EXPERIMENTS ON THE APPLICATION OF LASER SELECTIVE EXCITATION SPECTROSCOPY TO THE DIAGNOSTICS OF A POTASSIUM PLASMA

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ABSTRACT

A thermally tuned Q-switched ruby laser of moderate power, with emission wavelength at 6939Å, has been used to selectively excite the \( \frac{4^2P_{3/2}}{6^2S_{1/2}} \) transition in the atoms of a pure potassium plasma of low degree of ionization produced in a hot cathode diode. A pulse of intensified spontaneous emission has been observed at 6939Å, 6911Å and at a number of other wavelengths, as a result of the laser selective excitation. The characteristics of the plasma intensified emission pulse have been observed to depend on the local conditions of the plasma, at the point of intersection of the laser beam and the solid angle of observation, estimated by a Langmuir probe. In particular, it has been observed that the signal decay time decreases as the free electron density increases.

The possibility of using this type of interaction for the purpose of diagnosing the plasma has been studied experimentally. The results obtained indicate that using an adequately tailored laser pulse it should be possible to measure the electron density and the excitation temperature in a potassium plasma in the range \( 10^{12} - 10^{14} \text{ cm}^{-3} \) and 2000-3500 °K respectively. The spatial resolution associated with the measurement should be of the order of a few cubic millimeters, with a temporal resolution of better than \( 10^{-7} \text{ sec} \). Furthermore, because of the low power required for excitation, the plasma perturbation effects associated with the measuring technique are likely to be negligible.
# TABLE OF CONTENTS

1. INTRODUCTION  
   1.1 Brief Review of Laser Resonant Interactions  
   1.2 Purpose of the Thesis  

2. THEORY  
   2.1 Basic Concept of the Diagnostic Technique  
   2.2 Theoretical Model  
   2.3 Simplified Theoretical Model  
      2.3.1 Initial Pumping Stage  
      2.3.2 Decay Period Immediately After Saturation  
      2.3.3 Steady State and Final Periods  
   2.4 Limitations of the Theoretical Model and Discussion of Some Implications of the Analysis of the Simplified Model  

3. EXPERIMENTAL CONSIDERATIONS  
   3.1 Purpose of the Experimental Programme  
   3.2 Experimental Requirements  
      3.2.1 Plasma Requirements  
      3.2.2 Laser Requirements  
      3.2.3 Requirements Concerning the Reference Diagnostic Method  

4. DESCRIPTION OF EQUIPMENT  
   4.1 The Tunable Ruby Laser  
      4.1.1 Laser Performance  
   4.2 The Potassium Plasma Source  
      4.2.1 Some Problems Associated with the Development of the Plasma Source  
      4.2.2 Performance of the Plasma Source  
   4.3 Auxiliary Equipment  

5. DESCRIPTION OF EXPERIMENTS AND OF EXPERIMENTAL RESULTS  
   5.1 Experimental Procedure  
   5.2 Exploratory Experiments  
   5.3 Experiments Designed to Evaluate the Role of the Laser Parameters  
   5.4 Experiments for Quantitative Correlation of the Plasma Enhanced Emission Signal with the Plasma Parameters
6. DISCUSSION OF EXPERIMENTAL RESULTS

6.1 General Interpretation of Experimental Results

6.1.1 Discussion of the Role of the Laser Power
6.1.2 Discussion of the Role of the Laser Wavelength Mismatch
6.1.3 Discussion of the Dependence of the Signal Decay Time on the Plasma Free Electron Density

6.2 Discussion of Non-Ideal Effects

6.2.1 Realistic Dynamics of the Laser Pulse Evolution
6.2.2 Plasma Ionization Enhancement
6.2.3 Free Electron Heating by Inverse Bremsstrahlung
6.2.4 Photoionization by Single and Multiple Photon Absorption
6.2.5 Laser Beam Self-Focussing Effects
6.2.6 Radiative Splitting and Shifts of the Resonant Energy Levels
6.2.7 Laser Pulse Distortion Due to Saturation Effects in the Plasma

6.3 Practical Requirements Regarding the Laser Performance

7. CONCLUSIONS

7.1 Conclusions
7.2 Potential Advantages of the Method of Laser Selective Excitation Spectroscopy for Plasma Diagnostics
7.3 Suggestions for Future Work

REFERENCES

FIGURES

APPENDIX A, B, C, D, E
NOTATION

$A_{ij}$ spontaneous transition probability for the radiative transition $i \rightarrow j$

$a_o$ Bohr radius

$B_{ij}$ Milne coefficient for the stimulated transition $i \rightarrow j$

$c$ speed of light

$D_j$ Global depopulation rate coefficient of level $j$

$E_j$ energy

$E$ electric field strength

$e$ electron charge

$F$ photon flux (photons cm$^{-2}$ sec$^{-1}$)

$f_{ij}$ oscillator strength associated with transition $i \rightarrow j$

$g_j$ statistical weight of level $j$

$G$ photomultiplier gain

$h$ Plank's constant

$I_\nu$ laser intensity (watts cm$^{-2}$ sec$^{-1}$ std$^{-1}$)

$I_o$ laser peak equivalent isotropic power density (watts cm$^{-2}$ std$^{-1}$) *

$I_i^*, I_e^*$ dimensionless ion and electron current density respectively

$J^*$ plasma enhanced emission coefficient

$J_i, J_e$ ion and electron current density, respectively

$K_{ij}$ de-excitation rate coefficient due to electron collisions, associated with transition $i \rightarrow j$

$k$ Boltzmann constant

$k_\nu$ plasma absorption coefficient, at frequency $\nu$

$L_\nu$ line shape factor, at frequency $\nu$

$M_i$ ion mass

$m_e$ electron mass

$N_j$ atomic population density of level $j$

$N_e$ free electron density

* Throughout the text for brevity we have used wcm$^{-2}$ when referring to $I_o$, where in fact, it should be understood that the value quoted is the equivalent isotropic power, in wcm$^{-2}$ steradian$^{-1}$
 pressure

 cross section

 rate coefficient for induced radiative transitions

 Langmuir probe radius

 enhanced signal amplitude

 electron temperature

 excitation temperature

 temperature

 excited plasma volume

 excitation rate coefficient due to electron collisions associated with transition i → j

 self focussing length

 fine structure constant

 dielectric constant of vacuum

 frequency

 laser frequency

 wavelength

 wavelength difference

 atomic weight

 solid angle of observation

 mean free path for elastic collision between species i-j

 photocathode quantum efficiency

 Debye length

 normalized potential of the Langmuir probe

 time constant

 characteristic risetime constant

 characteristic decay time constant
1. INTRODUCTION

1.1 Brief Review of Laser Resonant Interactions

The advent of the laser in the early sixties and the subsequent progress at a remarkable rate, provided the technology which has made possible the beginning of a whole new area of research: the study of the interaction of powerful radiation fields with matter.

The sheer power of solid state lasers has been used to heat, vaporize and ionize matter (1) (2), and an ultimate goal in this direction is the production of dense, hot plasmas of interest in thermonuclear fusion research.

High power lasers have also been used for diagnostics of hot plasmas by Thomson scattering (3) (4) and for studies of cross sections by Rayleigh scattering (5). Non-linear optical effects such as harmonic generation, multiphoton absorption, self-focusing and stimulated Raman, Rayleigh and Brillouin scattering, are also among the new areas of study made accessible by high power lasers.

Most of the research indicated above is based on non-resonant interactions, where the laser's primary role is to supply radiation of high power density, its emission wavelength being of secondary importance.

Recent improvements in the field of tunable lasers (7), have laid open a new and potentially vast area of endeavour, based upon resonant laser-matter interactions. In this instance, the laser output wavelength is tuned to coincide with a specific atomic or molecular transition within a liquid, gas or plasma, which thus can be excited selectively.

As a result of such selective excitation, the energy level populations of the pumped transition are momentarily redistributed; the upper level population is enhanced as a consequence of the absorption of laser radiation, and this leads to an enhancement of the spontaneous emission from this level, which can be readily monitored. Collisional energy transfer from the upper level population to neighbouring levels, produces an enhancement of the spontaneous emission of other transitions, which can also be monitored.

It is important to realize that the volume of matter under observation, determined by the intersection of the laser beam and the solid angle subtended by the optics, can be made as small as desired, compatible with the requirement of having enough photons for detection of a meaningful, relatively noise free signal. In some instances, as we shall discuss in more detail later, this feature is extremely valuable, since it allows for detailed spatial resolution.

Resonant laser-matter interactions are of interest to several fields. For atomic and molecular physics, it provides a means for studying atomic constants, such as radiative decay constants (8), collisional energy transfer cross-sections (9) (10), molecular structure (11), as well as for studies of photoionization and photodissociation (12) and spectroscopy in general. The latter subject has been reviewed recently in a book by Demtröder (13).

The possibility of selective excitation of fluorescence is also of interest in luminescence spectroscopy studies (14) based on the analysis of fluorescence and phosphorescence of materials. This has an extremely wide
range of applications, including chemistry and biochemistry. In the laser field itself, the study of saturable absorbers (15) is based on the concept of selective excitation of solutions of organic compounds.

Some results have been already obtained in the field of environmental sensing (16) where laser tuned radiation was used to probe the atmospheric sodium content. In the field of gasdynamics and plasmadynamics, some applications have been suggested (7) (8) (17) for measurement of flow velocities and ionic drift motion.

Several applications of laser selective excitation have been proposed in the field of plasma physics. This subject has been reviewed recently by Dewey (7). For continuity of presentation and because of their connection to the present work, we shall indicate here the major developments in this area, including some recent work not included in Dewey's review.

Early in 1967, Measures (8) discussed the possibility of measuring the electron density and temperature within a plasma, as well as some other possible applications, using what he termed "selective excitation spectroscopy". As illustrative examples, he analyzed the diagnostics of potassium and barium plasmas. Part of this work will be considered in some detail later on. The present thesis originated at that time, in an attempt to test experimentally the basic ideas and study the practical details of the diagnostic scheme as applied to potassium.

Independently, Dimock, Hinnov and Johnson (17) considered the diagnostics of a barium plasma using a tunable dye laser. This was based upon an extension of the resonance scattering work done by Hofmann (18) using a conventional light source. The present work of Dimock, et al shows the advantages of using for this purpose laser radiation to increase the number of photons produced per available scatterer. They reformulated the problem considered by Hofmann to account for the fact that the high intensity laser radiation practically equalizes the upper and lower level populations of the pumped transition, and obtained practically the same formula as Measures (8) for the total number of photons emitted as a result of laser excitation. They discussed measurement of the electron temperature, the ion temperature and the ion density. Mention was also made of the possibility of measuring the ion drift motion and the electron thermal energy transport. The latter measurement is based on the important concept of free electron heating, as a result of inelastic collisions with laser enhanced populations.

In 1969, Oettinger and Dewey (19) discussed the possibility of plasma ionization enhancement by means of selective laser excitation. Their idea was to pump a transition having an upper level near to the continuum from which the excited atoms could easily be ionized by free electron inelastic collisions. A key feature of this scheme is the fact that the upper level of the transition excited by the laser must be above a certain energy bottleneck, for which the rate of de-excitation reaches a minimum, as postulated by Byron, et al (20) and Hinnov and Hirschberg (21). Above this bottleneck, the energy level populations are in equilibrium with the free electron density. Oettinger and Dewey carried out calculations for atomic oxygen, nitrogen, carbon and cesium, and found that it was possible to enhance free electron densities of the order of $10^{14}$ to $10^{16}$ cm$^{-3}$ by a factor of two, with present day laser technology.

Ideally, in order to enhance ionization as much as possible, one would like to pump a low lying level, in order to elevate an appreciable population density; the best possible choice for this purpose is the ground level. Unfor-
Unfortunately, the technique suggested by these authors has the practical disadvantage (at least for the moment) that the bottleneck energy level is always high (3 ev for cesium, 9 ev for carbon) and thus, in order to pump from the ground level, ultraviolet lasers would be required, with proper tuning ability.

In this connection, Oettinger and Dewey point out that the population of the lower level of the pumped transition will be replenished continually, as a result of population transfer from lower levels and in particular, from the ground level. Under these conditions, the vast population reserve of the ground state becomes available in order to sustain an appreciable enhancement in the degree of ionization, that otherwise would be impossible on the basis of the population of the pumped level alone.

We would like to point out that although physically the argument is correct, for proper quantitative evaluation of the population replenishment process, one has to consider carefully the collisional energy exchange time constants involved in each case and how they compare to the duration of the laser pulse.

The assumption of instantaneous replenishment, and its implication that all the population of the ground state is made available to laser excitation by a rapid collisional transfer to the lower level of the pumped transition is generally incorrect unless one works with extremely high electron densities, or high pressure or very closely spaced lower energy levels.

In general, we can say that the rate at which the pumped level population is replenished is determined by the slowest rate in the population transfer chain from the ground level. This rate corresponds to excitation of the first excited level, in general.

Under these conditions, if the duration of the laser pulse is short, compared to the rate constant for replenishment of the lower level of the pumped transition, the ionization enhancement will be negligible, since the number of absorbers initially available will be very small. On the other hand, sustained pumping with a long duration laser pulse, may in fact lead to appreciable ionization enhancement provided the recombination time constant is larger than the population replenishment time constant. These conditions are implicitly assumed in the paper of Oettinger and Dewey, since a steady state is considered. This argument also shows that the maximum possible absorption rate for laser photons, under steady state conditions, is determined by the rate at which population is fed to the lower level of the pumped transition from the lower excited levels. This indicates that it is not correct to choose an arbitrary absorption rate in order to sustain a two-fold increase of the free electron density, as done by Oettinger and Dewey in their equation (11), without proper consideration of the actual dynamics of the steady state condition.

Measures (22) has recently discussed the ionization enhancement of a potassium plasma, resulting from laser excitation of the resonant transition. In his analysis, he does not invoke the concept of excitation above the bottleneck energy level. The increase of the degree of ionization in this case is a consequence of (i) the free electron temperature elevation which results from the enhancement of the population of the resonance level, (ii) the effectively reduced ionization energy of the atom due to the high population maintained in the resonance state by the laser radiation. He showed that for initial plasma conditions of $T_e = 2000^\circ K$, $N_e = 10^{12} \text{ cm}^{-3}$, it is possible to elevate the electron
temperature and density to values of about $T_e = 3600\,^0K$, $N_e = 2.5 \times 10^{14}\,cm^{-3}$ respectively after pumping for 2.5 $\mu$sec at a laser power of $160\,W\,cm^{-2}/cm$ length.

While most of the work concerning resonant laser-plasma interactions is theoretical and speculative in nature, only a few references are found to experimental work.

Carpenter (23) has attempted to test Oettinger and Dewey's ideas for plasma ionization enhancement using a tunable laser. He selectively excited the $5^2D_{5/2}-5^2F_{7/2}$ transition in neutral cesium ($\lambda = 8081\AA$) using a 3.3' diethylthiatricarbocyanine iodide dye laser pumped by a Q-switched ruby laser. The cesium plasma was produced in an annular thermionic converter and any ionization enhancement could have been detected by monitoring the increase of discharge current after laser excitation.

Carpenter's experiment failed to confirm any significant current changes and he suggested, as an explanation for negative results, the fact that the dye laser output energy per angstrom was less than required for producing appreciable excitation.

Carpenter established a laser energy requirement criterion, on the basis of the number of laser photons needed to be absorbed to double the free electron density, assuming that all selectively excited atoms would be ionized by free electron collisions. Thus, if the energy of a laser photon is $h\nu_L$ and the free electron density of $N_e$, he concludes, assuming that the upper level is a perfect sink, that an energy of the order of $h\nu_L N_e$ is needed per unit volume to double the plasma degree of ionization.

In establishing this energy criterion, Carpenter implies that the plasma will be able to absorb this amount of energy in a time of the order of 10 nsec, corresponding to the duration of his laser pulse. Since for his experimental conditions ($T_e \sim 3000\,^0K$, $N_e \sim 10^{15}\,cm^{-3}$, $N_{\text{ground}} \sim 10^{16}\,cm^{-3}$) the population of the lower level of the pumped transition is about $4 \times 10^{13}\,cm^{-3}$ (24), which is far less than the $10^{15}$ absorbers/cm$^3$ he needs for accomplishing his purpose, it must be concluded that he relies on nearly instantaneous replenishment of the lower level population from the lower lying levels, as discussed earlier in connection to Oettinger and Dewey's paper.

This assumption is not made explicitly by Carpenter, nor does he discuss the corresponding relaxation time for lower level population replenishment. In view of the extremely short laser pulses he used, and the low number of initial absorbers available, it is highly questionable whether in fact his plasma could absorb the amount of radiation he anticipated in order to double the free electron density, even if his laser had been powerful enough. As a crude and optimistic estimate, if we take the collisional excitation rate from ground level to level 6p to be $X_{12} = 4.8 \times 10^{-10}\,cm^3/sec$ (Gryzinsky) (27) for $T_e = 3000\,^0K$, and assume that all the population reaching level 6p is instantaneously excited into level 5d, which we assume behaves as a perfect sink (i.e., we neglect radiative and collisional de-excitation), we obtain for the rate of population replenishment:

$$\left( \frac{dN_{5d}}{dt} \right)_{\text{replenishment}} = N_1 N_e X_{12}$$
For $N \sim 10^{16}$ cm$^{-3}$, $N_e \sim 10^{15}$ cm$^{-3}$, we can calculate the number of replenished atoms $N$, in 10 nsec, to be:

$$N \sim 4.8 \times 10^{13} \text{ atoms cm}^{-3}$$

This figure, even considering the initial number of excited atoms present in levels $6P_{1/2,3/2}$ and $5P_{3/2,3/2}$ is too low to account for any significant ionization enhancement. Although this estimate is extremely crude and depends on a cross section reliable only within a factor of 2-3, it illustrates the fact that under transient conditions, the mechanism proposed by Oettinger and Dewey can be very inefficient.

Quite recently, Burrell and Kunze$^{(10)}$ published what we believe are the first positive experimental results in this area; they succeeded in observing enhanced emission from helium atoms in a plasma, after excitation with a tunable dye laser at 4471Å. They were able to observe the enhanced emission at several wavelengths, as a result of collisional transfer of the excitation and this enabled them to estimate the appropriate collisional rates.

An important aspect of this work is that it demonstrated the possibility of using a metastable level as the lower level of the pumped transition, in virtue of the ability of such a state to store a large population. Consequently it is possible to obtain an appreciable signal even at low temperature. This extends the application of the selective excitation techniques to new elements, having a large resonance energy gap.

Some preliminary results have also been recently published by Measures and this author$^{(25)}$ concerning the selective excitation of a potassium plasma using a tunable ruby laser. A full discussion of this experiment is given in this thesis.

1.2 Purpose of the Thesis

Measures$^{(8)}$ has suggested, on theoretical grounds, the possibility of using laser selective excitation techniques for localized plasma diagnostics. It was the purpose of the present thesis to attempt to evaluate this possibility experimentally, using a suitable practical example.

We have studied the case of a potassium plasma excited by a tunable, Q-switched ruby laser, with emission wavelength at 6939Å coincident with the $4^2P_{3/2}-6^2S_{1/2}$ transition of neutral potassium.

An extensive experimental programme has been established, in order to test the basic concept of the diagnostic technique and understand the basic physics of the resonant laser-plasma interaction under realistic conditions.
2. THEORY

2.1 Basic Concept of the Diagnostic Technique

When radiation from a tunable laser is used to excite selectively a transition in the heavy species of a plasma, absorption of the laser photons produces an enhancement of the upper level population. If the laser intensity is high enough, stimulated emission will balance absorption and the particular transition is saturated. Under these conditions, the upper and lower level populations become locked together, and will remain so as long as the laser intensity is above saturation threshold. The two population densities are in the same ratio as their respective level degeneracies (8).

If the transition rates induced by the laser dominate over collisional and radiative rates associated with the upper and lower level of the pumped transition, it is easy to show (see Appendix A) that at the instant of saturation:

\[
\frac{J^*_{\text{max}}}{J} = \frac{1 + \frac{N_L}{N_u}}{1 + \frac{g_L}{g_u}}
\]

where \( J^*_{\text{max}} \) represents the maximum value of the enhanced spontaneous emission coefficient of the plasma, at the wavelength of excitation (or at an alternative wavelength corresponding to a different decay mode of the upper level), \( J \) is the emission coefficient at the same wavelength prior to laser excitation, \( N_L, N_u \) are the respective lower and upper level equilibrium populations (before laser excitation) and \( g_L, g_u \) are the corresponding level degeneracies. The enhanced emission is maximum at the instant of saturation.

If \( N_L/N_u \gg 1 \) which is likely to be the case for widely spaced levels at moderate temperature, then we can rewrite (1) as:

\[
\frac{J^*_{\text{max}}}{J} = \frac{\hbar \nu A_L}{4\pi(1 + \frac{g_L}{g_u})} N_L
\]

where \( A_L \) is the radiative transition probability and \( \hbar \nu \) is the spontaneous emission photon energy. It is also clear from Eq. (1) that \( N_L/N_u \) must be as large as possible, in order to obtain an appreciable intensification of the spontaneous emission. The need for large intensification can be appreciated, if we consider that \( J^* \) originates from a relatively small volume, whereas \( J \) corresponds to the emission of the whole plasma volume within the solid angle of observation.

The criterion which ensures \( N_L/N_u \gg 1 \) can be written in terms of the excitation temperature \( T_{\text{ex}} \) and the laser frequency \( \nu_L \), viz

\[
\hbar \nu_L \gg k T_{\text{ex}}
\]
Equations (1) and (2) are useful in discussing the possibility of localized temperature measurements.

From (1) we see that by measuring the relative intensification of spontaneous emission from a given plasma volume, it is possible to obtain the ratio of the populations of the two levels of the excited transition, prior to laser pumping. If a condition of LTE prevails in the plasma before laser excitation, the ratio $N_2/N_u$ can be used to determine the electron temperature; under non-equilibrium conditions, only the excitation temperature can be calculated. In any case, the measurement is complicated by the necessity of knowing the excitation volumes contributing to $J_{\text{max}}^*$ and $J$.

A more elegant technique was suggested by Measures(8) based on excitation of two transitions sharing a common upper level. Under these conditions, if one measures the intensification of spontaneous emission which results in each case, the ratio of the two is given by Eq. (2) as:

$$\frac{J_{\text{max}}^*(v_{13})}{J_{\text{max}}^*(v_{23})} = \left[ \frac{v_{13} A_{13} (1 + g_2/g_3)}{v_{23} A_{23} (1 + g_1/g_3)} \right] \times \frac{N_1}{N_2} \quad (4)$$

where we have considered generic levels 1,2,3 level 3 being the common upper level. Provided the volumes excited by the two laser beams are identical, the ratio $N_1/N_2$ can be obtained from a relative measurement.

If only one frequency is used, an interesting possibility arises if the lower level of the pumped transition is the resonance level and the resonance radiation is strongly self absorbed. Under these circumstances the population of the resonance level is in collisional equilibrium with the ground level population and is characterized by the electron temperature.

