2080 IF A$="H" THEN 1820
2090 IF A$="Y" THEN 2110
2100 GO TO 1170
2110 PRINT "WHAT IS THE NEW STARTING POINT ?";
2120 INPUT M3
2130 M1=M3
2140 PRINT "WHAT IS THE NEW END POINT ?"
2150 INPUT M4
2160 M2=M4

```

```

2170 PAGE
2180 GOSUB 1520
2190 GO TO 1970
2200 Z$="NONE"
2210 PRINT "DO YOU WANT THIS ON THE PLOTTER ? (N OR /RETURN/) ";
2220 INPUT A$
2230 IF A$="N" THEN 1640
2240 GO TO 2260
2250 REM***** TARGET ?
2260 REM---THIS BEGINS THE PLOTTING SECTION-----
2270 PRINT "MAKE SURE THE PLOTTER IS TURNED ON AND THE 'LOAD'"
2280 PRINT "BUTTON IS IN THE 'OUT' POSITION. WHEN THIS IS DONE"
2290 PRINT "PRESS THE 'RETURN' BUTTON.";
2300 INPUT A$
2310 GOSUB 4430
2320 IF A=8888 THEN 2270
2330 MOVE @1:0,B(R2)
2340 FOR I=R2 TO R1
2350 IF I>R1 OR I>G THEN 2380
2360 DRAW @1:I,B(I)
2370 NEXT I
2380 HOME @1:
2390 PAGE
2400 PRINT " DO YOU WANT OTHER NOTATIONS ON THE PLOTTER ?";
2410 PRINT " (N OR /RETURN/) ";
2420 INPUT A$
2430 IF A$="N" THEN 1640
2440 GO TO 3120
2450 RETURN
2460 POLL M,W;16
2470 RETURN
2480 REM*****PLOT
2490 AXIS @1:R3/X1,M3/Y1
2500 S2=2
2510 S3=3

```

```

2520 PRINT @1,17:S2,S3
2530 A=45
2540 PRINT @1,21:A,3
2550 PRINT @1:X#
2560 A=25
2570 PRINT @1,21:3,A
2580 PRINT @1,25:90
2590 PRINT @1:Y#
2600 PRINT @1,25:0
2610 S4=1
2620 S5=1.5
2630 PRINT @1,17:S4,S5
2640 X2=86/X1
2650 X3=VAL(N#)+R5*(R2-1)
2660 IMAGE 3D.1D
2670 FOR I=1 TO X1+1
2680 PRINT @1,21:(I-1)*X2+11,8
2690 R6=X3+(I-1)/X1*R5*R3
2700 PRINT @1: USING 2660:R6
2710 NEXT I
2720 Y2=55/Y1
2730 REM***** K IS THE VERTICAL SCALE NUMBER INCREMENT ***
2740 K=0.1
2750 FOR I=1 TO 11
2760 PRINT @1,21:10,(I-1)*5.5+9.5
2770 A=0.1*(I-1)
2780 PRINT @1: USING 2790:A
2790 IMAGE 1D.1D
2800 NEXT I
2810 HOME @1:
2820 PAGE
2830 PRINT "MOVE THE PEN TO THE PROPER PLACE FOR LABELS"
2840 PRINT "GGGGGGGGGGTHEN PRESS RETURN"
2850 PRINT "OR (N) NO LABELS";
2860 PRINT @1,21:60,60

```

```

2970 INPUT A$
2980 IF A$="H" THEN 4570
2990 S6=N3/30
2900 GIN @1:X4,Y4
2910 A=0
2920 A$=B$&L$
2930 A$=A$&H$
2940 A$=A$&Z$
2950 GOSUB 3080
2960 A$=C$&M$
2970 GOSUB 3080
2980 A$=D$&H$
2990 GOSUB 3080
3000 A$=E$&O$
3010 GOSUB 3080
3020 A$=I$&S$
3030 GOSUB 3080
3040 A$=K$&U$
3050 GOSUB 3080
3060 GO TO 4570
3070 HOME @1:
3080 MOVE @1:X4,Y4-S6*A
3090 PRINT @1:A$
3100 A=A+1
3110 RETURN
3120 GOSUB 3140
3130 GO TO 2480
3140 REM***** TARGET
3150 REM*****LABEL DISPLAY
3160 PAGE
3170 PRINT "THE CURRENT LABELS ARE:"
3180 PRINT "X AXIS ";X$
3190 PRINT "Y AXIS ";Y$
3200 IF R3=0 THEN 3220
3210 PRINT "X AXIS TIC MARKS EVERY ";R3/X1;" OF ";R3

```