Under such conditions, the ratio of the ground and resonance level populations is determined by a simple Boltzmann relation, and it is easy to show using Eq.(2) that:

$$J_{\text{max}}^* = \frac{g_2 A_{22} h \nu_{22}}{g_1 4\pi (1 + g_2/g_3)} \times N_1 \exp \left( - \frac{E_{21}}{kT_e} \right) \quad (5)$$

where again we have used generic levels 1,2,3 to indicate the ground, resonance and upper excited levels respectively, considering the laser to couple levels 2 and 3. $E_{21}$ is the energy of the resonance level.

Provided we know $N_1$, which for low degrees of ionization can be assumed equal to the neutral population, and provided we can calculate the volume excited by the laser, the electron temperature can in principle be readily determined. This technique has the inconvenience of requiring an absolute intensity measurement.

The preceding discussion is based on the assumption that the laser induced transition rates dominate over all other rates leading to excitation, or de-excitation, of the levels of the pumped transition. If the upper level is strongly collision coupled to a few neighbouring levels, such that we can assume
a nearly instantaneous population readjustment, and if the rates in and out of this set of levels are small compared to laser induced transition rates, the problem can still be reformulated and an equivalent set of equations (1) - (5) can be derived. Intermediate cases require detailed analysis of the dynamics of the transient perturbation.

As the transition excited by the laser saturates the absorption and the stimulated emission rates balance and the collisional and radiative decay from the upper excited level becomes important. As a result, the population of the set of laser-locked levels decays, with a time constant characteristic of the depopulating process for the upper level. In a low density plasma with a moderate degree of ionization, energy transfer processes occur mainly as a result of electron-atom inelastic collisions. If this is the dominating mechanism, it can be expected that high electron densities will produce a fast quenching of the upper level population, whereas at low electron densities, the quenching times will be longer and in the limit, the excited level will decay with a time constant equal to its radiative lifetime, provided atom-atom collisions are unimportant. Thus, we see that the enhanced spontaneous emission decay time can be related to the free electron density, in the conditions considered.

The preceding arguments indicate that under appropriate conditions, the plasma enhanced spontaneous emission which results from the laser excitation of a given atomic transition, provides important information about the conditions of the plasma. By measuring the enhanced emission signal peak amplitude it is possible to obtain information about the excitation or electron temperature, while the signal decay time can be related to the free electron density. This technique provides local measurements, since very small plasma volumes can be sampled. Furthermore, saturation of transitions can be achieved with quite moderate laser powers, which preclude serious perturbation of the conditions of the plasma.

2.2 Theoretical Model

A full simulation of the dynamics of selective laser excitation of a potassium plasma, under the conditions of interest for this experiment, has been performed by Fournier (26) as part of an independent thesis. Fournier has considered a twenty level model of the potassium atom, with electron collision rates calculated on the basis of the Gryzinski (27) cross-sections for forbidden transactions and Seaton's (28) cross-sections for allowed transitions. Zapesochny and Shimon's experimental excitation cross-section (29) was used for the resonance transition. Assuming a Collisional-Radiative model (30) for the plasma, which is considered optically thin except for all the resonance transitions, a system of twenty differential rate equations is set up and solved by the Runge-Kutta method (31) using a IBM 360/65 computer. Simultaneously, the temperature variation of the free electrons, as a result of the inelastic collisions with the laser excited atoms, can be calculated by proper consideration of the electron energy equation. Nevertheless in the preliminary analysis available at the time of writing the collisional rate coefficients were not readjusted for electron temperature changes. The implications of this aspect of the program will be discussed later.

The results obtained by Fournier, corresponding to the temporal variation of the enhanced spontaneous emission of the pumped transition are used in the interpretations of results of this experiment.
2.3 Simplified Theoretical Model

In an attempt to gain some physical understanding of the basic features of the laser excitation process, we have developed a simplified model for analysis of the problem. Although the model is based in a considerable simplification of the interaction it represents correctly the important initial period of excitation of the transition, up to the instant of saturation. Simple analytical formulas can be derived from this model that illustrate the role of the laser power and wavelength mismatch on the degree of enhancement of the upper level population. Also, insight is gained into the time correlation between the laser pulse and the enhanced emission pulse.

Figure 1 illustrates a realistic energy level diagram of neutral potassium (for clarity we only indicate the doublet nature of the \( ^4P \) term). The equivalent simplified energy level diagram adopted in our model is shown in Fig. 2. Here, we have retained the identity of only the relevant levels and grouped the rest in a manifold level system for simplicity of representation.

Because of the small energy gaps involved, levels \( ^4P_{3/2} \) and \( ^4P_{1/2} \), and levels \( ^6S_{1/2} \) and \( ^4D_{3/2,5/2} \) are considered to be strongly collision coupled for the conditions of interest in our experiment (\( N \sim 10^{12} \text{ cm}^{-3} \), \( N_e \sim 10^{14} \text{ cm}^{-3} \), \( T_e \sim 2500 \text{ K} \)). This is indicated by the double arrows in Fig. 2. Thus, each set of levels will be assumed as sharing the same population, in the ratio of their respective degeneracies. Proper consideration of collisional rate constants, indicates that levels \( ^4F_{7/2} \), \( ^4F_{5/2} \) are also closely coupled to levels \( ^6S_{1/2} \) and \( ^4D_{3/2,5/2} \), although not as strongly as these two.

In view of the little difference in the results when levels \( ^4P_{7/2,5/2} \) are considered fully coupled or uncoupled to levels \( ^6S_{1/2} \), \( ^4D_{3/2,5/2} \) and the fact that we obtain good agreement with the computer results in the latter case, we feel justified in considering only the levels indicated in Fig. 2.

2.3.1 Initial Pumping Stage

In the initial pumping stage the interaction of the laser resonant radiation with the atomic system can be adequately represented by a simple \( ^4 \)-level system. These levels are designated by the numbers 2, 2', 6 and 7 in Fig. 2. During this period, the laser induced rates dominate all other rates into or out of the \( ^4 \)-level system.

The corresponding rate equations can be written:

\[
\frac{dN^*}{dt} + \frac{dN^0}{dt} = R_{27} N^* \quad \frac{dN^0}{dt} = R_{72} N^* \quad (6)
\]

\[
\frac{dN^*}{dt} + \frac{dN^0}{dt} = R_{72} N^* \quad \frac{dN^0}{dt} = R_{27} N^* \quad (7)
\]

* All doublets other than the \( ^4P_{3/2,1/2} \) set are treated in the present text as a single level.
where:

\( N_6^*, N_7^*, N_2^*, N_7^* \), represent the perturbed population densities of levels \( 6^2S_{1/2}, 4^2D_{5/2}, 3/2, 4^2P_{3/2} \) and \( 4^2P_{1/2} \) respectively.

and

\[
R_{27} = B_{27} \int_{\nu} L_\nu I_\nu \, d\nu ; \quad R_{72} = \frac{g_2}{g_7} R_{27} \quad (8)
\]

\( R_{27} \) represents the rate constant corresponding to laser absorption, \( B_{27} \) being the Milne absorption coefficient, \( L_\nu \) the absorption line shape and \( I_\nu \) the laser intensity, at frequency \( \nu \). By conservation, we can also write:

\[
N_7^* + N_6^* + N_2^* \equiv N_2^* + N_2' \equiv N_2^0
\]

where \( N_2^0 \) represents the population density of the resonance doublet prior to irradiation and we neglect the initial population of levels 6 and 7. Because of our assumption of strong collision coupling between levels 6-7 and levels 2-2', we can write:

\[
N_2' = \frac{g_2'}{g_2} N_2^* \\
N_6 = \frac{g_6}{g_7} N_7^*
\]

(10)

Thus, using (6), (9) and (10), and introducing the proper numerical values for the degeneracies, we readily obtain:

\[
\ln \left[ 1 - \frac{N_7^*(t)}{N_7^{\text{max}}} \right] = - \int_{-\infty}^{t} R_{27}(t) \, dt
\]

(11)

where \( N_7^{\text{max}} = N_2^0/9 \) (see Appendix A), and \( t = 0 \) represents the time for which the laser pulse reaches its peak amplitude. An adequate representation of the laser temporal intensity distribution is:

\[
I_\nu = I_\nu(0) e^{-t/\tau_r}
\]

(12)

for \( t \leq 0 \). This has been confirmed experimentally, in a dynamic range of five orders of magnitude (see Fig. 13). \( \tau_r \) is the laser pulse risetime. Furthermore, under our experimental conditions, the laser line width is considerably smaller than the absorption line width of the \( 6^2S_{1/2} - 4^2P_{3/2} \) transition, which is \( 0.02A^0 \) (see Fig. 12 and Appendix B).
We can thus write:

\[ R_{27}(t) = B_{27} L_v I_\phi(o) e^{t/\tau}, \quad t \leq 0 \]  

(13)

We indicate by "v" the frequency of laser emission. We neglect changes of \( L_v \) during the absorption process. This is justified in view of the low laser power density necessary to saturate the transition, which is about 50 watts/cm\(^2\) in the present experiment, and the fact that the absorption line width is determined by doppler broadening (see Appendix B). Inserting the expression for \( R_{27}(t) \) in (11) and integrating, we obtain:

\[
\frac{N^*_7(t)}{N^*_7} = 1 - e^{-B_{27} L_v I_\phi(o) \tau \tau} e^{t/\tau}
\]  

(14)

which is valid for \( t \leq 0 \). Equation (14) represents the saturation behaviour of the upper level population of the laser pumped transition. The result is the same if one includes in the analysis the ground level, which indicates that feeding of population into level \( ^4P_{3/2} \) from this vast population source is negligible in the early stage of the process.

In Fig. 3 we indicate the saturation curve of the transition \( ^4P_{3/2} - ^6S_{1/2} \) as a function of laser peak power (i.e., \( t = 0 \)), calculated using Equation (14). Computer results obtained by Fournier, for the same laser pulse shape, are included for comparison. Figure 3 shows clearly the extreme non-linearity of the excitation process, and indicates a threshold of approximately 50W/cm\(^2\) for saturation of the transition. Since we have neglected to consider the population loss from the upper levels, the saturation power predicted by the simplified model is relevant only for the assumed laser pulse shape. In fact, for ideal loss free upper levels, saturation could be achieved with nearly zero laser power, in an infinitely long time. In general, in a realistic case, the saturation power will depend on the temporal characteristics of the laser pulse. Provided the conditions assumed in the simplified model prevail, Equation 14, or its equivalent for a different laser pulse shape, can be used in order to estimate this power.

Using Equation (14), we have also calculated the temporal variation of \( N^*_7(t) \), normalized to \( N^*_7 \), for different laser peak powers. Results are indicated in Fig. 4. The computer results are included for comparison, corresponding to a laser power density of \( 10^3W/cm^2 \), for the same pulse shape and \( Ne = 2 \times 10^{12}cm^{-3}, Te = 2500^\circ K \). The fact that our results, which are independent of the plasma condition, are in good agreement with the computer results, seems to indicate that under the present circumstances the laser induced rates indeed dominate over collisional rates, as we have assumed and that losses from the upper excited levels are negligible in this early period.

An interesting feature of Equation (14), is that it shows clearly the equivalence of laser power \( (I_\phi) \) and laser wavelength mismatch (represented by \( L_v \)) on the degree of enhancement of the upper level.
Finally, we have calculated the effects of laser wavelength mismatch for constant laser peak power, on the ratio \( \frac{N_7^*(t)}{N_7^*_{\text{max}}} \). For this purpose, we have computed the absorption line shape (see Appendix B) which we use to relate \( L_\nu \) to the laser wavelength mismatch \( \Delta \lambda \). Results are indicated in Fig. 5 for three different laser peak powers. We can see that as the laser power is reduced, the wavelength tuning requirements are considerably more severe, in order to saturate the transition.

2.3.2 Decay Period, Immediately After Saturation

As the transition excited by the laser saturates, the populations of levels \( ^2P_{3/2}, ^2P_{1/2}, ^2S_{1/2} \) and \( ^2D_{3/2,5/2} \) become locked together, because of the strong collisional and radiative coupling. Thus, these levels can be considered as sharing the same population, in the ratio of their respective degeneracies. A population loss (or gain) by any of these levels, will be felt immediately by the other members of the group who will readjust their respective populations to compensate for the change. Thus, as a result of collisional depopulation of the upper levels \( ^2S_{1/2}, ^2D_{3/2,5/2} \), the total population of these four locked levels will decay. The relatively unpopulated levels in the manifold (see Fig. 2) will receive this excitation energy. At the same time, the ground level will feed by electron collisions population to the resonance levels. The physical picture can be understood by reference to Fig. 2. We can think of the four locked levels as a single coalesced level, which is fed from the ground level and loses its population to the manifold of levels. Losses to ground level are negligible initially. Furthermore, we neglect in the early decay period any population feedback from the manifold levels, since we are assuming a very large population enhancement of the upper levels 7 and 6.

The model adopted for this period can be analyzed simply in terms of rate equations. In fact, except for the addition of levels \( ^2P_{1/2} \) and \( ^2D_{3/2,5/2} \), it is identical to the one considered by Measures(8).

We can write the corresponding rate equation for the change of the population of the four locked levels:

\[
\frac{dN_7^*}{dt} + \frac{dN_6^*}{dt} + \frac{dN_2^*}{dt} + \frac{dN_2^*}{dt} = -N_7^*(N_{D7} + A_7) - N_6^*(N_{D6} + A_6) + N_7N(X_{12} + X_{12'})
\]

where:

\[
D_7 = \sum_{n > 7} X_{7n} + \sum_{r < 7, r \neq 7_2} X_{7r} + X_{7c}
\]

and

\[
A_7 = \sum_{r < 7, r \neq 7_2} A_{7r}
\]
Here, \( X_{7n} \) represents the collisional excitation rate coefficient to a generic upper level, \( n \), \( X_{7c} \) is the ionization rate coefficient from level 7, \( K_{7r} \) represents the collisional de-excitation rate coefficient to a lower generic level \( r \), where levels 2 and 2' are excluded (since de-excitation to a level of the "locked" system does not represent a loss for the system). \( A_7 \) is the total radiative transition probability of level 7. \( X_{12} \) and \( X_{12'} \) represent the collisional excitation rate coefficients from the ground level to levels 2 and 2'. Since these two levels have approximately the same energy, we shall assume \( X_{12} \approx (\frac{6}{6 - 2'}) \cdot X_{12'} \).

Making use of the fact that the populations of the locked levels are in the ratio of their respective degeneracies, we can write Equation (15) in the form:

\[
\frac{9dN_7^*}{dt} = -N_7^* \left[ N_e (D_7 + 5D_6) + A_7 + 5A_6 \right] + X_7 \cdot \frac{N_e}{N} \cdot X_{12}
\]  

We shall define the depopulation time constant \( \tau \) as:

\[
\tau = 9[N_e(D_7 + 5D_6) + A_7 + 5A_6]^{-1}
\] 

Assuming \( N_1 \) constant, which is reasonable in view of the large population of the ground level compared to the excited level populations for the temperature of interest in this analysis (\( T_e \sim 2500^\circ K \)), we can solve equation (16) and we obtain:

\[
\frac{N_7^*(t)}{N_{7,\text{max}}} = e^{-t/\tau} + \frac{1}{3} X_{21} N_e \tau (1 - e^{-t/\tau})
\] 

In equation (18), we have made use of the relation:

\[
\frac{N_7^*(t)}{N_{7,\text{max}}} = \frac{N_e^0}{9} = \frac{N_e^2}{6} = \frac{X_{12} N_1}{6 X_{21}}
\] 

on the basis of the principle of detailed balance. For conditions where radiative decay is negligible compared to collision losses, the population of level 7 (\( 6^2S_{1/2} \)) decays initially with a time constant inversely proportional to \( N_e \), as indicated by equations (17) and (18). The contribution of the second term of equation (18) is negligible in this first period. The decay time also depends to some extent on the electron temperature, through the coefficients \( D_6 \) and \( D_7 \).

Consequently, provided we can estimate the electron temperature independently, and that \( D_6 \) and \( D_7 \) can be calculated, we can determine \( N_e \) from an experimental measurement of the enhanced emission decay time, corresponding to the early stage of the decay period.

Equation (18) also indicates that a steady state regime will be finally
attained, where losses from the upper excited levels are balanced by the rate of feeding from the ground level, provided the laser power is maintained above saturation threshold.

For steady state conditions, we obtain:

\[ N_7^* (D_7 + 5 D_6) = 1.5 X_{12} N_1 \]  

(20)

as indicated by Equation (18).

In closer approximation, after the initial decay period it is necessary to take into account the feedback of population into the levels 2, 2', 6, and 7, which arises from the increased populations of the levels of the manifold system. The resultant net depopulation rates will thus vary with time, as new levels of the manifold come into collisional quasi-equilibrium with levels 6 and 7. Consequently for these conditions, the decay characteristics of level 7 will not be represented by a pure exponential, as was the case in the early period, and the results derived from equation (18) will not apply.

2.3.3 Steady State and Final Periods

In view of the arguments given above, feedback of population to levels 2, 2', 6, and 7 and also the ground level from the manifold of levels is likely to be important. The computer results shown in Fig. 6 indicate that for large laser power densities (i.e., where there is an extended period for which the laser power exceeds the locking threshold) a steady state is reached, where the locked levels population remains constant. We tend to imagine this condition as determined by a balance between the losses from the locked levels and feeding from the ground level and also feedback from the manifold level. The condition that characterizes this stage is

\[ \frac{N_7^*}{N_7^{max}} = \text{constant} \]  

(19)

Finally, as the laser intensity falls below the excitation threshold, the locking of levels 7 and 6 with levels 2 and 2' is broken, and the upper levels, as well as the manifold of levels, decay. Typical computer results, taken from Fournier calculations are shown in Fig. 6 for two different laser powers. For the high power case, a steady state is reached. For the low power (near saturation threshold) the plateau condition does not exist.

2.4 Limitations of the Theoretical Model and Discussion of Some Implications of the Analysis of the Simplified Model

An important feature of the model assumed by Fournier, on which we base the analysis of our results, is that the collisional rate coefficients and the populations are not readjusted according to any variation that may be induced in the free electron temperature. For laser pulses for which time that the laser power exceeds the threshold for saturation is short (i.e., less than about 10 nsec) the perturbation of the electron temperature is predicted to be negligible.
However, if the time for which the laser power exceeds the excitation threshold is long and the electron density is high then the temperature is found to increase with time as long as the laser is above threshold. If proper temperature dependent rate coefficients are considered, it is estimated that ionization enhancement would result and this effect would impose a limit on the temperature increase (22).

It should be realized also that under real conditions, there will be an "enhanced" electron temperature profile in the volume excited by the laser, that will follow closely the laser spatial intensity profile. The temperature gradients are likely to be large, and if in addition the diameter of the excited plasma volume is small, diffusion of electrons out of the excited volume is likely to reduce further the effects of local temperature elevation.

From this discussion, it appears that a full evaluation of the effects of electron heating requires not only modification of the atomic model, but consideration of the particular geometry of the laser beam and of the detailed diffusion processes in the plasma. This aspect is far beyond the purpose of this thesis, and in practise this problem can be avoided by use of a short duration pulse.

It is sufficient to indicate here, that on the basis of the computer results, and for the typical laser powers used, the model should represent closely our experimental conditions to $N_e \approx 1 \times 10^{12}$ cm$^{-3}$. For higher electron densities, decay times calculated using computer results are likely to be overestimated. This is because under these conditions, collisional rates may in fact be higher, if the temperature increases appreciably. Thus the quenching of the upper level population will be faster.

A second limitation of the theoretical model is that it relies mostly on cross-sections for collisional excitation and de-excitation which are generally considered only accurate within a factor of 2 to 3. The lack of experimental data prevents improving on this point.

The analysis of the early excitation period with the simplified model, has illustrated clearly the extreme non-linearity of the saturation process. In particular we have observed that for the conditions considered ($N_e \sim 1 \times 10^{12}$, $T_e \sim 2500^\circ$K) an exponentially increasing laser pulse (with a 5 nsec risetime) saturates the transition at approximately 50W/cm$^2$. The time at which saturation takes place, relative to the time for which the laser pulse reaches its peak amplitude was found to depend on the laser power. The time difference increases with increasing power. If now we consider that the laser beam, in addition to its temporal profile, has a spatial intensity distribution with extremely large gradients, it is easy to see that saturation will occur at different times at different radii of the excited plasma volume.

Saturation of the transition will occur initially along the axis of the laser beam, where the power density is greatest. The region of saturation will then expand radially as the beam intensity increases.

This space-time correlation is indicated schematically in Fig. 7. Thus, although locally the temporal variation of intensified emission depends only on the local laser power and the local plasma conditions, the signal detected by a photomultiplier, observing the radiation from the excited volume, will represent a spatial average of the local signals. Awareness of this effect is crucial for the interpretation of results.
CHAPTER 3

3. EXPERIMENTAL CONSIDERATIONS

3.1 Purpose of the Experimental Programme

The general purpose of this experiment is to study the application of laser selective excitation techniques to plasma diagnostics. It should be pointed out that at the time this programme was started, in 1967-68, no previous work had been done on the subject. In fact, published experimental work first appeared in 1972 (11) (25). In view of this fact, much of the present experimental study is exploratory in nature, and a heavy emphasis has been placed in understanding the basic physics of the resonant laser-plasma interaction. As experience was gained, usually the hard way, much of our initial views on the subject have changed, although in some instances too late to make the necessary modifications in the experiment. This fact should be kept in mind when reading the next two chapters.

The experiment was designed to answer the following general questions:

1) Can the enhanced plasma emission signal be clearly identified above the inherent plasma radiation background and the scattered laser radiation contribution?
2) Does this enhanced emission behave in the general way expected?
3) Can intensification be observed on several spectral lines?
4) Is there any polarization of the enhanced emission?
5) What part do the laser beam parameters (power, wavelength, temporal and spatial profile) play in determining the characteristics of the enhanced emission signal?
6) How does the plasma condition effect the enhanced emission signal?

From the practical point of view it was important to evaluate the diagnostic potential of this form of laser-plasma interaction. In particular, since the cross sections which determine the decay characteristics of the enhanced plasma emission are not known accurately, it was of interest to measure experimentally the enhanced emission decay time under known conditions of the plasma, in order to evaluate these cross sections in a global fashion, and obtain a series of calibration curves $\tau = f(N)$ for a range of relevant temperatures. This will be referred to as the "calibration experiment". Finally, we wished to determine whether the laser excitation produced any serious observable perturbation to the plasma, such as ionization enhancement.

3.2 Experimental Requirements

In order to study the application of selective excitation diagnostic techniques, the plasma and laser used had to fulfill certain basic requirements. Furthermore, we required auxiliary diagnostic methods, able to evaluate independently the local plasma conditions.

3.2.1 Plasma Requirements

In order to produce a significant emission enhancement, Eq.(3) requires:
\[ h\nu_L \gg kT_{ex} \]

which indicates that the mean thermal energy of the free electron has to be small compared to the energy level difference of the excited transition.

Moreover, we require the lower level of this transition to be close to the ground level, in order to have a large signal. Alternatively, for the decay type measurements a highly populated high energy metastable level may be used, as indicated by Burrell and Kunze\(^{10}\).

The energy of the upper level, relative to the ionization energy, determines the relevant range of electron densities for which this technique can be used. If the upper level is near the continuum, collisional quenching may be very fast, even for moderate electron densities, and unless the detection system has an extremely high time resolution, it will not be possible to measure accurately the decay time. Moreover, the signal will be small due to the quenching.

Potassium has an adequate energy level diagram, and the \( ^4P_{3/2} \rightarrow ^5S_{1/2} \) transition, which corresponds to a wavelength of 6939\(\AA\),\(^*\) is within the range of tunability of a ruby laser.

In addition, a potassium plasma could be produced to have the conditions suitable for this study (i.e., \( T_e = 0.25 \text{ eV}, N_e \approx 10^{11}-10^{13} \text{ cm}^{-3} \), according to Ref. 8) furthermore, such a potassium plasma was of general interest in the MHD field. Thus, a potassium plasma was chosen as a suitable example to demonstrate the diagnostic application of selective excitation spectroscopy.

### 3.2.2 Laser Requirements

For this experiment a tunable, moderate power laser with good emission-wavelength reproducibility from shot-to-shot was required. Medium Power Tunable lasers were in a very primitive stage when this experiment was started. Abella and Cummins\(^{32}\), demonstrated the possibility of tuning a ruby laser over a range sufficient to encompass the 6939\(\AA\) line of potassium by control of the ruby temperature. In view of this we decided to use a ruby laser with thermal tuning as the source of excitation.

### 3.2.3 Requirements Concerning the Reference Diagnostic Method

In order to relate the measured signal characteristics with \( N_e, T_e \), as discussed in connection with the calibration experiment we required a well diagnosed plasma.

Since we were interested in the local plasma conditions, we looked for a reference diagnostic method which provided spatial resolution; the obvious choice was to use a Langmuir probe (33).

In view of the experimental complications when using a Langmuir probe in an alkali plasma, we decided also to obtain extra information by spectroscopic measurements of the radiative recombination continuum (34) into the

\[ ^4P_{3/2} \rightarrow ^5S_{1/2} \] transition is 6938.77\(\AA\).

For briefness of notation, we shall write this wavelength as 6939\(\AA\) throughout the text.
$^{4}\text{P}_{3/2,1/2}$ levels of potassium. From the spectroscopic measurement, the average value of $\text{Te}$ along the line of sight can be obtained. Electron densities can also be calculated from an absolute measurement of the recombination continuum intensity.
4. DESCRIPTION OF EQUIPMENT

4.1 The Tunable Ruby Laser

In view of the lack of commercially available units with proper wavelength tuning capability at the time this experiment was started, it was decided to design and build our own laser.

The design requirements were:

1. Laser emission wavelength: \( \lambda_L = 6939\text{A} \)
2. Wavelength Reproducibility: \( \Delta \lambda_L \sim 0.01\text{A} \)
3. Ruby Operation Temperature: \( T_r \sim -50^\circ\text{C} \) (32)
4. Ruby Temperature Stability: \( \Delta T_r \leq \pm 0.2^\circ\text{C} \)
5. Power Output: \( 10^5 - 10^6 \text{W peak} \)
6. Transverse Mode Output: \( T E M_{\infty} \)
7. Cooling System: Cold Nitrogen vapour flow
8. Mode of Operation: Q-switched

The laser wavelength reproducibility is estimated by requiring the emission wavelength to be within the Doppler half width of the 6939A absorption line of neutral potassium, which for our experimental conditions is about 0.02A (see Appendix B). As a consequence of taking the wavelength thermal coefficient for ruby to be \( \Delta \lambda / \Delta T_r = 0.053\text{A/}^\circ\text{C} \) \( @T_r = -50^\circ\text{C} \) (35), we arrive at the temperature stability requirement of \( \pm 0.2^\circ\text{C} \).

Several practical problems had to be solved before making the final design.

The finished version of the laser incorporated an elliptical, mirror polished aluminum cavity, of about 5 cm mean diameter. We used a linear xenon flashlamp EG & G type FX-47C-3 and a Linde S.I.Q. ruby rod, 90° axis orientation, 6.35 mm in diameter by 76 mm length, with a 90° prism end. The effective cavity length was 60 cm. The ruby and the flashlamp were placed at the focal axis of the elliptical cavity, for maximum pumping efficiency. A Raytheon LPS-12A laser power supply was used to drive the flashlamp. The unit was modified to match the flashlamp impedance.

The entire laser cavity was flushed with cold nitrogen vapour, which was produced using a small cryostat (36). The flow entered the cavity through a series of small nozzles at the rear end of the ruby rod, and left the cavity through a similar arrangement at the front end of the ruby. This cooling configuration ensures a nearly laminar flow over the ruby. Although in our design the flashlamp was directly in contact with the cold vapour, this had no detrimental effect in its pumping efficiency, as was verified by measuring the flash luminous intensity with a photodiode, down to a temperature of -70°C.
The temperature of the ruby was determined by the vapour rate of flow. A coarse flow control was obtained by adjusting the ohmic dissipation of the cryostat heater, by means of a variac. A finer temperature control was incorporated. It consisted of a small chromel heater at the entrance of the laser cavity which was automatically switched on whenever the inlet flow temperature fell below a pre-set value. As the temperature increased, a flow temperature sensor (thermistor), which formed part of an A-C wheatstone bridge, disconnected the heater.

By means of a combination of the coarse control and the feedback system, we have achieved an inlet flow temperature stability of about ± 0.5°C at any set temperature value down to -70°C. The ruby temperature itself was measured using a copper-constantan thermocouple and a thermistor.

The laser was Q-switched using a saturable absorber. The performance of cryptocyanine dissolved in methanol was found quite poor, due to deterioration of the solution after short periods of time. This led to poor reproducibility in the laser performance. We found more convenient Kodak's Q-switch solution 10220 (cryptocyanine in acetonitrile) regarding stability, although for good performance the concentration of the solution had to be adjusted periodically. Although this type of Q-switch is far from ideal regarding shot-to-shot wavelength reproducibility, it has the virtue of simplicity. The laser and associated equipment is illustrated in Figs. 8 and 9.

4.1.1 Laser Performance

(a) Wavelength Tuning and Shot-to-Shot Reproducibility

The laser output wavelength was analyzed using a Fabry-Perot interferometer of adjustable plate spacing. The interference ring pattern was photographed using Polaroid 413 infrared film. The photographic camera had its focal plane divided in two halves by means of a moveable diaphragm, such that only one half of the plate was exposed at the time. The dividing line was adjusted to coincide with the vertical line of symmetry of the interference ring pattern. In this way, we were able to record on the same film the laser light interferogram and a reference one corresponding to the 6939A line of potassium. The latter was obtained using an Osram Potassium spectral lamp with an adequate narrow band interference filter. Knowing the interferometer free spectral range, the laser wavelength mismatch could be readily calculated by measuring the fringe shift of a given order. Care had to be taken initially to avoid mixing of orders, and usually measurements were repeated using different free spectral ranges. In this way, we obtained the general dependence the laser emission wavelength on the initial ruby temperature. These results are indicated in Fig. 10. The operational temperature required to match the 6939A potassium absorption line was measured to be $T_R = -45.0°C$. At the same time, we studied the output wavelength reproducibility. Measurements on the basis of ten successive shots indicate that the mean square deviation about the average is 0.05A. This is illustrated in Fig. 11, where only five shots are shown. The main sources of irreproducibility are:

(i) Initial ruby temperature fluctuations from shot-to-shot.

(ii) Random shot-to-shot fluctuations of the Q-switching time. This implies different amounts of energy are absorbed by the ruby and consequently the ruby can have a different temperature at the instant of laser action.
Both of these problems can be alleviated by improving the temperature control system, and by replacing the passive Q-switch by an active one, such as a Pockels cell. Alternatively, we could have replaced the front mirror by a thermally tuned etalon for mode control (37).

Later experimental evidence indicated that the performance of the tuning system, as described above, was quite acceptable for practical purposes. Thus, no modifications were made.

(b) Laser Line Width

The line width was measured using the Fabry-Perot interferometer. The free spectral range was set to 0.024 Å, corresponding to a plate separation of 100 mm. The results are illustrated in Fig. 12. Since the absorption line width (0.02 Å) is practically equal to the free spectral range, we can clearly see that the laser line width is considerably smaller, which justifies our treatment of the laser emission as being single frequency, in the theoretical analysis in Eq. (13), to a first approximation. *

(c) Laser Pulse Temporal Characteristics

The pulse width can be adjusted by varying the concentration of the Q-switch solution. On the short side, we were able to obtain pulse widths (FWHM) of the order of 8-10 nsec at maximum concentration. The pulse shape was measured using a Hewlett-Packard PIN photodiode (5082-4220) with a claimed response time of the order of 1 nsec. The photodiode output was measured with a Tektronix 7704 oscilloscope, with a bandwidth of 150 MHz (2.4 nsec risetime).

In view of the **averaging** effect discussed in Section 2.4, it was deemed important to measure the shape of the laser pulse in its early build-up stage. This was done over a dynamic range of five orders of magnitude. The results are indicated in Fig. 13.

(d) Laser Pulse Spatial Characteristics

Quantitative analysis of the **averaging** effect requires a knowledge of the laser intensity distribution in the plasma at the point of observation. At the time we realized the need for this measurement, the plasma source had already been constructed and measurements at the real test section were not possible. Thus, we built an exact replica of the relevant forepart of the plasma source and scanned the mid-horizontal plane of the test section with a photomultiplier provided with a pinhole (0.3 mm dia.) and a stack of neutral density filters that were removed gradually as we moved away from the beam center.

Particular care was taken to reduce the laser stray light to a minimum when measuring the profile wing intensities. We were also careful to use exactly the same optics as employed in the real case.

Results of three independent scans are illustrated in Fig. 14. The spatial intensity profile, measured over a range of five orders of magnitude, was extrapolated down to seven orders, for the purpose of interpretation of results, as we shall discuss later.

It is important to note that the measured spatial profile corresponds to the instant of peak power. In fact, the spatial intensity profile will change

* However, a more rigorous analysis requires the consideration of hole burning effects.
with time (38) and to a very minor extent, the wavelength will also change, during the evolution of the Q-switched pulse. This subject will be discussed to some extent in Chapter 6.

(e) Laser Output Power

The laser power was monitored using a calibrated PIN photodiode (H-P 5082-4220). The calibration was performed by measuring simultaneously the laser pulse shape (with the photodiode) and the laser output energy with a TRG Ballistic Thermopile Model 100. The laser produced peak powers of the order of 5 MW. The laser beam was focused at the plasma test section, using a 730 mm focal length lens. At this point, after losses in the optics and windows, power densities of the order of 10 MW/cm² were obtainable. This is well in excess of the power density required for our experiment. Calibrated aqueous solutions of CuSO₄ were used to reduce the intensity of the beam. We have verified experimentally that such solutions behave as passive attenuators, without producing a distortion of the wavefront. Similar results have also been reported, confirming this point (39).

In general, the shot-to-shot peak power reproducibility was poor. Fluctuations were within a factor of 2 of the average power.

4.2 The Potassium Plasma Source

The plasma source main design requirement was that it should produce a plasma possessing the following conditions:

\[ T_e \sim 2000 - 3000 \text{°K} \]
\[ N_e \sim 10^{11} - 10^{13} \text{ cm}^{-3} \]

Whether the plasma was pulsed or D.C., or whether thermal equilibrium was established between the free electrons and the heavy species, were matters of secondary concern.

A survey of available plasma sources led to selection of the following candidates.

(a) Potassium Seeded Shock Tube (40)
(b) Isothermal Plasma Oven (41)
(c) Rarefied Potassium Seeded Flame (42)
(d) Plasma Low Voltage Arc (43)

We shall briefly discuss each possibility.

(a) The development of a potassium seeded shock tube has associated with it several major problems. The shock tube would have to be made of stainless steel, and enclosed in an oven, to prevent vapour condensation on the walls at temperatures of the order of 350°C. Furthermore, the seeding concentration has to be uniform, and seeding reintroduced after each shot, which means that after each shot a whole sequence of time consuming events would be necessary before attempting the next. This, added to the transient condition of the plasma, ruled out a shock tube approach to the problem.

(b) An isothermal plasma oven, such as the one developed by Agnew and
Summers for cesium, seems the ideal facility, producing a plasma in thermal equilibrium that can be studied at leisure. Nevertheless, their design is not adaptable to our experiment where two windows at right angles are required, one for the laser beam input, the other for observation purposes. Incorporating an observation window that would operate at 3000°C in potassium vapour is not possible, since under optimum conditions a window would tolerate only 1000°C at most, assuming a sapphire window that has to remain leak tight and transparent. Removing the window towards a cooler region would probably create large temperature gradients that would have to be overcome using a complicated heating system; when the temperature of the oven is 3000°C and we require leak-tightness for an element as corrosive as potassium, the task is really formidable.

(c) The possibility of producing a flame plasma, by seeding a rarefied flame with small amounts of potassium vapour is an appealing one, since the high temperature region is only at the burner exit and observation windows can be placed out in cooler regions where potassium attack will not be severe. Also, the plasma will be close to thermal equilibrium, and its condition can be easily regulated by modifying the seeding rate or the flame characteristics. Nevertheless, one serious drawback of this plasma source is the presence of the flame gases (i.e., acetylene-air), which constitute an impurity not easy to account for, since these foreign gases may accelerate the "quenching" of the excited level. Due to this uncertainty, this approach was not considered.

(d) Low Voltage Arc

In a low voltage arc, or plasma diode, the plasma is produced between two heated electrodes separated by a short gap. This is not a plasma in thermal equilibrium, at least for low vapour pressures of the order of 1 mm Hg, but it has the advantage of a cylindrical geometry and steady state conditions \( (N, T) \) that can be regulated by adjustment of the vapour pressure or the arc current. It is also possible to place the windows close enough to have a good field of view, yet removed enough from the high temperature arc region, so as to minimize the attack of the potassium. Its only drawbacks are those also associated with the previous system: difficulty in maintaining its leak-tightness due to potassium corrosion at temperatures around 350°C.

This plasma source seemed the most suitable one for the experiment, and it was selected accordingly.

The plasma source and details of some of its components are illustrated in Fig. 15.

The structural part of the discharge chamber was made of stainless steel. The two electrodes, also made of stainless steel, were heated indirectly using chromel wire heaters, insulated from the electrode surface by means of thin boron-nitride spacers. The electrodes were 20 mm in diameter, with an inter-electrode gap of 12.7 mm. Two sapphire observation windows were incorporated opposite to each other (Elmac, Model X6195-100A), mounted on Varian Conflat-flanges with OHFC copper gaskets. A third sapphire window was welded directly to the structure and was used for the laser input. The two
electrodes were insulated from the main body of the plasma source using special high purity Alumina ceramic spacers (Eimac).

The Langmuir probe was mounted on a bellows assembly which permitted it to scan the horizontal plane of symmetry of the interelectrode region. This system also facilitated the removal of the probe from the path of the laser beam when firing the laser. The probe output electrical connection was made using a sapphire feedthrough pin. The potassium was contained in a reservoir attached to the main structure. Under normal operating conditions, the whole system was enclosed in a two compartment oven. The major compartment contained the main body of the plasma source and was always kept at about 350 °C, to prevent condensation of the vapour on the walls or windows. The second compartment contained the potassium pod and its temperature was always lower than that of the rest of the system at least by 70 °C, and so it determined the potassium vapour pressure within the plasma source. The temperature at all the vital points of the structure was monitored using Chromel-Alumel thermocouples.

As the plasma source was finally assembled, it was connected to the vacuum system and evacuated to about 10⁻⁷ Torr. This was achieved after prolonged pumping and outgassing at 350 °C (the electrodes were heated to 900 °C). At this point, the potassium was distilled into the system. We used high purity potassium (99.95%) obtained from Mines Safety Appliances (M.S.A.) in a 3 gram glass ampoules. After distilling for about 20 hours, we isolated the system from the vacuum system (using a bakeable all metallic valve) and the plasma source was ready for operation. The discharge was produced using a Hewlett-Packard DC variable power supply, model 6433B (0-36V, 0-10 Amp).

4.2.1 Some Problems Associated with the Development of the Plasma Source

Preliminary estimates suggested that we would require to operate the plasma source at potassium vapour pressures near to 1 Torr. This corresponds to a temperature of nearly 350 °C (44). Subjecting the whole plasma source to such a high temperature, puts serious constraints on the materials to be used.

Another constraint on the design was related to the need of minimizing the amount of laser light scattered by the walls and other parts of the system, out of the observation window. This could possibly obscure the enhanced plasma emission signal which was expected to be fairly weak.

These two particular features of the design, i.e., material compatibility and the stray light rejection, will be briefly discussed.

(a) Materials Compatibility

Potassium vapour at temperatures near and above 300 °C is an extremely corrosive agent; its attack on materials proceeds via two principal types of corrosion: dissolutive corrosion, where the material is simply dissolved by the potassium, and reactions with impurities, whereby the presence of certain elements in a material, such as O, N, H accelerate the corrosion process; materials that in a pure state are compatible with potassium can become attacked provided certain impurities, especially oxygen, are present. In the case of glass, the attack at temperatures above 300 °C shows itself by the darkening of the glass to the extent that it becomes opaque after a short
exposure to the vapour. The darkening seems to be associated with diffusion of
the potassium into the glass (45); a secondary reaction also takes place and
this is the chemical reaction of the potassium with the glass, which changes
the expansion characteristics of the original glass. This can become a serious
problem in the case where the glass is sealed to Kovar, especially for large
seals, since now the mismatch leads to cracks in the glass and leaks develop.

Our initial design for the plasma source incorporated glass as the
major structural and electrical insulating element.

After a short operational period of 7 hours at a temperature of
350°C, the prototype model developed leaks in all glass-to-metal seals and
became useless.

A considerable time was subsequently spent trying to find a glass
that would not be affected by potassium. Tests were conducted with Corning
7052 glass and fused silica; surface treatments were tried in order to improve
the resistance, even special coatings resistant to alkali vapours were tried,
all without success. At this point, the whole concept of the plasma source
was revised and it was decided to develop a new model, incorporating stain­
less steel as the structural element, sapphire windows and special ceramic
electrical insulators. The new design falls in line with those used in
thermionic converters, and this is a field where a great deal of practical
experience and technical know-how has been accumulated in the past decade
regarding ceramic-to-metal seals for electrical insulation that can operate
within such corrosive environments up to extremely high temperatures.

Our ceramic-to-metal electrical insulator seals were developed
specially by Eimac for this application, and feature a 99.8% Al₂O₃ (alumina
oxide) ceramic cylinder, 3" O.D., 1.8" long, with a wall thickness of 1/8"
and provided with two Kovar flanges brazed to the ceramic using special
alloys.

The laser input and the observation windows were also specially
designed for a potassium vapour environment, at temperatures up to 350°C and
continuous service, and were manufactured also by Eimac.

The main limitation in the use of these components is oxidation of
some of the seal components, mainly Cb, when exposed to air at high tempera­
ture; hence, it is vital for an extended lifetime of the units, to operate
at a maximum temperature not exceeding 350°C, after which degradation of
the seal develops extremely rapidly and leaks appear. Should higher operational
temperatures be required, it is necessary to work in an inert gas atmosphere
or even ultrahigh vacuum; for this last case, seals can be operated at over
1000°C with extremely long lifetimes.

The special electrical insulator seals and windows used in this
design are shown in Fig. 15.

(b) Reduction of Laser Stray Light

For a 1 KW power laser pulse, of about 10 nsec width, the number of
photons emitted Nₗ is of the order: \( Nₗ \approx 3 \times 10^{13} \) photons. Although, as we discussed earlier, we only need 50 W/cm² in order to
saturate the pumped transition, in practical experimental conditions the laser wavelength tuning becomes extremely critical for those low powers (see Fig. 5). Besides, the excitation volume, which depends on laser power, is too small to produce a meaningful signal. A peak power of about 1 KW, which corresponds to an average power density of 40 KW/cm², was found to be a practical compromise in our experiment in order to overcome the two problems mentioned above. The number of photons associated with the intensified emission pulse, emitted by the plasma into the solid angle of observation when excited by a 1 KW peak power laser pulse at 6939A, in the same time interval, is considerably smaller. A typical, realistic number, based on later experience (see Appendix C) is: 

$$N_{\text{signal}} \sim 2 \times 10^6 \text{photons.}$$

We see that the ratio:

$$\frac{\text{input (} N_L \text{)}}{\text{output (} N_s \text{)}} \sim 10^7$$

and unless extreme care is taken in minimizing the stray laser light, the signal is likely to be masked. This situation is similar, although not as severe, to that encountered in Thomson scattering experiments. The disadvantage in our case is the compatibility of materials with potassium which limits the selection of light absorbing surfaces. Fortunately, a stack of razor blades, when looked at edge-on, offers a black surface, since the wedge between two consecutive sharp edges provides a multiple reflection path for the incoming light and only about 3% is reflected back. This property is not affected when the razor blade trap is exposed to potassium vapour. This is based on our observations after exposing one sample trap to potassium vapour at 350°C for a period of 100 hours.

An extensive program was set up to optimize the parameters of the plasma container that would lead to minimum stray light contribution. The study was made using a model of the planned chamber that could be modified by adding or removing light stops, light traps, or even by changing its physical dimensions at will. Hence, the effect of each element of the design could be studied independently. A He-Ne laser was used to simulate the ruby laser, and the laser stray light intensity received into the solid angle of observation was monitored using a photomultiplier.

For the accepted design, we measured:

$$\frac{\text{laser stray scattered light}}{\text{laser input light}} \equiv \eta = 6 \times 10^{-10}$$

This means that we can expect about $2 \times 10^4$ photons from the laser to be scattered out into the observation solid angle, when the input number is $3 \times 10^{13}$. Since our signal has about $2 \times 10^6$ photons, the design proves to be satisfactory. Fortunately spectral discrimination can be used to further improve the signal to noise ratio. This is possible, since all transitions arising from the upper level of the laser excited transition experience the same relative intensity enhancement. The $6^2S_{1/2}$ level of potassium has two alternative decay modes

$$6^2S_{1/2} \rightarrow 4^2P_{3/2} (\lambda = 6939\text{A})$$

$$6^2S_{1/2} \rightarrow 4^2P_{1/2} (\lambda = 6914\text{A})$$
This means that we can monitor the emission at 6911A and still obtain all the relevant information.

Typical stray rejection values of spectral components are:

- Narrow Band Interference Filter $\eta \sim 10^{-2} - 10^{-3}$
- Monochromator $\eta \sim 10^{-4}$
- Double Monochromator $\eta < 10^{-6}$
- Filter + Monochromator $\eta \sim 10^{-6}$

These values were obtained experimentally using either a potassium spectral lamp or a He-Ne laser where possible, and measuring the amount of stray radiation at 6911A when the spectral discriminator in question was illuminated with 6939A radiation. When the He-Ne was used, stray radiation readings were done at 6300A. This shows that a proper design of the plasma source, coupled to a monochromator used as a spectral discriminator should completely overcome the problem of stray laser light.

The plasma source was designed on the basis of this experience, and the results have proved successful.

4.2.2 Performance of the Plasma Source

Langmuir probe measurements (see Appendix D) indicate free electron densities in the range $10^{12} - 10^{14}$ cm$^{-3}$, with fractional degrees of ionization of 1 - 10%. Recombination continuum measurements indicate electron temperatures of the order of 2000°K. The plasma source conditions could be varied by regulating the potassium vapour pressure, the electrode temperature and the voltage across the discharge. Typical performance characteristics are indicated in Figs. 16 to 19.