```

3220 PRINT "THE LABELS ARE:"
3230 A$=B$&L$
3240 PRINT A$
3250 A$=C$&M$
3260 PRINT A$
3270 A$=D$&N$
3280 PRINT A$
3290 A$=E$&O$
3300 PRINT A$
3310 A$=I$&S$
3320 PRINT A$
3330 A$=K$&U$
3340 PRINT A$
3350 REM***** FORGOT THE NUMBERS
3360 PRINT "JJJDO YOU WANT TO CHANGE ANY LABELS ? (Y OR /RETURN/) ";
3370 INPUT A$
3380 IF A$="Y" THEN 3410
3390 GO TO 3400
3400 RETURN
3410 PAGE
3420 PRINT "IF YOU WANT TO MAKE A CHANGE ENTER THE CHANGE"
3430 PRINT "THEN /RETURN/ IF YOU DO NOT WISH TO CHANGE"
3440 PRINT "JUST /RETURN/"
3450 PRINT "X AXIS ";X$
3460 INPUT A$
3470 IF A$="" THEN 3490
3480 X$=A$
3490 PRINT "Y AXIS ";Y$
3500 INPUT A$
3510 IF A$="" THEN 3560
3520 Y$=A$
3530 PRINT "IF YOU DO NOT LIKE THE SPACING,"
3540 PRINT "THE DISPLAY MUST BE RE-SIZED,"
3550 PRINT "TO RESIZE: ENTER /R/ THEN /RETURN/."
3560 PRINT X1;" X AXIS TIC MARKS AT ";R3/X1;" SPACING ";

```

```

3570 A7=0
3580 INPUT A$
3590 IF A$="" THEN 3630
3600 IF A$="R" THEN 4550
3610 X1=VAL(A$)
3620 IF N3=0 THEN 3680
3630 PRINT Y1;" Y AXIS TIC MARKS AT "M3/Y1;" SPACING "I
3640 INPUT A$
3650 IF A$="" THEN 3680
3660 IF A$="R" THEN 4550
3670 Y1=VAL(A$)
3680 PRINT L$;
3690 INPUT A$
3700 IF A$="" THEN 3720
3710 L$=A$
3720 PRINT M$;
3730 INPUT A$
3740 IF A$="" THEN 3760
3750 M$=A$
3760 PRINT N$;
3770 INPUT A$
3780 IF A$="" THEN 3800
3790 N$=A$
3800 PRINT O$;
3810 INPUT A$
3820 IF A$="" THEN 3840
3830 O$=A$
3840 REM
3850 REM
3860 REM
3870 REM
3880 REM
3890 REM
3900 REM
3910 REM

```

```

3920 REM
3930 REM
3940 REM
3950 REM
3960 PRINT S$;
3970 INPUT A$
3980 IF A$="" THEN 4000
3990 S$=A$
4000 REM
4010 REM
4020 REM
4030 REM
4040 PRINT U$;
4050 INPUT A$
4060 IF A$="" THEN 4080
4070 U$=A$
4080 GO TO 3150
4090 PRINT "DO YOU INTEND TO DO MATH ON THE ARRAY IN MEMORY (Y OR N) ";
4100 INPUT A$
4110 IF A$="Y" THEN 4160
4120 IF A$="N" THEN 4140
4130 GO TO 4090
4140 GOSUB 1380
4150 GO TO 1460
4160 PAGE
4170 PRINT "GGGGGGGG***** WARNING *****"
4180 PRINT
4190 PRINT "THE LABELS MAY BE INVALIDGGGGGGGGGGGGGGGGGGGG"
4200 PRINT " "
4210 PRINT "1 SUBTRACT DATA ON TAPE FROM CURRENT DATAJ"
4220 PRINT "2 SUBTRACT CURRENT DATA FROM DATA ON TAPEJ"
4230 PRINT "3 OOPS, I SCREWED UPJ"
4240 PRINT "INPUT THE NUMBER OF YOUR OPERATION";
4250 INPUT A
4260 GO TO A OF 4270,4330,4390

```

```

4270 DELETE C
4280 DIM C(G)
4290 C=B
4300 GOSUB 1380
4310 B=C-B
4320 GO TO 1460
4330 DELETE C
4340 DIM C(G)
4350 C=B
4360 GOSUB 1380
4370 B=B-C
4380 GO TO 1460
4390 PRINT "0 9000"
4400 PAGE
4410 FOR I=1 TO 20
4420 PRINT "GGGGGGGG UNLOAD THE PRINTER GGGGGGGG"
4430 GIN @1:A,I
4440 MOVE @1:A+A/100,I+I/100
4450 GIN @1:A1,I1
4460 IF A=A1 AND I=I1 THEN 4490
4470 A=9999
4480 RETURN
4490 PAGE
4500 FOR I=1 TO 3
4510 PRINT "GGGGGGGGGGGGGGGGGG UNLOAD IT DUMMY GGGGGGGGGGGG"
4520 NEXT I
4530 A=8888
4540 RETURN
4550 A7=99
4560 GO TO 1130
4570 PRINT "DO YOU WANT TO RE-GRAPH ? (Y OR /RETURN/)"
4580 INPUT A$
4590 IF A$="Y" THEN 1720
4600 GO TO 1640

```