Several possible discharge modes could be produced. These ranged from seemingly glow type discharges, characterized by a very low current and reasonably high voltage ($i_L < 100$ mA, $v_L \sim 40$ volts) to very definite arc discharges ($i_L \sim 10$A, $v_L \sim 5$V). The visual characteristic modes of the low voltage arc could be clearly seen: anode glow, ball of fire, cathode glow mode.

The stability of the plasma conditions was investigated. For low current discharge modes, conditions were quite stable over periods of several hours (3-4 hours), as measured with the Langmuir probe. The stability of the discharge current was also investigated by measuring the voltage drop across a resistor in series with the discharge with an oscilloscope. We did not observe any obvious instabilities or oscillations in the frequency range investigated (1 MHz). For high currents, of the order of 1 Amp and above, stability deteriorated and conditions could change appreciably in a short time. All relevant measurements regarding laser selective excitation were done in the low current discharge mode.
4.3 Auxiliary Equipment

We would like to discuss briefly the time resolution characteristics of the detection system. This system comprises the photomultiplier-oscilloscope assembly.

We used a RCA C70042K developmental photomultiplier in most of the final measurements and a RCA 7265 phototube in the early stages of our experiment. From the point of view of time response, the developmental tube was superior, and this is the reason it was selected for final measurements. We investigated extensively the time response of both tubes under pulsed conditions. For this purpose, we used a red (6700A) light emitting diode (LED) (Monsanto MV 10A) driven by a Tektronix III pulse generator (5 x 10^{-10} sec risetime). Using an adequate driving circuit for the LED we were able to produce step-like light pulses of extremely fast risetime. Although we were not able to measure the exact risetime of the light pulse itself, we did measure the electrical pulse risetime across the LED using a sampling oscilloscope, which was about 1 nsec. If we believe the LED manufacturer's specifications, which claim 1 nsec risetime, our light pulse risetime should thus be close to that figure.

Using this arrangement, we measured a risetime (10%-90%) of about 9 nsec for the C700 42K phototube alone, using a sampling scope. This risetime is for step-like pulses. For 5-function type pulses the risetime is considerably faster, although in our experimental conditions we are closer to the first.

The combined PMT-oscilloscope (Tektronix 7704, 150 Mz) was measured to be about 10 nsec (10%-90%). The performance of the 7265 PMT was considerably worse; we measured a risetime of about 18 nsec for the combined system.

Alternatively, we used a double beam oscilloscope (Tektronix 556) when it was necessary to observe the time synchronization between the plasma signal and the laser pulse, or the plasma emission at other wavelengths. Optimum time resolution in this case was of the order of 12 nsec. The photomultiplier's saturation behaviour under pulsed conditions was also studied, in order to avoid exceeding the response linearity range. Both tubes exhibited about the same shot-noise behaviour. This characteristic can be quite serious in an experiment of this sort, where only a few photoelectrons are produced at the photocathode. To attempt to overcome this problem, we used a double bounce prism (46) mounted on the face of the 7265 phototube. Optical contact was made using silicone oil. In this way, we enhanced the quantum efficiency of the tube by a factor of two, as measured experimentally. We were not very successful in enhancing the quantum efficiency of the C70042K phototube by more than 35%. Consequently, this tube was not modified.
In this section we shall be concerned mainly with a description of the experiments and of the experimental results. Except in a few cases, no attempt will be made to interpret or discuss the results. This is left for the next chapter.

5.1 Experimental Procedure

The experimental arrangement is illustrated in Fig.20. The typical procedure and sequence of events corresponding to the laser excitation experiments can be described as follows.

The Ruby laser radiation at 6939A was focussed by a 730 mm focal length lens into the volume of plasma under observation. The laser output was linearly polarized with the electric vector oriented vertically. For each shot we measured both the laser peak power, and the laser output wavelength. The peak power was measured to within 5 to 10% using a calibrated photodiode and the wavelength was monitored using a Fabry-Perot interferometer and a reference potassium spectral lamp. The accuracy of this latter measurement was estimated to be about 5% of the instrumental free spectral range.

The plasma conditions prior to the laser excitation were monitored with a langmuir probe, at the volume of interest. The volume excited by the laser could be easily identified by using a He-Ne laser to simulate the main ruby laser beam. Two removable pinhole stops were used for this purpose. The position of these stops was determined initially using the ruby laser, and each pinhole was located exactly at the centre of the spot produced by the laser. Although the pinholes were removable, they could easily be relocated in proper position when required. Thus, the He-Ne laser was easily aligned along the same optical axis of the main beam. By moving the langmuir probe until the probe tip intersected the He-Ne beam, we automatically located the volume excited by the laser.

The plasma intensified emission, resulting from the laser excitation, was collected by a lens, and the radiation at a particular wavelength was selected using a monochromator (Spex, Model 1700-II), or alternatively, a narrow band (3A, FWHM) interference filter at 6911A.

The spectral interval passed by the monochromator was 20A. This was a result of using a 2 mm wide slit setting, necessary in order to observe a reasonably large plasma volume. The cross sectional area of the plasma, normal to the line of sight, was 4.8 mm high by 5.0 mm wide, as seen through the 6911A filter. This area was determined by a rectangular stop, placed just ahead of the filter. The cross sectional area of the plasma seen through the monochromator was 1.7 mm wide by 4.8 mm high. All these measurements are accurate to about 5%.

The magnification of the optics was 1.2 while the solid angle of observation was 0.057 steradians. The accuracy of both measurements was about 5%. The radiation selected by the monochromator, or the filter, was detected by a photomultiplier and the corresponding signal displayed on an oscilloscope. A fast polaroid camera was used to photograph the signal, using 410 type polaroid
film. Usually, the signal was displayed simultaneously on two oscilloscopes: a Tektronix Model 7704 (150 MHz bandwidth) for fast time resolution, and a Tektronix dual beam Model 556 (50 MHz) where the plasma signal was displayed simultaneously with the laser pulse, monitored by the photodiode. In this way, we measured the laser peak power and at the same time, could correlate in time the two signals. In general, the accuracy of signal peak amplitude measurements from the polaroid pictures is estimated to be 5-10%. The signal decay time measurements are believed accurate within 10%.

The fast RCA C70042K photomultiplier was always used in conjunction with the narrow band filter, to detect the radiation at 6911A. The RCA 7265 phototube was used in conjunction with the monochromator. For all measurements, except when investigating the enhancement of different transitions (other than \(6^2S_{1/2} \rightarrow 4^2P_{1/2}\)) we used the C70042K phototube.

As a regular practice we checked the level of laser stray light reaching the photomultiplier (by turning the discharge off (no plasma). This precaution was necessary in order to ensure that the signals to be measured had no contribution from the original laser photons. This was verified only once, for a given set of experimental conditions, and rechecked again when changing to a new setting.

In order to measure absolute light intensities, both photomultipliers were calibrated "in situ" for absolute measurements, using a calibrated tungsten strip lamp. The losses in the optics were included in the calibration. This calibration was required in order to perform electron temperature measurements, as discussed in Section 2.1 (see Equation (5)).

Unfortunately, the observation window of the plasma source that we selected initially for our measurements, became completely opaque after a short time of exposure to the alkali. The second, alternative window, suffered no obvious attack at all. The darkened window was relatively close to the discharge plasma, in an attempt to gain as much solid angle as possible, while the second window was recessed. This might be indicative that close contact of a sapphire window with an alkali plasma, presents a more severe constraint, regarding compatibility, than the contact with pure potassium vapour, at approximately the same temperature. No attempts were made to investigate this point. Removal and cleaning of the darkened window represented an extremely time consuming operation. The potassium had to be re-distilled out of the plasma source; the plasma source itself had to be opened up, cleaned thoroughly with solvents, in order to eliminate all traces of potassium; then the chamber had to be resealed, evacuated and outgassed before potassium could be distilled back into the system. Since this procedure also involved a risk of possible after-damage to the special ceramic-to-metal seals if the cleaning was not perfect, we decided to sacrifice the temperature measurements for the safer alternative of moving all observation equipment to the clean, auxiliary window.

5.2 Exploratory Experiments

The "exploratory experiments" had the main purpose of evaluating qualitatively the main feature of the laser-plasma interaction process. The results can be summarized as follows:

(a) Plasma enhanced emission signals have been observed at 6911A, 6939A and at a number of other wavelengths (4044A, 4850A, 5350A, 5782A, 5800A and
5832A), when the plasma was excited at 6939A. The signals do not exhibit linear polarization characteristics. The signals disappear by detuning the laser (see Fig. 21), when the discharge is switched off, or when observing at wavelengths slightly different from the specified lines. These results provide very strong evidence that the signals observed result from a population enhancement of the \(6^2\Sigma_{1/2}\) level created by laser selective excitation. The enhanced emission from some of the other spectral lines indicates that collisional excitation transfer to other excited levels occurs in a short time.

(b) Laser stray light was found to pose no problem for signal detection. In fact, for laser peak powers of about 1 kW, or lower, the stray light detected at 6939A is negligible, compared to the enhanced plasma emission at the same wavelength.

Furthermore, the enhanced peak emission signal at 6911A is about three orders of magnitude larger than the plasma background emission at the same wavelength.

Thus, there is no problem of identification of the plasma enhanced emission against the laser or plasma background light. Nevertheless, photoelectron noise in the photomultiplier can be a problem, especially when observing intensification of collisionally excited transitions. For excitation with a 1 kW laser pulse, and plasma conditions given by \(T_e \sim 2500\) K, \(N_e \sim 10^{12}\) cm\(^{-3}\), we obtained about \(7 \times 10^3\) photons at 6911A absorbed at the photocathode within the detection time of the phototube (~10 nsec) (see Appendix C). This value is typical of the signal peak amplitude. Under these conditions, the photoelectron statistical noise, for a typical quantum efficiency of 6% (extended red C70042K response) was calculated to be about 5%. Although this is not too serious at 6911A and 6939A, because of the large intensification factor, measurements at other wavelengths are seriously impaired, as illustrated in Fig. 22. These estimates also illustrate that the use of lower powers, under our present experimental conditions, will lead to increased shot noise in the 6911A and 6939A signals. Fortunately, the problem of photoelectron noise can be overcome to some extent by improving the efficiency of the observation optics.

(c) The decay time of the enhanced emission at 6911A was found to depend strongly on the discharge current, which controls the degree of ionization. For low currents (50 mA) the signal decay time is long, of the order of 50 nsec. For currents of the order of 500 mA, or higher, the decay time cannot be resolved with our present instrumentation, and we obtain just the instrumental resolution time of 10 nsec. By proper adjustment of the discharge current in the range 50 - 500 mA, we could obtain a continuous change in the signal decay time between the indicated values. Limiting results are illustrated in Fig. 23. We have also observed the effects of changes in the discharge current on the characteristics of the enhanced emission at other wavelengths. Typical results are illustrated in Fig. 22 for radiation at 580QA. Since we observed with a bandwidth of 30A, it was difficult to separate the relative contribution of the \(5^2P_{3/2}-4^2P_{3/2}\) and \(7^2S_{1/2}-4^2P_{3/2}\) transitions, which have
emission wavelengths at 5812A and 5801A, respectively.

The measured signals at 5800A have been time synchronized with respect to the enhanced emission at 6911A. Both measurements were made simultaneously, using the two photomultipliers. It is interesting to note that for high current (2 Amps), both signals reach peak almost simultaneously. This indicates that for high electron densities (here we identify high discharge current with high electron density) excitation transfer is extremely fast. For intermediate currents (0.750 Amps) we can clearly see a difference between the time for peak of both signals, which indicates a slower energy transfer process. For low currents (0.3 Amps) the noise in the upper signal and its small amplitude precludes any observation in this respect.

We can clearly see from this sequence, the lengthening of both signals decay time as the current decreases. The amplitude of the signal at 5800A keeps decreasing with decreasing discharge current, until it finally vanishes. This behaviour can be interpreted by considering that the excitation energy transfer to these highly excited levels becomes more and more ineffective as the free electron density decreases.

In general, the magnitude of the signal at 5800A and other observed wavelengths (except for 6911A and 6939A) was no more than a factor of 2-3 above the background. Finally, we attempted to determine whether atom-atom quenching collisions played a significant role in the observed decay characteristics of the enhanced emission at 6911A. This was done by measuring the plasma signals for different discharge currents, while keeping the potassium vapour pressure constant in the plasma source. We obtained in this way signal decay times ranging from 50 nsec, down to the instrumental resolution time of 10 nsec. This confirms as expected that electron-atom collisions play the dominant role in the quenching of the laser excited transition. The relative contribution of atom-atom collisions for low electron densities could not be resolved experimentally.

The conclusions of this aspect of the experiment will be summarized as follows. The characteristics of the plasma enhanced emission at different wavelengths depend strongly on the free electron density. In particular, the decay time of the plasma enhanced emission at 6911A decreases with increasing electron density. These results support the view, at least in principle that this type of interaction can be used for diagnostic purposes, as suggested by Measures.

5.3 Experiments Designed to Evaluate the Role of the Laser Parameters

We have studied experimentally the dependence of the enhanced plasma signal peak, and of the signal decay time, at 6911A, on the following laser parameters:

(a) Laser power
(b) Laser wavelength
(c) Laser temporal pulse width

The results are summarized next.
(a) Results Concerning Power Dependence

The experiment was done by gradually attenuating the laser beam intensity using a series of aqueous solutions of copper sulfate (CuSO\textsubscript{4}), contained in a 1 cm absorption cell. The solutions were calibrated for transmission at 6939 Å. The accuracy of calibration is about 20-30%. In this way, we varied the laser power density between about $10^7$ W/cm\textsuperscript{2} and $10^8$ W/cm\textsuperscript{2}. Measurements were not continued below this power due to the difficulty of tuning the laser; in fact, tuning becomes quite critical at low powers.

The results are indicated in Fig. 24. For this experiment, we selected only those laser shots whose wavelength was coincident with the potassium absorption line to within the experimental error. The accuracy of this wavelength measurement was estimated to be ± 0.01 Å. The interferometer free spectral range was 0.25 Å. In this way, we ruled out any possible effects of laser wavelength mismatch, on the measured signals. The results indicate a logarithmic dependence of the peak signal on laser power, while the decay time is practically independent of laser power, at least for the conditions considered, where $\tau_d \sim 40$ nsec. This is illustrated in Fig. 25. In fact, measurements performed for short $\tau_d$ values (10-15 nsec) do indicate some dependence on laser power. The interpretation of these results will be given in Chap. 6.

(b) Results Concerning Wavelength Dependence

In this case we kept the laser power constant and arbitrarily mismatched the laser wavelength. The resulting plasma signal peak amplitudes and decay times were measured. The peak signal amplitudes were corrected for laser peak power fluctuations using the results obtained in the previous section. In order to avoid mixing of orders when interpreting the Fabry-Perot interferograms, we used a free spectral range of 0.6 Å, except for measurements near the absorption line center, where a value of 0.3 Å was used.

While making these measurements we monitored periodically the plasma parameters with the Langmuir probe to make sure that the measurements were made under identical plasma conditions. This was also done in the case of the power dependence measurement described earlier. This is also the reason why we operated in a low current mode discharge, since the plasma conditions were quite stable. Correspondingly, the enhanced emission decay time is long. The results of the wavelength mismatch experiment are shown in Fig. 26 for two different laser powers. As already anticipated, the laser tuning requirements become more severe at low power. The decay time, in this instance ($\tau_d \sim 50$ nsec) was found to be independent of the laser mismatch as illustrated in Fig. 27. This effect was also anticipated, since as we have seen in the theoretical discussion (see Eq. (14), Section 2.3.1), the effects of laser power and laser wavelength mismatch are equivalent. For short decay times, this is no longer true. This is illustrated in Fig. 28.

(c) Results Concerning the Laser Pulse Width

Finally, we changed the laser pulse width by changing the concentration of the Q-switch solution. We have compared the resulting plasma signals, under identical plasma conditions, for a laser pulse width of 9 nsec and a pulse width of 23 nsec (FWHM). The results are indicated in
The characteristics of both signals are markedly different. The long laser pulse, although its power is lower, produces a signal with a longer decay time, while its amplitude in the leading edge portion is also larger.

As we shall discuss in the next chapter, all these experimental results can be readily interpreted in terms of a spatial averaging of emission from different volume elements of the plasma, resulting from the laser spatial and temporal intensity inhomogeneity and the non-linearity of the interaction process.

5.4 Experiments for Quantitative Correlation of the Plasma Enhanced Emission Signal Characteristics with the Plasma Parameters

The purpose of this "calibration experiment" was to relate the peak signal amplitude and decay time, with the actual local plasma conditions at the volume of interest measured independently with the Langmuir probe. The Langmuir probe measurements deserve detailed description. This is done in Appendix D.

At this point, we shall only note that we experienced severe experimental problems in performing the probe measurements. This is attributed mainly to potassium contamination effects on the probe insulator. As a result, there is some doubt regarding the accuracy of these probe measurements. In particular, the electron temperatures calculated from probe data are considerably higher than temperatures calculated spectroscopically from the radiative recombination continuum (see Appendix E). In fact, probe calculated temperatures (4000-6000K) are not consistent with the measured degree of ionization (1-10%), assuming that the plasma is in collisional-radiative equilibrium. The temperatures measured spectroscopically (~2000K) seem to be closer approximation, although, we should keep in mind that they represent spatially averaged values.

Regarding the probe measurements of the electron density, we have no additional evidence to corroborate or disprove the accuracy of the results. The fact that temperature measurements seem to be in error does not necessarily mean that the density measurements should be incorrect. Accordingly, we shall accept the electron densities indicated by the probe as representative of the plasma density, although, subject to the condition that results should be interpreted with caution.

Regarding the electron temperature, we shall make the assumption that its value is compatible with the measured degree of ionization, assuming the plasma is in collisional-radiative equilibrium. We would expect temperatures of the order of 2500-3000K.

Although the lack of precise knowledge of the absolute plasma conditions affects the quantitative aspect of the experiment we feel that the qualitative indications of this work form a useful contribution to the field since to date there has been no other experiments of this nature. Indeed, as will be seen from the discussion of the results, the physics of this resonant laser-plasma interaction is rather complex and under the present experimental conditions the information obtained is not of the simple nature envisioned at the start of this project. It is clear now, that the main contribution of these experiments has been to improve our understanding of the basic interaction process, for which, exact knowledge of the plasma conditions is not an absolute requirement.

In the experiment to be described in this section, we varied the plasma conditions by controlling the discharge current. For each condition, measurements
were made of the plasma enhanced emission signal and of the corresponding probe characteristic. These two measurements were made one immediately after the other, in order to avoid any changes in the plasma conditions. The laser peak power density was about $10^5 \text{W/cm}^2$. The measured signal decay time was correlated with the measured electron densities. The results are illustrated in Fig. 30. No attempt was made to correlate the signal peak amplitude to the local electron temperature, for the reasons indicated above. Moreover, the window darkening problems discussed in section 5.1 prevented the use of the photomultiplier calibrated for absolute light intensities.

It is clear from the experience gained in these experiments, that temperature measurements based on the absolute value of the signal peak amplitude require precise control of the laser wavelength and to a lesser extent, the power output, in order to avoid the fluctuations of the signal peak associated with detuning and power variations, indicated in Figs. 24 and 26.
CHAPTER 6

6. DISCUSSION OF EXPERIMENTAL RESULTS

6.1 General Interpretation of Experimental Results

As we have already discussed in Section 2.4, the nonlinearity of the excitation process, and the temporal and spatial characteristics of the laser intensity, add to produce a considerable inhomogeneity of the enhanced emission coefficient in the volume of the plasma excited by the laser. The physical situation was illustrated schematically in Fig. 7.

Consequently, the signals observed experimentally result from the spatial averaging of the enhanced spontaneous emission, from different points of the excited volume that lie within the field of view of the photomultiplier. The temporal characteristics of the signals will depend strongly on the laser power, as well as on the beam spatial and temporal characteristics.

It is important to realize, that the averaging effect discussed herein is inherent to any nonlinear absorber excited by a laser with an inhomogeneous intensity distribution. Consequently, care should be exercised in analyzing results of experiments of this nature. High resolution spectroscopy based on saturation effects is also likely to be prone to these effects.

We shall discuss here the interpretation of the results in terms of an average of local signals. The temporal characteristics of these local signals are determined by the laser beam's local power, and also by the local plasma conditions. Thus, the behaviour in time of these signals can be obtained from a computer simulation program, as discussed earlier in Section 2.2. Accordingly, assuming radial symmetry, one can easily calculate the temporal behaviour of the plasma enhanced emission coefficient at different radii, on the basis of the computer calculations of Fournier (assuming we know $N_e$, $T_e$ at each radius) and of the measured spatial and temporal laser intensity profiles. Since the present computer program does not provide a convenient time correlation between the local plasma emission signal and the laser pulse, for powers above $10^5 \text{W/cm}^2$ we have used the results derived from Equation (14) of Section 2.3.1 and illustrated by Fig. 4. Proceeding as indicated above, we have obtained the results illustrated in Figs. 31, 32 and 33 for a laser peak power density of $10^6 \text{W/cm}^2$. Perfect wavelength matching and uniform plasma conditions characterized by $T_e = 2500 \text{K}$, and $N_e = 10^{13}$, $2 \times 10^{12}$ and $5 \times 10^{11} \text{cm}^{-3}$ respectively were assumed. From these results, one can immediately calculate the radial distribution of the plasma enhanced emission coefficient for any given instant of time. This is illustrated in Figs. 34 and 35 where we have represented the dynamics of the evolution of the radial profile of the enhanced emission coefficient, for a laser peak power of $10^7 \text{W/cm}^2$, zero wavelength mismatch, and $N_e = 2 \times 10^{12} \text{cm}^{-3}$, $T_e = 2500 \text{K}$. We neglect in this calculation, the possible perturbation effects associated with such a high laser power density. These effects will be discussed in Section 6.2. In addition, we are assuming that the plasma conditions are essentially uniform in the volume excited by the laser. Although this is a reasonable assumption for low powers, where the excited volume is small compared to the total plasma volume, it is questionable for a high power of the order of $10^7 \text{W/cm}^2$, where the radius of the volume excited to saturation is estimated to be about 10 mm. Unfortunately, we do not have experimental information about the plasma uniformity.
over such a large characteristic length.

It should be realized that only non-uniformity of the plasma along the line of sight is of concern, since the cross section of plasma observed, perpendicular to the direction of observation is small. Thus, the problem is not as severe as in the case of full three-dimensional inhomogeneities.

Furthermore, the results calculated in this chapter, assuming a uniform plasma, seem to agree closely with the experimental results.

In view of these arguments, we shall neglect the consideration of plasma inhomogeneities in the following discussion, and assume that reasonably uniform conditions prevail in the volume of plasma under observation, prior to laser excitation.

For a particular time $t_1$, the signal $S(t_1)$ produced by the photomultiplier is proportional to the integral (47):

$$S(t_1) = 4 \int_0^x dx \int_x^R \frac{J^*(r, t_1) r \, dr}{(r^2 - x^2)^{1/2}}$$

where $J^*(r, t_1)$ is the plasma enhanced spontaneous emission coefficient at the radius $r$, $R$ being the radius for which $J^* = J$ (the unperturbed emission coefficient), and $2x_0$ is equal to the height of the observed plasma volume. For simplicity, the proportionality constant has been taken equal to unity, since we are interested in the normalized signals. This has the advantage that no absolute calibration factors are required, since we deal with relative magnitudes only. The geometry of the problem is illustrated in Fig. 36.

Using the results illustrated in Figs. 31 to 35 or the equivalent ones for lower laser powers, we have calculated numerically the integral (20) for a number of cases of practical interest.

We shall use these calculated results, in order to illustrate the discussion of the experimental results described in the previous chapter.

6.1.1 Discussion of the Role of the Laser Power

Figure 37 shows the plasma enhanced emission signal normalized to its maximum amplitude and calculated as indicated in the previous section for different laser powers. We have assumed $Ne = 2 \times 10^{12} \, \text{cm}^{-3}$, and $Te = 2500 \, \text{K}$.

The main features of the calculated signals agree closely with those observed experimentally, as discussed below.

(a) Excitation of the plasma with high power ($>10^5 \, \text{W cm}^{-2}$) pulses produces signals with a long risetime ($\sim 100 \, \text{nsec}$) while for low powers (or high power pulses that are detuned) the signals exhibit a fast risetime, of the order of 20 nsec. This behaviour is clearly illustrated by Fig. 37 and by the experimental results presented in Figs. 39 and 40.

(b) The decay time of the calculated signals depends on the laser power.
Nevertheless, this dependence is quite weak, since for the case illustrated, $\tau_d$ changes only by about 25% for a change in power of 4 orders of magnitude. For short decay times, we would expect a larger percentual change in $\tau_d$, for the same range of power variation. These conclusions, obtained from the analysis of the calculated results, are consistent with the results obtained experimentally, i.e., when $\tau_d$ was large ($\tau_d \sim 40-50$ nsec), its value remained approximately constant, within experimental uncertainties, for laser powers between $10^4$-10$^7$W/cm$^2$ (see Fig. 25). Nevertheless, for small values of $\tau_d$ ($\tau_d \sim 10-15$ nsec), the decay time was observed to increase with laser power. The same arguments can be used in discussing the effects of wavelength mismatch.

The increase of the signal decay time with the laser power is a direct consequence of the laser radiation exceeding the saturation threshold for a longer period of time. The central core of the plasma region excited by the higher laser power will thus radiate over a longer period and lead to an increase of $\tau_d$. The same physical argument can be applied to explain the signal leading edge characteristics for different laser powers.

(c) The observed peak of the enhanced plasma emission signal (at 6914A or 6939A) and that of the laser pulse always appear to coincide. Calculated and experimental results are again consistent on this point. This behaviour is simply a result of the fact that the excitation volume is a maximum at the time the laser intensity reaches its peak amplitude.

(d) The signal leading edge characteristics depend mainly on laser power, while the signal decay characteristics depend mainly on the free electron density. The point is clearly illustrated in Fig. 37. This effect is particularly striking for high electron concentrations, where decay times can be very fast while the signal risetime can be considerably slower.

(e) The enhanced emission signal decay time and risetime were found to be quite sensitive to changes of the laser pulse temporal width. A long laser pulse should produce a correspondingly "wider" plasma emission pulse. This is consistent with the results shown in Fig. 29.

We have calculated the plasma enhanced emission peak signals corresponding to different laser peak powers for perfect wavelength matching.

The results are illustrated in Fig. 24, where they are compared to the equivalent experimental results described in Section 5.3a. The calculated values were normalized to the peak amplitude corresponding to $10^5$W/cm$^2$ and fitted to the experimental points. The agreement between the results is good, and the fact that the calculated and experimental values depart from the straight line for powers of $10^7$W/cm$^2$ is particularly encouraging. The logarithmic dependence of the plasma peak signal on laser power, can be easily understood by considering that the square of the radius of the laser beam, which determines the volume of plasma excited depends on the logarithm of the laser power, for a gaussian beam. If we neglect the inhomogeneity of $J^*(r)$, and assume $J^*(r) \propto$ constant over the excitation volume, then:

$$S_{\text{peak}} \propto r_{\text{th}}^2$$

(21)
and

\[ r_{th}^2 = 6^2 \log_e \left( \frac{I_0}{I_{th}} \right) \]  

(22)

for \( r_{th} < x \) (see Fig. 36) where \( S^*_{peak} \) represents the signal peak amplitude, \( r_{th} \) is the radius of the laser beam associated with the threshold power density, \( I_{th} \approx 50 \text{ W/cm}^2 \), and \( I_0 \) is the actual laser peak power density. 6 is the gaussian 1/e attenuation radius. Although the assumption \( J^*(r) = \text{const.} \) is not strictly correct, it is at least a reasonable first approximation that enables us to illustrate in a simple, qualitative way, the role of the laser spatial intensity profile in the determination of the signal peak amplitude.

The situation represented by Eq.(22) is likely to hold for powers up to \( 10^5 \text{ W/cm}^2 \), for which \( r_{th} \approx x \) (see Fig. 14). For higher powers, the departure of the laser beam intensity profile from a pure gaussian distribution, presumably due to light scattering from the laser input window, produces an anomalous increase of the excitation volume, which is responsible for the sudden increase of the signal peak amplitude. This increase is compensated to some extent, at least up to powers of \( 10^6 \text{ W/cm}^2 \), by the "truncation" of the volume of observation by the two horizontal planes separated by \( 2x \). This reduces the square law dependence of the excitation volume on the beam radius, to a nearly linear dependence.

Finally, for very low laser powers, the assumption of \( J^*(r) = \text{constant} \) breaks down, and again we have a departure from the behaviour represented by Eq. (22).

The full power dependence curve, for perfect wavelength matching, has been calculated, as discussed in Section 6.1 and it is illustrated in Fig. 38. Here again, results are for \( N_e = 2 \times 10^{12} \text{ cm}^{-3} \), \( T_e = 2500 \text{ K} \). The peak plasma enhanced emission signals have been normalized here with respect to the signal corresponding to a power of \( 6 \times 10^6 \text{ W/cm}^2 \) (150 kw). These results are discussed in the next section.

6.1.2 Discussion of the Role of Laser Wavelength Mismatch

Equation (14) of Section 2.3.1 indicates that changes in the laser peak power \( I_0(0) \), or changes in the absorption line shape factor \( L_v \) (produced by laser mismatch), will have exactly the same effect on the degree of enhancement of the plasma emission, provided the product \( I_0(0) L_v \) remains constant.

The calculated results indicated in Fig. 38, which represent the dependence of the enhanced emission peak amplitude \( S^*_{peak} \) on laser peak power \( I_0(0) \) for perfect wavelength matching \( (L_v = L_v(0)) \), can also be used to study the dependence of \( S^*_{peak} \) on the laser wavelength mismatch. For this purpose, it is useful to define an effective laser power \( I_{eff}(0) = I_0(0) L_v/L_v(0) \).

If \( I_0(0) = 6 \times 10^6 \text{ W/cm}^2 \) = constant, and we allow \( L_v \) to vary, then Fig. 38 represents the plot of \( S^*_{peak} \) vs. \( L_v/L_v(0) \) for \( I_0(0) = 6 \times 10^6 \text{ W/cm}^2 \) (150 kw peak power). Using the calculated absorption line shape (Appendix B), we can
relate $L/L_0$ with the laser wavelength mismatch $\Delta \lambda$. Knowing the relationship between $L/L_0$ and $\Delta \lambda$, we can immediately calculate $S^\text{peak}_{\text{peak}}$ vs. $\Delta \lambda$, using Fig. 38.

We have performed this calculation for laser peak powers of $6 \times 10^6$ W/cm$^2$ and $4 \times 10^4$ W/cm$^2$. Results are indicated in Fig. 26, where we include the experimental results obtained for the same laser powers and $N_e \sim 10^{12}$ cm$^{-3}$, $T_e \sim 2500^\circ$K.

Agreement between the calculated and experimental results is quite good, considering the complexity of the laser-plasma interaction, and the limitations of the physical model used. We should point out that the absorption line shape is likely to be distorted as a result of the radiative resonant perturbation by the laser, at high powers. This will be discussed in section 6.2.6. The perturbation is not expected to affect our results for the condition of perfect resonance, since saturation occurs at low laser powers. For large mismatch, saturation takes place at high powers, and some decrease of $L_v$ is expected under these conditions. The value of $L_v$ at the saturation threshold will depend on the laser power density, wavelength mismatch and on the characteristics of the original absorption profile. This dependence is complex (49), and it has not been calculated in this thesis. Nevertheless, the fact that our experimental data agrees closely with the results based on a simple model that does not include this perturbation effect seems to indicate that the net effect of the perturbation is not too severe.

The results discussed here illustrate the interesting possibility of measuring an absorption line profile $L/L_0$ on the basis of the experimental measurement of $S^\text{peak}_{\text{peak}}$ vs. $I_0(\text{o})$ for $\Delta \lambda = 0$, followed by the measurement of $S^\text{peak}_{\text{peak}}$ vs. $\Delta \lambda$ for a constant $I_0(\text{o})$, where $\Delta \lambda$ is the laser wavelength mismatch.

6.1.3 Discussion of the Dependence of the Signal Decay Time of the Plasma Free Electron Density

The experimental results described in Section 5.4 indicate that the free electron density plays a dominant role in determining the signal decay time.

Under our present experimental conditions, this role tends to be less dominant and obscured by the spatial averaging effect discussed in Section 6.1. Nevertheless, the fact that we are still able to produce changes of the signal decay time by a factor of five, is quite encouraging from the diagnostics point of view. This is even more so, if we consider that the troublesome spatial inhomogeneity of the plasma enhanced emission can be eliminated by proper design of the experiment, on the basis of the experience gained herein.

Using the approach indicated in Section 6.1, we have calculated the decay time, $\tau_d$, of the signals, assuming an electron temperature $T_e = 2500^\circ$K, and for three different conditions of the free electron density: $N_e = 10^{13}$, $2 \times 10^{12}$ and $5 \times 10^{11}$ cm$^{-3}$.

The results are indicated in Fig. 30, together with the experimental results. The dotted line corresponds to the decay times that would be measured if the averaging effect was absent. These values were obtained by measuring the decay time from a computer plot, for a laser power density of 50 W/cm$^2$, corresponding to the excitation threshold. For such a power level the averaging
effect disappears, in the sense that the enhanced emission from all points of the excited volume will decay simultaneously.

Agreement between the experimental and calculated results is good for electron densities of the order of $10^{12}$ cm$^{-3}$. The fact that the experimental decay time for $N_e > 10^{12}$ cm$^{-3}$ is shorter than calculated, can be explained in terms of a possible electron temperature increase due to superelastic electron collisions with laser excited atoms. This possibility was discussed in Section 2.4.

The importance of the spatial averaging effect in determining the observed decay time is clear from Fig. 30. This illustrates the strong dependence of $\tau_d$ on the laser temporal and spatial characteristics.

Finally, as a complement to the results of this section, we have compared the calculated and experimental plasma emission signal for conditions $T_e \sim 2500$ K, $N_e \sim 10^{12}$ cm$^{-3}$ in both cases, for two different laser powers. The results are indicated in Fig. 39 and 40. It is seen that remarkably close agreement is obtained. The slightly sharper leading edge of the experimental profile can be explained in terms of Fig. 13, where it is seen that the experimental laser pulse has a sharper leading edge than the exponential approximation used in the calculation. The small discrepancy in the tail of the experimental and calculated profiles (for $t > 50$ nsec) is attributed to a similar difference between the real and the assumed laser pulse decay characteristics.

6.2 Discussion of Non-Ideal Effects

In the previous section, we have presented a model for interpretation of the results obtained in the experiments. We have shown that on the basis of the theoretical results obtained by Fournier and of the measured temporal and spatial characteristics of the laser, it is possible to account for all of the basic features of the results obtained experimentally. Furthermore, the quantitative agreement between the experimental and calculated results is good.

Nevertheless, we have neglected to consider a number of effects which might have had some influence under our experimental conditions. It is the purpose of the present section to analyze briefly those non-ideal effects, and attempt to clarify the situation.

6.2.1 Realistic Dynamics of the Laser Pulse Evolution

A comprehensive theoretical analysis of the generation dynamics of a fast Q-switched (i.e., using a pockels cell) laser has been performed by Letokhov and Suchov(50). Their results have been confirmed in a qualitative fashion, by the experimental work done by Korobkin, et al(51) (52) for lasers with active and passive Q-switches, and also by the work of Ambartsumyan et al(53).

From these results we may expect the following performance for the
output of our passive Q-switched laser:

(a) The laser emission does not take place simultaneously over the full emitting face of the ruby, but rather individual regions emit at slightly different times. The emission begins on the axis of the crystal and propagates towards the periphery of the rod, in a time of the order of the total laser pulse duration. These localized bursts of emission are shorter than the total, integrated laser pulse.

(b) The laser beam divergence changes considerably during the pulse evolution. Korobkin and co-workers (51) have measured the time resolved beam divergence of a passively Q-switched laser during a period of \( \sim 20 \) nsec corresponding to peak emission. According to their results, the laser beam divergence changes by more than one order of magnitude in that time period.

(c) The laser emission wavelength has been observed (51), (54) to shift towards the blue, in the period of maximum power output. Shifts are small, of the order of 0.006 - 0.007 Å.

The situation is aggravated if we consider that for our experiment, we are concerned about the laser output characteristics not only at the time of peak emission, but also in both the leading edge and the tail of the laser temporal profile. For completeness we would require to know all the relevant information in a period of 150 - 200 nsec, centered at the time of peak laser emission. Although some work has been done (55), (56) which indicates that it is feasible to obtain this information experimentally, a study of this nature is a major project by itself and beyond the purpose of our thesis.

Since we are using a focussed laser beam, it is expected that the temporal irregularities of the laser emission, due to contributions of different regions of the ruby rod, will be averaged out. This applies also to the emission irregularities associated with the filamentary nature of the ruby laser radiation (57). Thus, we expect that the temporal characteristics of the laser radiation at any point in the plasma will be nearly the same and equal to the measured temporal variation of the laser pulse. The reported laser frequency shifts are quite small, and in first approximation it seems reasonable to neglect their effects.

However, the effects of the variation of the laser beam divergence can be quite important, in view of the relevance of the spatial shape of the laser beam in our experiment.

In all our calculations, we have used the laser intensity distribution at the time of peak, which was measured experimentally. If now we consider that this intensity distribution can vary with time during the evolution of the Q-switched laser pulse, an analysis of the laser-plasma interaction process would become extremely difficult.

Theoretical and experimental results indicate that the beam divergence is a minimum near to the time of peak emission, and consequently, the measured
intensity distribution should correspond to this condition. For other times, the increase of the laser beam divergence over the value attained at peak power, leads to two opposing effects in our experiment.

On the one hand, the power density decreases for small radius, near to the beam centre, thus leading to a time delay during the initial process of saturation of the laser excited transition. During the enhanced emission decay period, it reduces the time for the laser power to drop below saturation threshold. Both effects tend to produce a narrower signal. On the other hand, due to increased beam divergence, the power density in the wings of the laser intensity profile will be larger and consequently, for large radius, the effect is just the opposite of the one discussed above. Although it is not possible to estimate which contribution will dominate in our case, the opposing nature of the effects associated with the variation of the laser beam divergence leads us to expect that this parameter will only play a secondary role in determining the shape of the convoluted signals. This seems to be confirmed by the good agreement between our experimental and calculated data.

6.2.2 Plasma Ionization Enhancement

In section 1.1 we discussed Oettinger and Dewey's (19) ideas about ionization enhancement, based on laser excitation of a transition with an upper level above the energy bottleneck for which the level populations are in near Saha equilibrium with the free electrons. Cool and Zukoski (58) have evaluated the approximate bottleneck energy for potassium, in the density range $10^{13} - 10^{15}$ cm$^{-3}$ and electron temperatures of 2000-3000$^\circ$K. Their estimate is that this energy lies between the energies associated with levels $6^2S_1/2$ and $5^2P_{3/2,1/2}$.

In our experiment we not only satisfy Oettinger and Dewey's criterion, of pumping above the bottleneck, but our laser intensity is high enough for the transition to be saturated. This leads to a slightly different situation with respect to the case considered by these authors, since saturation of the transition implies that a balance between absorption and stimulated emission is attained. Under these conditions, a steady state is established where the population fed to the upper level of the pumped transition results mainly from collisional excitation from the ground level (see Section 2.3.3). This latter process is slow for the conditions of interest. The same situation is also likely to apply in the model of Oettinger and Dewey, after the initial population of the pumped level is exhausted.

Assuming a ground level population $N_1$~$10^{15}$ cm$^{-3}$, a free electron density $N_e$~$10^{13}$ cm$^{-3}$, and an excitation rate coefficient $X_{12}$~$10^{-10}$ cm$^3$sec$^{-1}$ corresponding to $T_e$~2500$^\circ$K (26), we can estimate the maximum possible rate of excitation into the upper level of the laser pumped transition:

$$\frac{dN_{upper}}{dt} \sim N_1 N_e X_{12} = 10^{18} \text{ atoms cm}^{-3} \text{sec}^{-1}$$

(23)

Even assuming all these atoms could be ionized by electron inelastic collisions for the time of interest of our experiment ($T$~$10^{-7}$ sec) the perturbation of the free electron density is negligible. Furthermore, the population excited into levels $6^2S_{1/2}$ and $4^2D_{3/2,5/2}$, is not all elevated into the continuum, as just
assumed. Some population will be used to bring the population in the adjacent levels into some kind of pseudo equilibrium. These arguments are supported by the computer calculations of Fournier which indicate no appreciable free electron density modification during the excitation process, for short duration pulses.

Thus, we conclude from this analysis that the mechanism discussed by Oettinger and Dewey is not likely to produce any serious perturbation in the degree of ionization. Nevertheless, the enhanced populations of levels $^6S_1/2$ and $^4D_{3/2}, 5/2$ which result from laser excitation, represent an energy source for the free electrons. As discussed in section 2.4, superelastic collisions can increase appreciably the free electron temperature, which in turn may increase the degree of ionization. This is essentially the mechanism proposed by Measures (22) for the enhancement of the degree of ionization of a plasma, except that he optimized the interaction by considering laser excitation of the resonance transition.

Proper analysis of the problem is difficult, since the temperature perturbation is established over a very small volume, where the perturbed radial temperature profile has large gradients, and electron diffusion must be taken into account. Fortunately, according to the results of Fournier (26), for electron densities below about $2 \times 10^{12}$ cm$^{-3}$, the maximum temperature increase is less than 350$^\circ$K for an initial temperature of 2500$^\circ$K in the time of interest of our experiment (100 nsec).

Consequently, we may conclude that the plasma perturbation effects associated with superelastic electron collisions will be negligible to a first approximation, in our experiments for $N_e < 10^{12}$ cm$^{-3}$. However, for higher electron densities some form of perturbation may in fact be present for the high power (i.e. long effective duration) cases. This seems to be supported by the fact that under these conditions the experimental decay times are observed to be shorter than the values calculated using the computer results. In the near future the computer programme will be modified to account for these perturbation effects, and it may then be possible to obtain good agreement between the theory and the experiment for the high power laser interaction with a dense plasma.

However, it is important to note that the temperature perturbation effect can be minimized considerably by using short laser excitation pulses. Under these conditions, an adequate power can be used to saturate the transition, while keeping the amount of energy transferred to the plasma to a minimum.

Consequently, although under our present experimental conditions the effects of electron heating may be of some concern, it is estimated that this problem does not represent a limitation to the diagnostic technique, when using a laser of proper characteristics.

6.2.3 Free Electron Heating by Inverse Bremsstrahlung

The free electrons can gain considerable amounts of energy from a powerful laser radiation field, by the process of inverse bremsstrahlung (59). In this process, an electron can absorb the energy of a photon provided it is in the field of an atom, such that the momentum is conserved. Inverse
bremsstrahlung is believed to be closely connected to the mechanism of gas breakdown by laser beams (60).

Wright (59) has developed a simple formula which estimates the rate of energy gain \( \frac{dE}{dt} \) per electron, from a laser field having a photon flux \( F \).

He obtains:

\[
\frac{dE}{dt} \sim n \nu N \sigma^{5/2} F
\]

where \( \nu = 1.78 \text{ eV} \) is the photon energy for a ruby laser, \( N \) the neutral number density \( (N \sim 10^{15} \text{ cm}^{-3}) \) and \( \sigma \) is the cross-section for atom-electron elastic collisions averaged over the electron energy distribution \( (\sigma \sim 3 \times 10^{-16} \text{ cm}^2) \) (see Appendix D). \( F \) is the photon flux \( (F \sim 3 \times 10^{25} \text{ cm}^{-2} \text{ sec}^{-1} \) for \( I_o = 10^7 \text{ W/cm}^2 \)).

Thus we obtain:

\[
\frac{dE}{dt} \sim 8 \times 10^6 \text{ eV sec}^{-1}
\]

and for a time \( t \sim 10^{-8} \text{ sec} \) we obtain:

\[
E \sim 0.08 \text{ eV}
\]

This means that a free electron can absorb about 0.08 eV in 10 nsec if the laser power density is \( 10^7 \text{ W/cm}^2 \). The energy gained by the free electron will be lost to some extent by elastic collisions with neutral atoms and by inelastic collisions, mainly excitation collisions to the resonance level. Losses are likely to be very small in the initial time interval considered.

From this example we may conclude that for most conditions in our experiment, where power densities were of the order of \( 10^5 \text{ W/cm}^2 \) this effect should be negligible. Although in a few cases we used powers close to \( 10^7 \text{ W/cm}^2 \), it should be realized that under those circumstances, the plasma volume where perturbations might have been important was less than 5% of the total volume observed. This is based on the measured laser beam spatial profile and on the assumption that for power densities below \( 10^6 \text{ W/cm}^2 \) the electron heating can be totally neglected. Thus, we estimate that free electron heating by inverse bremsstrahlung should not affect our results appreciably: Diffusion of electrons to regions of lower laser power densities should further minimize the effects discussed.

6.2.4 Photoionization by Single and Multiple Photon Absorption

Two possibilities need to be considered in this case. In the first place, the enhanced populations of levels \( 6^2S_{1/2} \) and \( 4^2D_{3/2,5/2} \) may be elevated directly into the continuum by absorption of laser photons. In second place, ionization may occur as a result of three photon absorption from ground state atoms. Let us consider first the direct photoionization of a 4d electron as an example. For this purpose, we estimate the cross-section for photoionization using the hydrogenic approximation formula (61):

\[
\sigma(4d) = \frac{64}{3^{3/2}} \alpha \pi a_o^2 \left( \frac{E_H}{\nu} \right)^3 \frac{Z^4}{n^5} g_n(\nu)
\]

where \( \alpha \) is the fine structure constant \( (\alpha = 7.30 \times 10^{-3}) \), \( E_H = 2m_e \nu^4/h^2 \) is the ionization energy for hydrogen \( (E_H = 13.53 \text{ eV}) \), \( a_o \) is the Bohr radius \( (a_o = 5.29 \times 10^{-10} \text{ cm}) \), \( \nu \) is the laser photon energy \( (\nu = 1.78 \text{ eV}) \), \( n \) is the principal quantum number \( (n=4) \) and \( Z \) is the effective charge acting on the electron \( (Z \approx 1) \). \( g_n(\nu) \) is the Gaunt factor, which we assume of order unity.
We obtain: \[ \sigma(4d) \sim 3 \times 10^{-18} \text{ cm}^2 \]

The percent of atoms photoionized after excitation to the 4d level, in a time interval \( \tau \sim 10\text{ nsec} \) corresponding to laser peak power is:

\[ N_{\text{photoionized}}(\%) \sim 100 \sigma(4d) F \tau \quad (25') \]

where \( F \) is the photon flux (\( F \sim 3 \times 10^{25} \text{ cm}^{-2} \text{ sec}^{-1} \), for \( I = 10^7 \text{ W/cm}^2 \)). Using equation (25'), we conclude that nearly 100% of the atoms excited into level 4d will be photoionized for laser peak powers of \( 10^7 \text{ W/cm}^2 \). Although the photoionization cross-section is just an estimate, it serves to illustrate the fact that under the present conditions, photoionization effects can be important for high laser powers.

We can apply here the same arguments used in discussing the effects of superelastic electron collisions in section 6.2.2. Photoionization from the upper excited levels is not likely to be important for laser powers below about \( 10^6 \text{ W/cm}^2 \). At higher powers, we can expect a transient perturbation at the time when the laser reaches peak power, if our estimate of the photoionization cross-section is correct. The perturbation would have the effect of a transient extra loss for the upper levels, and consequently the enhanced emission would decrease for those regions where the laser power exceeds \( 10^6 \text{ W/cm}^2 \). Since the enhanced population of the \( 6^2S_1/2 \) and \( 4^2D_{5/2,3/2} \) is small compared to the free electron density, the perturbation in the degree of ionization will be negligible. Moreover, in view of the small volume that would be affected, relative to the total volume contributing to the signal, it is estimated that direct photoionization will not affect seriously our interpretation of results.

The possibility of ionization by simultaneous absorption of three laser photons should also be considered. For 1.78 eV photons, the three photon ionization rate for potassium has been calculated by Bebb (62). If we call \( F \) the photon flux in photons cm\(^{-2}\) sec\(^{-1}\), and \( N_1 \) the number of neutral atoms in the ground state, per c.c., then number of ionized atoms per unit time is of the order:

\[ N_{\text{photoionized}} \sim 5 \times 10^{-80} N_1 F^3 \text{[cm}^{-3}\text{sec}^{-1}] \]

For \( N_1 \sim 10^{15} \text{ cm}^{-3} \), \( F \sim 3 \times 10^{25} \text{ cm}^{-2} \text{ sec}^{-1} \) (\( I_0 = 10^7 \text{ W/cm}^2 \)), we obtain:

\[ N_{\text{photoionized}} \sim 10^{12} \text{ atoms cm}^{-3} \text{ sec}^{-1} \]

For the times of interest in this experiment (\( 10^{-8} \text{ sec} \)) the effects of three photon ionization are completely negligible.

### 6.2.5 Laser Beam Self Focusing Effects

Javan and Kelley (63) pointed out the possibility of self focusing of light with frequency near an absorption line, due to the strong dependence of the index of refraction on the intensity of the radiation field near resonance. More recently, Grischkowsky (64) has reported self focusing of a raman-shifted ruby laser beam (\( \lambda = 7665\text{A} \)) by potassium vapour.

The focusing length is given by (63)(64)
\[ Z_f = d \left( \frac{n_0}{160n} \right)^{1/2} \]  

where \( d \) is the beam diameter, \( n_0 \) the refractive index of the unperturbed vapour, and \( n_0n \) is the refractive index change produced by the laser radiation, which can be written as:

\[
\delta n = \frac{\pi N |d_{12}|^4}{(4\pi\varepsilon_0)^2n^2(\omega\nu_0)^2} \times E^2
\]

where \( N \) is the number of absorbers (the number density of atoms in the \( 3/2 \) level, in our case) \( \nu_0 \) the absorption line central frequency, \( \nu \) the excitation frequency, \( E \) the electric field strength of the laser light, \( h \) is Plank's constant, and \( |d_{12}| \) is the matrix element of the dipole moment in the field direction. Equation (27) applies in the wings of the absorption line, where \( \nu \neq \nu_0 \). As indicated by Javan and Kelley, excitation with frequencies above the line frequency lead to focussing, whereas in the opposite case the beam is defocussed. This behaviour has been confirmed to some extent by Grischkowsky.

In our experiment, the focussing length calculated using equation (26) is large \( (Z_f \sim 130 \text{ cm for } I_o = 10^4 \text{ W/cm}^2, \Delta \lambda \sim 0.1 \text{ A, } N \sim 10^{12} \text{cm}^{-3}) \) because of the small number of absorbers. For excitation closer to resonance \( (\Delta \lambda < 0.1 \text{A}) \) damping effects will be important. It is estimated that this will prevent the self-focussing of the beam to any appreciable extent. It should be considered that \( N \sim 10^{12} \text{cm}^{-3} \) only in the plasma region, which has a characteristic length of about 2 cm. Focussing effects in the vapour in the input arm of the plasma source are totally negligible, since there \( N < 10^3 \text{cm}^{-3} \).

Furthermore, because of the dependence of this self-focussing effect on the excitation frequency relative to the absorption frequency, we would expect to have observed its presence by some asymmetries being produced in the laser tuning experiments described in section 5.3b.

In conclusion, we estimate that laser focussing, or defocussing, due to the intensity dependent anomalous dispersion is not likely to be important in our experiment.

6.2.6 Radiative Splitting and Shifts of the Resonant Energy Levels

The absorption of intense resonant radiation by an atomic system may distort the shape of the atomic absorption and emission lines. The perturbation may be interpreted, from the point of view of stationary perturbation theory, in terms of the splitting of the atomic energy levels in resonance with the laser radiation, and subsequent transitions between these sublevels.

The subject has been discussed extensively by Rautian \((49)\) and papers by Autler and Townes \((65)\) and Bonch-Bruevich and co-workers \((47\) and \(66)\) among others. In general, it can be said that the shape of the absorption and spontaneous emission lines will depend on the laser power density, the wavelength mismatch between the laser radiation and the excited transition, and on the decay characteristics of the two levels associated with the atomic transition in the absence of laser radiation. The perturbation of stationary atoms, where
the thermal motion of the radiating and absorbing species is neglected, is the simplest case to consider.

The general physical picture in this instance, for approximately equal lifetimes of the upper and lower levels of the transition, can be described as follows. For perfect resonance the upper and lower levels split into two sublevels each, displaced in opposite directions from the original level by an energy (66)

$$|\Delta E| = \left[ \frac{e^2 h f_{12}}{(4\pi e_0)^2 m c \nu_{12}} \right]^{1/2} I_o^{1/2}$$

(28)

where $f_{12}$ is the oscillator strength of the transition, $I_o$ is the laser power density, $\nu_{12}$ is the frequency of the excited transition and the other parameters are the usual atomic constants.

Thus, we see that the splitting is linearly proportional to the laser electric field strength, and the effect can be interpreted as a linear stark effect, produced by a resonance oscillating electric field (66).

The resultant line shape is symmetrical, and it is composed of a central maximum with two adjacent secondary maxima. These secondary maxima are smaller in amplitude, and have frequencies

$$\nu = \nu_{12} \pm \frac{2|\Delta E|}{h}$$

(29)

respectively.

The analysis of the line profile involves some cumbersome algebraic operations. It has been calculated analytically by Rautian (49) for the relatively simple case of equal level lifetimes and exact resonance, for stationary atoms. He concludes that the peak amplitude of the central maximum of the line shape is reduced by a factor of two as the laser power becomes infinitely large.

For our experimental conditions, when the laser is in perfect resonance with the transition $^4S_{3/2} - ^6S_{1/2}$, the energy perturbation can be written:

$$|\Delta E| = 2.8 \times 10^{-8} \sqrt{I_o} \text{ [eV]}$$

(30)

where $I_o$ is in W/cm$^2$. For the highest powers we used ($10^7$ W/cm$^2$) we obtain

$$|\Delta E| \sim 9 \times 10^{-5} \text{ eV}$$

which is certainly very small from the point of view of perturbation of the general structure of the atomic energy levels of the atom.

The shift of the secondary maxima, in terms of wavelength, is given by

$$\Delta \lambda = 2.2 \times 10^{-4} \sqrt{I_o} \text{ [Å]}$$

(31)

where $I_o$ is in W/cm$^2$. Thus, for powers of the order of $10^7$ W/cm$^2$ we can produce appreciable shifts, of the order of 0.6 Å. Perturbations of this type have been observed experimentally in potassium vapour by Bonch-Bruevich and collaborators (47),
Bradley(67), Kirin et al(68) and Golubev et al(69), although transitions involving only one of the perturbed levels were investigated.

It is expected that under perfect resonance conditions the effects just discussed will not represent a perturbation for the particular process we are interested in. As indicated by equation (30) the energy level splitting is totally negligible for the threshold power density of 50W/cm². Once saturation has been achieved, a decrease of the absorption coefficient due to the perturbation produced by the laser at higher power will not affect the total emission as the saturation effect will have already ensued. Furthermore, since we are looking at a large spectral interval (3A minimum) it is expected that distortion (broadening) of the emission line width will have no effect on the observed signal.

The analysis of the perturbation when the laser emission is not exactly in resonance with the atomic transition is even more complex especially regarding the calculation of the resultant line shape. The general features of the perturbation can, however, be described as follows:

The resultant line shape is asymmetrical with respect to the frequency \( \nu_{12} \). The three maxima discussed earlier shift and change in amplitude depending on the laser detuning and on the laser power. In the limit of large detuning, one of the two original secondary maxima shifts to the central frequency \( \nu_{12} \) and its shape coincides with the unperturbed line shape of the transition, while the remaining maxima vanish.

The corresponding shifts of the atomic energy levels can be interpreted in terms of a quadratic stark effect, in the oscillating field of the laser (65 and 66). The splitting of the levels can be interpreted in terms of laser induced multi-photon transitions (66).

A net effect of the perturbation is a reduction of the absorption coefficient for laser radiation because of the intensity dependent shift of the energy levels. This mechanism has been discussed recently by Zon et al(70) in connection with the possibility of inducing transparency of a resonant absorber by virtue of the intensity dependent "detuning" which results when the system is exposed to strong radiation fields.

Proper quantitative evaluation of the perturbation under these conditions requires calculation of the absorption coefficient of the plasma, which is a function of the mismatch, the laser intensity and the atomic characteristic of the two levels associated with the transitions as mentioned earlier. This problem is discussed by Rautian(49), although it is beyond the scope of this thesis to attempt such a calculation.

We shall indicate in this connection that this problem is likely to affect only our interpretation of the detuning experiments, where for large mismatch we require very high laser powers to saturate the transition. Nevertheless, the fact that our results calculated on the basis of the unperturbed line profile seem to agree closely with the experimental data for large laser mismatch, seems to indicate that the net effect of the perturbation is not too serious.

6.2.7 Laser Pulse Distortion Due to Saturation Effects in the Plasma

A light pulse can suffer a distortion in its temporal and spatial profiles
when it travels in a saturable absorber (71). For this effect to be appreciable, the change of the transmission characteristics corresponding to the unsaturated and saturated condition must be large.

The low-light level absorption coefficient \( k \) for a uniform potassium plasma is given by:

\[
k \approx \frac{h \nu}{\Delta \nu} N B_{\nu} L_{\nu}
\]

where \( N \) represents the number of absorbers, \( n \), \( \Delta \nu \) is the number density of atoms in the \( \frac{3}{2} \) level, \( B_{\nu} \) is the relevant Milne absorption coefficient and \( L_{\nu} \) is the absorption line shape factor.

For our case, considering absorption at 6939A in the \( \frac{3}{2} \) transition, we have

\[
B_{\nu} = 2.28 \times 10^{12} \text{ m}^2 \text{ Joule}^{-1} \text{sec}^{-1},
\]

\[
L_{\nu} = 7.7 \times 10^{-10} \text{ sec},
\]

assuming a doppier broadened absorption linewidth of 0.02A (FWHM) (see Appendix B) and \( N \approx 10^{12} \text{ cm}^{-3} \) (estimated experimental maximum). Thus, we obtain:

\[
k = 0.4 \text{ cm}^{-1}
\]

Consequently, the typical minimum transmission factor to be expected in the low intensity limit at 6939A for a length \( L = 1.0 \text{ cm} \) of plasma (this is the distance the laser beam has to travel through the plasma to reach the central point of the observation volume) is estimated to be about 67%. In view of this relatively small difference (33%) between the saturated and unsaturated transmission coefficient of the absorber, we would expect that pulse distortion effects will be small. Consequently, the temporal and spatial characteristics of the exciting laser pulse are expected to be the same as measured experimentally.

It is interesting to note here that if in fact pulse shaping effects were important, we would expect to observe faster rise and decay times in the plasma enhanced emission signals, with respect to the values calculated. This effect has been observed experimentally, although it is believed to be connected mainly with the difference between the assumed and measured laser temporal pulse shape, as illustrated in Fig. 13.

The estimates made above also indicate that the enhanced emission photon mean free path will be of the order of 2.5 cm. Thus, no re-absorption effects will have to be taken into consideration.

We can conclude this discussion of the non-ideal effects, by indicating that the perturbations considered in the proceeding sections would not impair the application of the diagnostic technique when a short, low power laser pulse with appropriate frequency is used. These conditions, are in general, considered by Measures(8) in his original discussion of the diagnostic application. Finally, we wish to point out that we have neglected to consider any losses of excited atoms by diffusion from the observed volume. This assumption cannot be easily shown to be reasonable, in view of the short interaction time (\( \Delta t \approx 10^{-7} \text{ sec} \)), by considering that the transit time for an atom across the region of excitation \( (L \approx 0.5 \text{ cm}) \) is \( \tau \approx 10^{-5} \text{ sec} \). Collisions would extend this transit time. Furthermore, we have neglected the effects of background gases and molecular potassium, because of the expected small concentration of these species.
6.3 Practical Requirements Regarding the Laser Performance

The experience gained in this work indicates that the laser performance has to be optimized in order to make full use of the capabilities of the selective excitation diagnostic technique.

The laser requirements can be summarized as follows:

a) The laser pulse duration should be sufficiently short that the entire region excited above threshold should saturate within a couple of nanoseconds, where upon the laser power should rapidly drop to a sub-threshold value, so that distortion of the pulse shape due to spatial averaging (see Fig. 7) is negligible. Under these circumstances, the signal decay time should be independent of the detailed laser temporal and spatial profiles.

Furthermore, and most important, the perturbation of the free electron temperature will be minimized, as discussed in section 6.2.2.

b) The laser pulse spatial intensity distribution should be as nearly rectangular as possible over the beam diameter. This would also eliminate the radial inhomogeneity of the plasma emission coefficient, and in particular, that associated with the plasma region which is excited below the saturation threshold (i.e., the wings of the radial profile of the enhanced emission coefficient). Consequently, the peak amplitude of the signal would be determined unambiguously, for the purpose of temperature evaluation. We should note here that the wing contribution to the radial emission coefficient should not affect the signal decay time, under the conditions discussed in (a) above, provided the decay characteristics of the low intensity wings are the same as that of the uniform core region. The computer results support this view.

c) The laser power density should be high enough to saturate the transition over a volume sufficiently large that the enhanced emission signal should be identified clearly above the background. However, the power should also be low enough to avoid undesirable perturbations, as discussed in section 6.2.3 - 6.2.6.

d) The laser tuning should be controlled within close limits. This would reduce the fluctuations associated with the signal peak amplitude indicated in Figs. 5 and 26. Moreover, the laser power associated with the saturation threshold would be quite moderate, and preclude any serious plasma perturbation.

Alternatively, it should be possible to use a laser with uniform emission over a wavelength interval of the order of several times the absorption linewidth. Under these conditions the excitation process would not be critically dependent on rigorous tuning.

e) The laser power output should be as stable as possible. However, in view of the weak dependence of the signal peak amplitude on the laser power as indicated in Fig. 24 this requirement is not too critical. In particular, if a laser beam with a nearly rectangular intensity distribution is used, the effects of power fluctuations would be further reduced.
To illustrate the improvement that can be obtained by fulfilling the requirements discussed above let us consider the same experiment we have performed, but with a laser having a pure exponential form with a rise and fall time constant of $\tau = 0.5$ nsec. We assume that the laser spatial profile is the same, and that we have single frequency emission. We shall assume further that the laser frequency reproducibility is the same as in our experiment.

In this instance, for perfect resonance, we can rewrite equation (14) in the form:

$$\frac{N^*_7(t)}{N^*_7} = 1 - e^{-8.8 \times 10^{-3} I(0)}$$

at $t = 0$ \hspace{1cm} (33)

On the basis of this equation we estimate that 10% saturation would be achieved with about 10 W/cm$^2$, and nearly full saturation at 500 W/cm$^2$. Thus, assuming a peak power of $10^4$ W/cm$^2$ and a threshold excitation power of about 500 W/cm$^2$, we can calculate the time interval $\Delta \tau$ for which the laser power exceeds the threshold power. During this time, spatial averaging effects may be important. We obtain $\Delta \tau \approx 3$ nsec compared to $\Delta \tau \approx 64$ nsec which we obtain for $I(0) = 10^6$ W/cm$^2$ with our present laser characteristics. This example illustrates that considerable improvement can be obtained by shortening the laser pulse. Moreover, our estimate of a power of $10^4$ W/cm$^2$ can be relaxed considerably by improving on the wavelength reproducibility.

If instead of a single frequency laser we consider a laser with emission over a broad frequency range but again with an extremely short rise time* the threshold power for saturation increases considerably. Let us consider the same example discussed above, but assume that the laser linewidth is 1A. By rewriting the equation that interprets the saturation of the $4^2P_{3/2} - 6^2S_{1/2}$ transition under those circumstances, we obtain 10% saturation at about 500 W/cm$^2$ and full saturation at nearly $3 \times 10^4$ W/cm$^2$. Because of the larger emission linewidth, the tuning requirements are considerably relaxed and we can operate at saturation threshold. Under these conditions, $\Delta \tau$ is nearly the same as in the previous example.

It is also of interest to estimate the minimum size of the volume of plasma that can be excited, in order to obtain a meaningful signal. This would determine the required laser pulse minimum diameter $(r_\text{th})_{\text{min}}$. From our estimates (see Appendix C) a total of $n \sim 2.3 \times 10^{17}$ photons sec$^{-1}$cm$^{-3}$ is emitted by the plasma at 6911A as a result of laser selective excitation at 6939A. Typical optimum experimental conditions can be defined as:

- solid angle of observation: $\Omega = 0.2$ sr;
- optics transmission: $T = 7.8 \times 10^{-2}$,

based on the following loss estimates: 2 windows (8% ea), 1 lens (8%), 1 monochromator (filter) (50%), photocathode loss (80%).

Thus the number of photons absorbed by the photocathode in the detection

* This could be achieved for instance by using a tunable dye laser with a fast external electro-optical shutter.
time $\tau = 10$ nsec is

$$n_{\text{photocathode}} = n V^* \frac{\Omega}{4\pi} T \tau = 3.1 \times 10^6 V^* \text{photons},$$

where $V^*$ is the excitation volume in [cm$^3$]. If we require a signal to noise ratio $S/N = 50$, and assume that the noise is determined principally by photoelectron statistics, we obtain:

$$S/N = (3.1 \times 10^6 V^* \eta)^{1/2}$$

where $\eta$ represents the photocathode quantum efficiency. Recent improvements by RCA in commercial phototubes have raised photocathode quantum efficiencies at 6939A from about 2-3% for an S-20 response trialkali photocathode, to about 26% for the new Quantacon tubes (type C31034A) with a gallium arsenide photocathode. Thus, we shall assume $\eta = 0.26$. In which case the minimum plasma excitation volume necessary to produce the relevant $S/N$ is $V^* \approx 3 \times 10^{-3}$ cm$^3$.

If we consider a cylindrical volume of 0.1 cm length along the beam, the appropriate minimum radius resolved is given by $(r_{\text{th}})_{\text{min}} \approx 0.1$ cm. Thus, the spatial resolution is extremely good.

We conclude from this analysis that by reducing the laser pulse duration to one nanosecond or less, and by improving both the wavelength tunability and the spatial intensity distribution of the laser beam it should be possible to perform local measurements of temperature and electron concentration with extremely good spatial and temporal resolution. This applies particularly in the case of potassium.
CHAPTER 7

7. CONCLUSIONS

7.1 Conclusions

An extensive experimental programme has been conducted to evaluate the application of laser selective excitation spectroscopy to the diagnostics of a potassium plasma.

1. It has been shown that a tunable laser of quite moderate power can be used to excite selectively an atomic transition in a plasma, and produce an enhancement of the spontaneous emission at discrete wavelengths, that can be easily monitored experimentally.

2. It has been shown that the enhanced spontaneous emission contains information about the local plasma conditions. In particular, we have observed that the signal decay time decreases from about 50 nsec to nearly 10 nsec as the free electron density increases from $2 \times 10^{12} \text{cm}^{-3}$ to about $3 \times 10^{13} \text{cm}^{-3}$.

3. Good agreement has been obtained between the experimental and calculated results. On the basis of a computer program and of a simple analytical model, which took into consideration the laser beam spatial and temporal intensity inhomogeneities, we have been able to account to a good degree of approximation for the general observed behaviour of the plasma enhanced emission signals.

4. It has been indicated that a proper laser performance is essential in order to take full advantage of the potentiality of this diagnostic technique. In particular, short laser pulses should be used (less than 1 nsec) of nearly rectangular spatial intensity distribution, in order to overcome the problems associated with the non-linearity of the laser interaction. This would simplify considerably the interpretation of the signals and reduce to a negligible amount the energy gained by the free electrons as a result of superelastic collisions with laser excited atoms. Furthermore, it has been indicated that the laser operation should be restricted to conditions of nearly perfect resonance excitation (i.e., within the half width of the absorption line). This would avoid the fluctuations of the signal peak amplitude associated with the laser detuning, while keeping the laser peak power required to saturate the transition to a low value ($10^2 - 10^3 \text{W/cm}^2$).

5. It has been indicated that for a laser of proper characteristics, the plasma perturbation effects associated with the measuring process should be negligible.

6. It has been indicated that by using a laser of adequate characteristics it should be possible to measure the electron density and excitation temperature, under conditions similar to those of this experiment, with a spatial resolution at about 3 mm$^3$ (a cylindrical plasma element 1 mm long with a 1 mm radius) and a temporal resolution of better than $10^{-7}$ sec. For the particular case of potassium, it is estimated that present day equipment should allow the measurement of $N_e$ from $10^{12} \text{cm}^{-3}$ to about $10^{14} \text{cm}^{-3}$ with an accuracy of 20-30%.

It is believed that the present results, together with the results reported by Burrell and Kunze\(^\text{10}\) constitute the first experimental evidence of
the possibility of using laser selective excitation for performing measurements in plasmas. In particular, Burrell and Kunze concentrated their work in the aspect of the evaluation of excitation transfer cross sections, whereas this work has been concerned mainly with the evaluation of the free electron density, i.e., the plasma diagnostics application.

Although further technical improvements are necessary, as well as quantitative measurements under well diagnosed conditions, it is hoped that a significant contribution has been made by demonstrating the general feasibility of the diagnostic technique, and in particular, the relevance of the laser performance regarding the practical application of the method.

7.2 Potential Advantages of Laser Selective Excitation Spectroscopy for Plasma Diagnostics

Because of its inherent ability to perform measurements with spatial and temporal resolution, the present technique should constitute a valuable complement to the diagnostic methods of Langmuir probes and Thomson scattering. Some potential advantages over these methods are listed below.

1. This diagnostic technique could be applied for localized measurements where high time resolution is required. In particular, the temporal resolution is better than that associated with Langmuir probes (10^-6 sec) by at least one order of magnitude.

2. It could be applied to plasmas of either low or high degree of ionization (in the latter case by selective excitation of ionic transitions). This represents an advantage over Langmuir probes, which are restricted mainly to operation in plasmas of low degree of ionization.

3. Since no material probe is inserted into the plasma, this technique could be used under conditions where the gas temperature is too high for use of a Langmuir probe. In particular, under severe environments such as an alkali plasma, this technique should have a definite advantage over a Langmuir probe. This is illustrated by the difficulties we have experienced in this experiment regarding the probe performance.

4. The fact that a laser of moderate power can be used avoids serious perturbation of the plasma. The low power required compared to that for Thomson scattering is an advantage from this point of view. Furthermore, the problems associated with laser stray light rejection are orders of magnitude less severe than in Thomson scattering experiments, because of the low laser power required and in particular, since alternative decay modes of the upper excited level can be observed. In certain instances these lines can be chosen to be displaced by tens or hundreds of angstroms from the laser excitation wavelength.

5. The determination of the excitation temperature and the electron density can be accomplished by a single measurement. By using a repetitively pulsed laser it should be possible to study the evolution of the plasma parameters under transient conditions at a given point in the plasma or the spatial profiles of the plasma parameters under steady state conditions, by simply scanning the plasma volume with the laser beam.
6. In Thomson scattering, the measurement of low electron densities is seriously impaired due to the small number of scattered photons. The present technique has the advantage that the plasma enhanced emission amplitude does not depend directly on the free electron concentration and thus, the range of measurements can be extended to lower densities.

On the other hand, an inherent difficulty of the present technique is that the interpretation of the results depends on collisional excitation and de-excitation rate coefficients, and their temperature dependence, which in general are not known quite accurately. At low electron densities, radiative transitions and atom-atom collisions may complicate to some extent the analysis of the results.

7.3 Suggestions for Future Work

Although our experiment has illustrated the application of selective excitation diagnostics for the particular case of a potassium plasma, the general ideas should also apply to other elements as well, provided they have an adequate energy level diagram and a tunable laser with proper characteristics is available.

A survey of possible candidates is certainly required. Furthermore, potassium itself is an extremely interesting element, since it can be added as an impurity trace, and it is widely used for seeding in the field of magnetogasdynamic power generation, because of its low ionization potential. Consequently, it would be interesting to study experimentally whether the process we have studied for pure potassium, is affected by the presence of foreign molecules or atoms.

In the case of a potassium seeded plasma a proper calibration experiment, under conditions where the laser characteristics make no contribution to the results, should be performed. In particular, the role of atom-atom and atom-molecule collisions should be investigated in more detail, to determine the relative contribution of these processes on the quenching of the upper level populations, for low electron densities.

As indicated by Burrell and Kunze, the measurement of collisional energy transfer cross-sections is a very interesting possibility, which should certainly be studied in depth experimentally.

In conclusion, as discussed in the introduction of this thesis and as illustrated by the present experiments, laser selective excitation spectroscopy provides a powerful new technique for probing the characteristics and structure of matter. It is expected to see considerable progress in this field in the future.
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APPENDIX A: SATURATION OF A TWO LEVEL ATOMIC SYSTEM BY RESONANT LASER RADIATION

When moderately powerful laser radiation is used to excite selectively an atomic transition in a gas, the transition rates induced by the laser (absorption and stimulated emission) may dominate over collisional and radiative decay rates.

These conditions will likely prevail when:

\[ \int \nu I_{\nu} d\nu \geq 10 \frac{N_e(D_u + D_l)}{B_{lu}} \left( A_u + A_l \right) \tag{A-1} \]

where \( I_{\nu} \) is the laser intensity, \( I_{\nu} \) is the line shape factor, \( N_e \) is the electron density, \( D_u \) and \( D_l \) represent the upper and lower level global depopulation rate coefficients respectively (see section 2.3.2, Eq.(15)). \( A_u, A_l \) are the upper and lower level total spontaneous transition probabilities respectively, and \( B_{lu} \) is the Milne coefficient for absorption.

Under these circumstances, it is possible to a first approximation, to analyze the transient excitation process that leads to saturation of the transition on the basis of a simplified rate equation, where only laser absorption and stimulated emission are considered.

For these conditions, the rate of change (increase) of the upper level population is given by:

\[ \frac{dN^*_u}{dt} = N^*_l B_{lu} \int \nu I_{\nu} d\nu - N^*_u B_{lu} \int \nu I_{\nu} d\nu \tag{A-2} \]

where \( N^*_u, N^*_l \) are the populations of the upper and lower levels respectively. \( B_{lu} \) is the stimulated emission Milne coefficient. The asterisk (*) indicates perturbed values, as opposed to unperturbed values prior to laser excitation.

When the transition saturates, absorption is balanced by stimulated emission and \( dN^*_u/dt = 0 \).

Thus, at the instant of saturation, we have:

\[ N^*_l B_{lu} = N^*_u B_{lu} \tag{A-3} \]

and making use of the fact that

\[ B_{lu} = \frac{\varepsilon_l}{\varepsilon_u} B_{lu} \tag{A-4} \]

where \( \varepsilon_l, \varepsilon_u \) are the lower and upper level degeneracies, we obtain:

\[ \frac{N^*_u}{N^*_l} = \frac{\varepsilon_u}{\varepsilon_l} \tag{A-5} \]

which corresponds to an effective infinite temperature distribution.
When \( g_u \approx g_\ell \), the laser practically equalizes the population of the two levels.

Considering now the conservation equation: (which is applicable to a two level atom, or more generally during the initial redistribution period)

\[
N^u + N^\ell = N_u + N_\ell
\]

where \( N_u, N_\ell \) are the upper and lower populations, prior to laser action, it is easy to show that:

\[
\frac{N^u}{N_u} = \frac{1 + \frac{N_\ell}{N_u}}{1 + \frac{g_\ell}{g_u}}
\]

(A-7)

Thus, the enhancement of the upper level population at the instant of saturation of the transition becomes independent of the laser power, and depends only on the ratio \( N_\ell/N_u \), determined by the conditions in the gas prior to laser excitation.
APPENDIX B: CALCULATION OF THE ABSORPTION LINE PROFILE AT 6939Å FOR NEUTRAL POTASSIUM VAPOUR

The absorption line profile, prior to laser excitation, will be determined by the following broadening mechanisms:

a) Doppler Broadening
b) Resonance Broadening
c) Stark Broadening
d) Natural Broadening

We shall assume the following conditions prevail in the plasma:

gas temperature: \( T_g \approx 700^\circ K \)
vapour pressure: \( P \approx 10^{-2} \) Torr (potassium pod temperature \( 483^\circ K \))
electron temperature: \( T_e \approx 3000^\circ K \)
electron density: \( N_e \approx 10^{12} \) cm\(^{-3}\)

On this basis, we shall consider each contribution independently.

We shall calculate the width between the line centre and the point corresponding to half-maximum intensity. As usual, we shall use the term "half-half width" for this quantity.

a) Doppler Broadening

Doppler broadening results from the thermal motion of the radiating atoms. The doppler half-half width is given by:\(^1\):

\[
\Delta \lambda_D = 3.58 \times 10^{-7} \lambda_o \left( \frac{T}{\mu} \right)^{1/2} \text{ Å}
\]  

where \( \lambda \) is the line centre wavelength in angstroms, \( T \) the gas temperature in K, and \( \mu \) is the atomic weight. For our case, \( \mu = 39 \).

We obtain:

\[
\Delta \lambda_D = 1.05 \times 10^{-2} \text{ Å}
\]

b) Resonance Broadening

Resonance broadening results from the interaction of radiating atoms of the same species, when the upper or lower level of the spectral line have allowed dipole transitions to the ground state \(^2\). For the case of potassium, the \( 4^2P_{3/2} \) level fulfills this requirement. The resonance half-half width is given by Ali and Griem\(^2\) as:

\[
\Delta \lambda_{RES} \approx 3\pi \left( \frac{g_1}{g_2} \right)^{1/2} \frac{e^2 f_{12} \lambda_{12}^3 N}{m_e c^2 (4\pi \varepsilon_o)}
\]

\[ (2) \]
where \( g_1 \): degeneracy of ground level \((g_1 = 2)\)

\( g_2 \): degeneracy of resonance level \((g_2 = 4)\)

\( f_{12} \): oscillator strength of the resonance transition \((f_{12} = 0.682)\) \((3)\)

\( N \): number of neutrals \((N = 1.62 \times 10^{20} \text{ m}^{-3})\)

\( m_e \): electron mass \((m_e = 9.1 \times 10^{-31} \text{ kg})\)

\( \varepsilon_0 \): dielectric constant of vacuo \((4\pi\varepsilon_0 = 1.11 \times 10^{-10} \text{ Farad m}^{-1})\)

\( \lambda_{12} \): resonance wavelength \((\lambda_{12} = 7.665 \times 10^{-7} \text{ m})\)

\( c \): speed of light \((c = 3 \times 10^8 \text{ m sec}^{-1})\)

The values within the brackets correspond to the case of interest and for these values we obtain \( \Delta \lambda_{\text{RES}} = 6.6 \times 10^{-5} \text{ A} \). Recent experimental work by Lewis et al\((4)\) suggests that we should multiply this quantity by a factor of 1.3. Thus, we obtain:

\[
\Delta \lambda_{\text{RES}} = 8.6 \times 10^{-5} \text{ A}
\]

c) **Stark Broadening**

Stark broadening is produced by the electric fields associated with the charged particles of the plasma. The stark half-half width is given by \((1, 5)\):

\[
\Delta \lambda_{\text{STARK}} = \left[ 1 + 1.75 \times 10^{-4} N_e^{1/4} \alpha(1-0.068 N_e^{1/6} T_e^{-1/2}) \right] 10^{-16} w N_e \quad (3)
\]

where: \( \alpha \): ion broadening parameter \((\sim 0.104)\)

\( w \): electron impact half-half width \((\sim 0.45 \text{A})\)

The parameters \( \alpha \) and \( w \) have been calculated for many atoms by Griem\((5)\). However, the values corresponding to the 6939A line of potassium are not available, and so we have taken those corresponding to the transition \( ^4D_{3/2} - ^4P_{3/2} \), at 6964A. Considering that levels \( ^2S_{1/2} \) and \( ^2D_{3/2} \) have practically the same energy, it is estimated that the error associated with the calculated stark width will not be large.

We obtain for the accepted values:

\[
\Delta \lambda_{\text{STARK}} = 4.7 \times 10^{-5} \text{ A}
\]

d) **Natural Broadening**

Natural broadening results from the finite lifetime of the excited states. The natural half-half width is given by:
Here, \( c \) is the speed of light \( (3 \times 10^8 \text{ m sec}^{-1}) \)

\( \lambda \) is the transition wavelength \( (6.939 \times 10^{-7} \text{ m}) \)

\( \tau_u, \tau_e \) are the lifetimes of the upper and lower levels of the transition respectively.

Under our present experimental conditions, the lifetimes of the upper level is determined mainly by electron-atom inelastic collisions. A typical measured value for the conditions of interest is \( \tau_u \sim 4 \times 10^{-8} \text{ sec} \).

The lifetime of the lower level is likely to be long, because of trapping of resonance radiation. The collisional lifetime is equally long.

Thus, for our purposes, only the upper level lifetime is of concern:

We calculate:

\[
\Delta \lambda_{\text{nat}} = 3.2 \times 10^{-5} \text{ A}
\]

From this analysis, we see clearly that the doppler broadening will be dominant. The absorption line shape will thus be a gaussian, with a half-half width of the order of \( 0.01 \lambda^o \), while the profile wings will be Lorentzian, determined by the remaining broadening processes.

**Calculation of the Absorption Profile**

The normalized line shape factor \( L/L' \) of the resultant absorption profile, can be calculated using the formula (6):

\[
\frac{L_V}{L'_{\nu_0}} = \exp \left( -\xi \right)^2 - 2\pi^{-1/2} \left[ 1 - 2\xi F(\xi) \right]
\]

where \( L'_{\nu_0} \) is the shape factor for a line with pure doppler broadening for \( \nu = \nu_0 \) and:

\[
a = \frac{\Delta \lambda_{\text{res}} + \Delta \lambda_{\text{stark}} + \Delta \lambda_{\text{nat}}}{\Delta \lambda_D} (\ln 2)^{1/2}
\]

Here, we have made use of the fact that the line profiles determined by natural, resonance and stark broadening mechanisms will be of the dispersion (Lorentz) type. Thus, the combined shape will also be a dispersion profile, with a half-width equal to the sum of the individual half-widths (5).

\( \xi \) is a variable defined as:

\[
\xi = \frac{\lambda - \lambda_0}{\Delta \lambda_D} (\ln 2)^{1/2}
\]

where \( \lambda_0 \) is the line centre wavelength and \( \lambda \) is the variable. \( F(\xi) \) are functions calculated by Miller and Gordon (7). For our case:
\[ a = 1.3 \times 10^{-2} \]
\[ \xi = 79 (\lambda - \lambda_0) \] (A)

where \( \lambda_0 = 6939 \text{Å} \)

Using Eq. (5) we have calculated the ratio \( \frac{L_v}{L_{vo}} \). The results are indicated in Table B-1, for values of \( \xi \) up to 12. For values of \( \xi > 12 \), a suitable approximation to the line profile is given by Born (6)

\[
\frac{L_v}{L_{vo}} = \frac{a}{\pi^{1/2}(a^2 + \xi^2)} \left(1 + \frac{3}{2\xi^2}\right)
\] (8)

Using this approximation, we have calculated the ratio \( \frac{L_v}{L_{vo}} \) up to \( \xi = 60 \). The results are indicated in Table B-2 and the line profile is illustrated in Fig. B.1.

References

APPENDIX C: CALCULATION OF NUMBER OF PHOTONS EMITTED BY THE PLASMA DUE TO LASER EXCITATION

It is the purpose of this appendix to calculate the maximum number of photons $n$, produced per cm$^3$ per second, as a result of laser excitation.

We shall estimate this number from experimental results, as well as from theoretical considerations.

**Estimation of $n$ from theoretical considerations**

Let us assume the plasma conditions to be:

$$N_e \sim 10^{12} \text{ cm}^{-3}, \quad T_e \sim 2500^\circ \text{K}, \quad N_{\text{neutral}} \sim 5 \times 10^{14} \text{ cm}^{-3}$$

If, in addition, we assume that due to the imprisonment of resonance radiation the ground level and the resonance level of the neutral potassium atom are in collisional equilibrium the ratio of the populations will be given by the Boltzmann distribution corresponding to the temperature of $2500^\circ \text{K}$.

The population of the $4P_3/2, 1/2$ levels is thus estimated to be $N_2^0 \sim 5 \times 10^{11} \text{ cm}^{-3}$. Now the saturation peak population of the $6S_{1/2}$ level is $N_{7\text{ max}}^* \propto N_2^0/9$ as was discussed in section 2.3.1.

Thus, we estimate: $N_{7\text{ max}}^* \sim 5 \times 10^{10} \text{ cm}^{-3}$

In which case the enhanced number of photons radiated spontaneously at 6911A per cm$^3$ of excited plasma, per second, into $4\pi$ steradians, is:

$$n = N_{7\text{ max}}^* A_{72}$$

where $A_{72}$ is the transition probability coefficient ($A_{72} = 2.7 \times 10^6 \text{ sec}^{-1}$).

Here, we have neglected the spatial averaging effect. This will not make a difference larger than a factor of two, as indicated by the results in Figs. 34 and 35. Thus, we estimate: $n \sim 1.3 \times 10^{17} \text{ photons cm}^{-3} \text{ sec}^{-1}$

**Estimation of $n$ from experimental considerations**

For the plasma conditions, $N_e \sim 10^{12} \text{ cm}^{-3}, \quad T_e \sim 2500^\circ \text{K}$, based on our Langmuir probe measurements, excitation of the plasma with a laser peak power of 40KW/cm$^2$ with perfect wavelength matching, creates a peak anode current (in the C70042K photomultiplier) of about $2 \times 10^{-3} \text{ amp}$, at a typical gain of $3 \times 10^3$ (according to manufacturer specifications).

We can thus calculate the photon flux per second reaching the photocathode, corresponding to the maximum plasma emission:

$$n_{\text{photocathode}} t = \frac{i_a}{\eta G_a}$$

where $n_{\text{photocathode}}/t$ represents the number of photons absorbed at the photocathode.
per second, $i$ is the anode peak current, $\eta$ is the photocathode quantum efficiency at 6911Å (about 6%), $e$ is the electron charge and $G$ is the phototube gain ($3 \times 10^5$).

We obtain:

$$ \frac{n_{\text{photocathode}}}{t} = 7 \times 10^{11} \text{photons sec}^{-1} $$

In order to calculate the number of photons $n$ produced at the plasma, we need to estimate the plasma volume contributing to the signal, as well as the solid angle of observation and losses in the optics.

The excited plasma volume is approximately a cube in this case. The area of the cube perpendicular to the line of sight is known (see section 5.1) and it is equal to 0.48 $\times$ 0.50 cm$^2$. The side of the cube in the direction of the line of sight, is of the order of the laser diameter corresponding to threshold power (50 W/cm$^2$) when the laser peak power is $40$ kW/cm$^2$. From Fig.14 we measure this diameter to be about 0.60 cm. Thus we estimate: the plasma excited volume $V_{\text{exc}} \approx 0.14$ cm$^3$.

The remaining factors are estimated as follows: solid angle: $\Omega = 0.057$ sr, optics transmission: $T = 4.5 \times 10^{-3}$.

The losses of the system have been estimated according to the following account: one plasma observation window (8%), one oven window (8%), one focusing lens (8%), one beam splitter (50%), one narrow band interference filter at 6911Å (94%), reflection at the photomultiplier face window (4%), losses due to transmission and reflection at the photocathode (80%).

Under these circumstances

$$ n = \frac{4\pi}{\Omega V_{\text{exc}} T} \frac{n_{\text{photocathode}}}{t} $$

and we obtain: $n = 2.3 \times 10^{17}$ photons cm$^{-3}$ sec$^{-1}$.

The agreement between the experimental and the theoretical values is surprisingly good, considering the coarse estimates used in the calculation.

We can see from the analysis of the losses in the optics that almost one order of magnitude improvement might be achieved by replacing the lossy interference filter by a monochromator or another more suitable filter.

From these results, we can easily estimate the signal photoelectron noise. The number of photons absorbed at the photocathode within the integration time of the instrument $\tau \approx 10$ nsec is: $n_{\text{photocathode}} \sim 7 \times 10^3$ photons.

If the quantum efficiency at 6911Å is 6%, the signal to noise ratio is of the order:

$$ S/N \sim (0.06 n_{\text{photocathode}})^{1/2} = 20.4 $$

Thus the noise in the signal should be about 5% which is in agreement with the noise observed experimentally. Finally, it is useful to estimate the total number of photons, corresponding to the enhanced plasma emission that fall into the solid angle of observation $\Omega$ in a time $\tau \sim 10$ nsec. This number, $n_{\Omega}$ is:
\[ n_\Omega \sim n \frac{\Omega}{4\pi} V^* \tau = 1.7 \times 10^6 \text{ photons} \]

This number is used for evaluation of the plasma source stray laser light rejection ability, in Section 4.2.1.

References


APPENDIX D: LANGMUIR PROBE MEASUREMENTS

D.1 Introduction

Although Langmuir probes are extremely simple devices to construct and to use from a purely practical point of view, interpretation of the probe data can be extremely complex and subject to errors arising from a variety of subtle experimental complications.

In its simplest form, a Langmuir probe is a small electrical conductor, of cylindrical, spherical or plane geometry of a well-defined area. The conductor is held by an insulating support, usually glass or ceramic. When inserted into a plasma and connected to an adequate electrical circuit, this probe will draw a current $i_p$ from the plasma, which will depend on the potential difference between the plasma and the probe, $V$. The functional dependence $i_p$ vs. $V$ is called the probe characteristic and can be recorded using an X-Y recorder or an oscilloscope. The electron temperature and the electron density can be calculated from the probe characteristic.

Reviews of probe theories and general discussion of application techniques and associated errors can be found in a number of standard references (1, 2, 3).

There is in practice a wide range of plasma-probe conditions, that can be characterized in terms of the following parameters:

$$\frac{\lambda_{e-n}}{r_p}, \frac{\lambda_{i-n}}{r_p}, \frac{r_p}{\lambda_D}, \frac{T_e}{T_i}, \chi_p$$

where $\lambda_D$ is the Debye length ($\lambda_D \approx 70 \sqrt{\frac{T_e/N_e}{m_e}}$ in MKSQ units) $r_p$ is the probe radius, $\lambda_{i-n}$ and $\lambda_{e-n}$ are respectively the ion and electron mean free path for elastic collisions with neutrals, $T_i$ and $T_e$ are the ion and electron temperature respectively and $\chi_p = eV_p/kT_e$ is the normalized probe potential.

The range of conditions characterized by $\frac{\lambda_{i-n}}{r_p}, \frac{\lambda_{e-n}}{r_p} \gg 1$ is called the collisionless regime. Probe data can be interpreted under those conditions using the theory of Laframboise(4) which covers a wide range of variation of the additional parameters $r_p/\lambda_D$ ($0 \leq r_p/\lambda_D \leq 100$) and $\chi_p (|\chi_p| \leq 25)$. This condition is typical of very low density gas discharges.

The opposite extreme where $\frac{\lambda_{i-n}}{r_p}, \frac{\lambda_{e-n}}{r_p} \ll 1$ and $\frac{\lambda_{i-n}}{\lambda_D} \ll 1$, $\frac{\lambda_{e-n}}{\lambda_D} \ll 1$ is called the continuum regime. Su and Lam(5) have presented an adequate continuum theory for spherical probes, and Cohen (6) has considered in addition elliptical probes. The case of cylindrical probes has no analytical solution in this regime, but results can be obtained for them as limiting solutions of the case of long, slender elliptical probes.

The continuum regime is typical of flame plasmas and plasmas of interest for magnetogasdynamic power generation.
The intermediate or transition regime is less well documented. A number of theories (7,8,9) and (10) have been presented where the plasma is divided arbitrarily into different regions. In each region a certain model is assumed to calculate the charged particle flow to the probe, and the solutions are matched at the boundaries. The accuracy of the matching procedure is open to question. A few authors (11,12,13,14) have presented general formulations based on a kinetic theory approach that are intended for arbitrary mean free paths and Debye lengths.

Presently, the most complete formulation for the transitional regime is that of Talbot and Chou(15). Their analysis is based on earlier work by Chou, Talbot and Willis(14). Kirchhoff, Peterson and Talbot(16) have conducted recently experimental work, in order to check the theory, with a good degree of success.

D.2 Experimental Complications in Alkali Plasmas

In an alkali plasma, the formation of alkali layers due to adsorption (17) on the probe surface has the effect of lowering the collector work function. Since the degree of contamination and thus the work function, depends on the probe temperature, which varies during the recording of the characteristic, it is possible to have a number of hysteresis effects which distort the results and lead to error. This problem has been adequately illustrated in a paper by Wehner and Medicus(18). The problem can be avoided by using appropriate measuring techniques as discussed by Waymouth(19).

Furthermore, if the probe work function is low, thermionic emission can be important, even at moderately low probe temperatures. This problem usually leads to an overestimation of the electron density when using ion collection data. Consequently, the probe must be kept cold, and as clean as possible.

Finally, materials which are normally insulators will become conductors to some extent, as a result of alkali vapour contamination (20,21,22). This effect does not result simply from the condensation of a metallic layer of alkali (this, of course, will produce surface conduction) which can be easily avoided, but rather from absorption of the alkali by the insulator and further ionc diffusion(22).

Effects of insulator conductivity in probe measurements have been reported by Bullis(23). In this work, we have experienced exactly the same problem discussed by this author, which leads to an overestimation of the electron temperature. Bullis was able to overcome this problem by using a special design for his probe. In our case, we found that by sputtering the probe before measurements, for periods of about five minutes, the effects associated with insulator conductivity would disappear initially.

After about 100 hours of operation, contamination was quite severe and extended periods of cleaning were required, until finally, contamination effects could not be eliminated. All the relevant measurements were conducted during the initial period of operation of the probe.
D.3 Description of Probe, Circuitry and Mode of Operation

We used a cylindrical tungsten probe mounted on a pyrex glass insulator. The probe geometry and dimensions are indicated in Fig. D-1.

The probe was mounted on a bellows assembly driven by a control screw; this allowed the probe to scan radially the central plane of the interelectrode region, and to be removed completely out of the field of view when firing the laser.

The probe, bellows and positioning screw formed a single unit, attached to the main body of the plasma source by means of a Varian Conflat flange with an OHFC copper gasket. Although by design the probe could be replaced if necessary, the problems associated with this operation, as discussed in section 5.1, made the replacement quite impractical. We wish to stress this point in particular, by indicating that we had considerable difficulties in preserving the leak tightness of the plasma source, in the initial stages of this experiment, as well as in keeping the unit in satisfactory operating conditions. These difficulties are inherent to a potassium plasma, in view of the corrosive nature of this element at a temperature of about 350°C.

Consequently, it should be realized that an operation as simple as the replacement of a Langmuir probe under normal circumstances can become involved under the present conditions and further, it implies some risk regarding the leak tightness of the special windows and insulator seals if the cleaning operation mentioned in section 5.1 is not performed properly.

The probe electrical circuit is indicated schematically in Fig. D-2. Connection to this circuit was possible by means of an alkali-resistant electrical feedthrough, provided by Mine Safety Appliances (MSA). This unit featured a single stainless steel pin (1/32" dia.) brazed into a sapphire insulating jacket provided on the outside with an "inverted-hat" type ss-flange for structural mounting.

In view of the contamination problems resulting from the operation of the probe in an alkali-vapour environment, it was necessary to obtain the probe characteristic under pulsed conditions, to ensure a constant probe work function during the measurements. Our experimental approach follows closely the technique suggested by Waymouth. A voltage pulse generator was used to provide sawtooth pulses of variable slope, and amplitude up to 150 V. In this way, the probe characteristic could be recorded in a time interval ranging from 1 sec to 10^{-3} sec. A D.C. bias power supply allowed us to keep the probe under constant ion or electron bombardment between measurements, for cleaning purposes.

The probe trace was displayed in an X-Y oscilloscope and photographed with a polaroid camera.

We found it convenient to clean the probe by sputtering, since this seemed quite effective in reducing the insulator contamination effects. Just before recording the characteristic, the probe tip was heated to an almost orange-white-colour, to eliminate any possible surface contamination layers. The probe was allowed to cool for about five seconds and then it was pulsed positive and the characteristic was recorded.
D.4 Theory and Procedure Used to Reduce Probe Data

Preliminary examination of the charged particles mean free paths indicates that the probe operates in the transitional regime, where $\lambda_i/n_p \sim 1$ (see Section D.6).

Accordingly we have used Talbot and Chou's theory for interpretation of our results.

The ion current density for a cylindrical probe of length $l$ is given by these authors as:

$$J_i = eN_e \left( \frac{kT_e}{2\pi M_i} \right)^{1/2} I_i^* \left( \frac{\lambda_i-n}{r_p}, \frac{T_e}{T_i}, \chi_p, \frac{r_p}{\lambda_D}, \frac{l}{r_p} \right)$$  \hspace{1cm} (D-1)

whereas the electron current density in the electron retarding region is given by:

$$J_e = eN_e \left( \frac{kT_e}{2\pi m_e} \right)^{1/2} e^{-\chi_p} I_e^* \left( \frac{\lambda_{e-n}}{r_p}, \frac{T_e}{T_i}, \chi_p, \frac{r_p}{\lambda_D}, \frac{l}{r_p} \right)$$  \hspace{1cm} (D-2)

where $M_i$ is the ion mass and $m_e$ is the electron mass. The functional dependence of $I_i^*, I_e^*$ on the plasma-probe parameters is given analytically (15,16).

Kirchhoff et al. (16) have plotted $I_i^*$ at ten dimensionless units below the floating potential, as a function of $r_p/\lambda_D$, using $\lambda_i/r_p$ as a parameter. Results are presented only for $T_e/T_i = 1, 10$ and for $l/r_p = 100$. Nevertheless, the dependence of $I_i^*$ on $l/r_p$ is very weak, and to a first approximation it is acceptable to use the results plotted for $l/r_p = 100$.

Electron densities have been determined on the basis of these results, from the ion saturation characteristics. Since for this purpose we require to know $\lambda_D$, we estimated $N_e$ initially from the electron current at the plasma potential, using the standard formula:

$$N_e = \frac{i_e(V_p = 0)}{eA \left( \frac{kT_e}{2\pi m_e} \right)^{1/2} I_e^*}$$  \hspace{1cm} (D-3)

Here, $i_e(V_p = 0)$ is the total electron current at the plasma potential, $A$ is the probe collecting surface, and $I_e^*$ is given by (16):

$$I_e^* = \left\{ \frac{1 + \frac{\lambda_{e-n}}{r_p}}{\ln \left( 1 + \frac{\lambda_{e-n}}{r_p} \right) - 1} \right\}$$  \hspace{1cm} (D-4)
$I_e^*$ can be interpreted as a correction factor which takes into account the fact that the electron flow to the probe is not strictly collisionless. In the standard collisionless theory, $I_e^* = 1$.

The value of $N_e$ obtained from the electron collection data was used to calculate a first approximation to $\lambda_D$, designated $\lambda_D(0)$. With $\lambda_D(0)$, we estimated $I_1^*(\chi_f-10)$ from the results of Kirchhoff et al., and calculated again $N_e$ from ion saturation data:

$$N_e = \frac{1}{e A (k T_e/2 \pi M_1)^{1/2}} \frac{i_i^*(\chi_f-10)}{I_1^*(\chi_f-10)} \quad (D-5)$$

$i_i^*(\chi_f-10)$ is the measured ion current, at ten dimensionless units below the floating potential. With $N_e$, we recalculated $\lambda_D$, designated $\lambda_D(1)$, and iterated to obtain a new value of $N_e$, and so on. The process converges very rapidly.

The electron temperature was determined as usual from the slope of the semi-log plot $\log_i$ vs. $V_p$. In mathematical form,

$$\frac{d(\log_i)}{dV_p} = \frac{1}{2.3 (k T_e/e)} \quad (D-6)$$

This value of the temperature was used to calculate $N_e$. The values of the parameters necessary for evaluation of $I_i^*(\chi_f-10)$ are given in TablesDI andDII. The results obtained are indicated in TableDIII. We note that the ion temperature is taken equal to the atom temperature, which we assume is 7000°K.

D-5 Discussion of Experimental Results

Observation of the experimental results, indicates a large discrepancy between the values of electron density determined from electron and ion collection data.

A number of experimental effects can be responsible for the difference. The electron density can be underestimated, when using electron collection data, due to electron reflection at the probe surface and because of plasma depletion effects. Reduction of the probe collecting area due to melting effects should also be considered.

It is expected that if electron reflection was important, the characteristic should exhibit some peculiarities near and above plasma potential, which were not observed. Plasma depletion effects are important when the probe collects a current which is appreciable compared to the main discharge current, since in this case electron losses at the probe surface start competing with other loss mechanisms in the plasma, with the result that the electron density is lowered near the probe.
In our case, the electron current at the plasma potential was of the order of 5 to 18% of the main discharge current. It is estimated that this may lead to an appreciable plasma perturbation. In fact, in some cases, the main discharge current was observed to decrease when the probe was biased near to the plasma potential.

On the other hand, the ion saturation current may be erroneously high because of electron ejection effects from the probe surface, or effects associated with the increase of the probe effective collecting area.

Secondary electron ejection may be produced by several mechanisms, such as thermionic emission, collisions of metastable atoms with the probe, photoelectric emission due to resonance radiation and kinetic ejection of electrons due to collisions with energetic ions. We have estimated that the effect of mechanisms are likely to be negligible under our experimental conditions.

The increase of the probe collecting area due to the conductivity of the insulator is estimated to be important, unless proper precautions are taken. As mentioned earlier, we were able to produce apparently normal traces after sputtering the probe for several minutes, at voltages of the order of -100 to -200 volts. This is indicated in Fig. D-3 where two traces are shown corresponding to identical probe conditions. In the first case, we recorded the trace with an uncleaned insulator. The second trace was recorded after five minutes of cleaning. Visual glow effects could be observed on the probe insulator when the probe was biased with the main discharge off, indicating that a discharge to the insulator surface was taking place, as illustrated in Fig. D-3. Evidence seems to indicate, as observed also by Bullis (23), that the perturbation is associated mainly with ion collection, whereas the electron part of the characteristic is not affected to any large extent. We shall accept the electron density readings from the ion saturation data as more reliable, in view of the high electron current collected by the probe near to the plasma potential and of the observed perturbation of the main discharge. Nevertheless, these results should be interpreted with some caution, considering the possibility of remnant conduction of the insulator.

The results should be more correct in the sense of relative measurements, since it is expected that errors associated with the insulator contamination will be systematic.

The electron temperature determined from the semi-log plot method seems to be rather high, to be compatible with the degree of ionization that is determined using the measured $N_e$ value and the neutral concentration $N$ (which can be calculated from vapour pressure tables (24), since we know the temperature of the potassium pod). The fact that the electron temperature was also higher than the temperature determined from the spectroscopic measurements (see Appendix E) seems to indicate a general tendency of the probe to overestimate the electron temperature. The actual mechanism responsible for this effect is not clear, although the lowering of the electron density in the vicinity of the probe could be one possible cause, in addition to alkali contamination effects.

Some experiments had been planned, in an attempt to elucidate the problems associated with the probe operation under these conditions. In particular, we were interested in performing detailed measurements using the probe and radiative recombination spectroscopy under different plasma conditions and obtain at least some indication about the reliability of the relative probe measurements.
Unfortunately, the general deterioration of the insulator and a subsequent air leak in the sapphire feedthrough which rendered the plasma source inoperative, prevented these measurements. Consequently, for calculation and comparison purposes, we adopted the criterion of assuming an electron temperature that is compatible with the measured degree of ionization, assuming the plasma is in collisional radiative equilibrium. Using the results of Fournier(36), we have evaluated the corresponding temperature for each case. On this basis, the electron density determined from the ion saturation data has been corrected using equation D-5. The correction does not take into account the small increase of $I_i \left(10\right)$ associated with the new temperature. This should not introduce an error larger than 20% which in view of general uncertainties of the measurement can be neglected to a first approximation.

In view of the problems associated with the Langmuir probe operation, it is difficult to determine the accuracy of these measurements, from an absolute point of view. The measurements should represent, at least, a good order of magnitude estimate of the conditions of the plasma. In fact, our results are consistent with those obtained for similar devices(37). Furthermore, the qualitative evaluation of the laser-plasma interaction does not depend critically on the precise knowledge of the plasma conditions. Consequently, we feel reasonably justified, to first approximation, in using the results indicated in Table III for the analysis of the laser excitation experiment.

D-6 Calculation of Charged Particle Mean Free Paths

(a) Electron-Neutral Mean Free Path

By definition:

$$\lambda_{e-n} = \frac{1}{N_0 Q_{e-n}}$$  \hspace{1cm} (D-7)

where:

$$N_0 \left[ \text{cm}^{-3} \right] = 9.62 \cdot 10^{18} \frac{P}{T} \left[ \text{mmHg} \right]$$

is the neutral number density

$$Q_{e-n} \left( \text{cm}^2 \right)$$

is the total elastic scattering cross-section for electron neutral collisions.

Karule (25) has calculated $Q_{e-n}$ for low energy electrons in a potassium vapour. Recent experimental work by Visconti et al (26) indicates very good agreement between the theory and the measured cross sections.

From the results presented in Ref. (26), we obtain

$$Q_{e-n} \left( 0.25 \text{ ev} \right) = 3.5 \times 10^{-14} \text{ cm}^2$$

The electron-neutral mean free path is:

$$\lambda_{e-n} \left( \text{mm} \right) = \frac{2.1 \times 10^{-2}}{P(\text{Torr})}$$  \hspace{1cm} (D-8)

(b) Calculation of Ion-Neutral Mean Free Path

By definition
\[ \lambda_{i-n} = \frac{1}{\sqrt{2} N_0 Q_{i-n}} \]  

(D-9)

where \( N_0 \) is the same as before and \( Q_{i-n} \) is the corresponding diffusion cross-section for the ions. It is generally accepted that the fundamental interaction mechanism between ions and their parent atoms is charge exchange (27). The diffusion cross-section \( Q_{i-n} \) is related to the change transfer cross-section \( \sigma \) by (27) (28)

\[ Q_{i-n} = 2\sigma \]  

(D-10)

In order to take into account the variations of \( Q_{i-n} \) with ion velocity, one should properly define the averaged cross-section \( \bar{Q}_{i-n} \) over a Maxwellian distribution characterized by the temperature \( T \):

\[ \bar{Q}_{i-n} = \frac{1}{2} \int_{0}^{\infty} x^2 Q_{i-n}(x) \exp(-x) \, dx \]  

(D-11)

where

\[ x = \frac{1}{2} \frac{M_i V_r^2}{kT} = \frac{\text{particles relative energy}}{\text{particles thermal energy}} \]

\( V_r \) is the relative velocity. An alternative procedure (29) is to evaluate \( Q_{i-n} \) for a relative velocity:

Both methods yield practically the same value of \( Q_{i-n} \) in our case. We have evaluated \( \sigma \) for \( V_r = 2.12 \left( \frac{2kT}{M_i} \right)^{1/2} \) using the extrapolated results of Sheldon (30).

It should be pointed out here, that the experimental values of \( \sigma \) obtained from different authors are in large discrepancy, by nearly a factor of two. We have accepted, rather arbitrarily, the results of Kushnir et al (31) reported in Sheldon’s paper. This is based on the fact that these results agree closely with those of Palyukh and Savchin (32) and also rather closely with the theoretical results of Smirnov (33) reported in (32). Thus, for \( T = 700 \, ^\circ \text{K} \), we accept

\[ \sigma = 7.0 \times 10^{14} \, \text{cm}^2 \]

and we obtain

\[ \lambda_{i-n} (\text{mm}) = \frac{5.2 \times 10^{-3}}{P(\text{Torr})} \]

From a conservative point of view, the uncertainty in this result should be a factor of two.

Since the theory of Talbot and Chou (15) is based on an inverse fifth power law for particle encounters, and the mean free path is in fact defined for perfectly elastic hard spheres gas model, these authors suggest that the mean free paths to be used in their theory should in fact be defined as:

\[ \lambda_{i-n} = \frac{3\pi}{4} \lambda_{i-n} \]

\[ \lambda_{e-n} = \frac{3\pi}{8} \lambda_{e-n} \]
where $\lambda_{i-n}$, $\lambda_{e-n}$ are the real mean free paths, calculated in the preceding part.

These mean free paths have been calculated for our experimental conditions, and are shown in Table 1.

(c) Other Mean-Free Paths: $\lambda_{e-e}$, $\lambda_{e-i}$, $\lambda_{i-i}$, $\lambda_i$-

The case of "self-collisions" between charged particles was considered by Spitzer (34), who defined an effective collision mean free path for $90^\circ$ deflection for like particles of temperature $T$ and number density $N$:

$$\lambda = 1.8 \times 10^9 \frac{T^2}{N^nA}$$

where $\Lambda = 1.24 \times 10^7 \sqrt{\frac{T}{N}}$; all quantities given in MKSQ units.

When $M_i \gg m_e$, it is possible to show that

$$\lambda_{e-e} \approx \lambda_{e-i} \text{ and } \lambda_{i-e} \gg \lambda_{i-i}$$

From Sonin's tabulated results (35) we obtain that

$$\left(\frac{\lambda_{e-e}}{r_p}\right)_\text{min} \approx 50$$

for the relevant conditions in our experiment. Thus, the electrons can be assumed in a collisionless regime with respect to encounters with charged particles. For ion-ion collisions the situation is different, since

$$\left(\frac{\lambda_{i-i}}{r_p}\right)_\text{min} \approx 0.05$$

though this does not seem to affect largely the ion collection (16) (35).

REFERENCES


32. B. M. Palyukh, L. S. Savchin, High Temp. 6, 613 (1968).


APPENDIX E: SPECTROSCOPIC TEMPERATURE MEASUREMENTS FROM THE RADIATIVE RECOMBINATION CONTINUUM

Radiative recombination involves free-bound transitions, which give rise to continuum emission. The continuum spectrum is characterized by bands corresponding to recombination into particular levels. Each band has an associated head where the recombination intensity is a maximum. The head is connected with the recombination of the slower electrons having an energy just equal to the binding energy of the particular level.

In the case of potassium, there are two recombination bands in the visible part of the spectrum. They correspond to recombination into the $^2P_{3/2,1/2}$ levels, with a bandhead at approximately 4500Å, and recombination into $^5D_{5/2,3/2}$ levels, with a bandhead at 7300Å.

A typical radiative recombination spectrum of potassium is shown in Fig. E.1. The spectrum illustrated is not corrected for the phototube (S-20) response. For a Maxwellian electron distribution, it is possible to show that the spectral intensity is given by:

$$I(\lambda) = \text{const.} \cdot T_e^{-3/2} N_e^2 \lambda^{-3} \exp\left( - \frac{hc}{\lambda kT_e} \right)$$

(E-1)

where $I(\lambda)$ is the radiation intensity at wavelength $\lambda$, $h$ is Plank's constant, $k$ is Boltzmann's constant, $c$ is the speed of light, and $N_e, T_e$ are the plasma electron concentration and electron temperature respectively. Thus, we can write for given plasma conditions:

$$\ln [I(\lambda) \lambda^3] = \text{constant} - \frac{hc}{\lambda kT_e}$$

(E-2)

Since $I(\lambda) \propto hcn/\lambda$, where $n$ is the number of photons at wavelength $\lambda$, and the photomultiplier signal $S(\lambda)$ (measured in volts, for instance) is proportional to $n/\lambda$, where $\eta(\lambda)$ is the photocathode quantum efficiency, we can write:

$$I(\lambda) = \text{constant} \cdot \frac{hc S(\lambda)}{\eta(\lambda) \lambda}$$

(E-3)

consequently:

$$\ln \left( \frac{S(\lambda) \lambda^2}{\eta(\lambda) \lambda} \right) = \text{const.} - \left( \frac{hc}{kT_e} \right) \frac{1}{\lambda}$$

(E-4)

If we plot $\log(S(\lambda)\lambda^2/\eta(\lambda)\lambda)$ vs. $1/\lambda$, we obtain a straight line whose slope is inversely proportional to $T_e$. The electron temperature can be readily calculated from:

$$T_e = \frac{hc}{2.3k} \left( \text{slope} \right)^{-1}$$

(E-5)

We have measured experimentally the radiative recombination spectrum into the $^4P_{3/2,1/2}$ levels of potassium for three different discharge currents: 1.0, 2.0 and 3.0 Amps. The recorded spectrum was used to determine $T_e$ as indicated above.
Unfortunately, the Langmuir probe insulator contamination at the time these measurements were performed, was quite severe, and this fact precluded a direct comparison of results, which would have been extremely valuable. Nevertheless, we were able to compare the spectroscopic measurements with earlier probe data for similar conditions, obtained at a time when the probe operation was estimated to be reasonable free from contamination effects.

We should also note the recombination spectrum of potassium was observable for discharge currents above 1 Amp, which were considerably higher than the currents of interest in the laser excitation experiment.

Consequently, spectroscopic measurements are relevant only in as much as they provide a reference for comparison with probe data, under similar plasma conditions. We have obtained the following results.

<table>
<thead>
<tr>
<th>Discharge current</th>
<th>1A</th>
<th>2A</th>
<th>3A</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_e$ (spectroscopic)</td>
<td>1100$^0$K</td>
<td>1400$^0$K</td>
<td>1900$^0$K</td>
</tr>
</tbody>
</table>

Under similar conditions, for a discharge current of 1.5A, the Langmuir probe indicates $T_e = 6300^0$K, and a degree of ionization of 29%.

Assuming that the plasma is in collisional-radiative equilibrium, the measured degree of ionization would correspond to an electron temperature of about $3400^0$K.

Of course it should be born in mind that the spectroscopic measurements represent an average value, which has contributions from the cooler peripheral regions of the discharge.

Consequently, the spectroscopically measured values are in principle compatible with the temperature of ($T_e \sim 3400^0$K) needed to account for the degree of ionization. The probe reading, on the contrary, is too high to be compatible with the measured degree of ionization. Considering also the large discrepancy of this value with respect to the spectroscopic measurements, and the problems associated with the alkali contamination, it is estimated that the temperature indicated is in error.

We conclude from the present comparison that the probe readings are likely to overestimate the actual electron temperature. The exact mechanism that leads to this effect is not clear, although it is estimated to be connected to potassium contamination effects, as discussed in Appendix D.

References


Normalized Plasma Enhanced Emission at 6939 A

Computer Results
- $N_e = 5 \times 10^{11}$ cm$^{-3}$
- $N_e = 2 \times 10^{12}$ cm$^{-3}$
- $N_e = 10^{13}$ cm$^{-3}$
- $T_e = 2500^\circ K$

FIG. 3
CALCULATED POWER SATURATION CHARACTERISTICS OF PLASMA ABSORPTION AT 6939 A
FIG. 4  CALCULATED TEMPORAL CORRELATION BETWEEN THE LASER PULSE AND THE ENHANCED PLASMA EMISSION AT 6939 Å, FOR DIFFERENT LASER PEAK POWERS
FIG. 5
CALCULATED DEPENDENCE OF PLASMA ENHANCED EMISSION AT 6939 Å ON LASER TUNING
\[ \frac{J^*(t)}{J_{\text{max}}^*} \]

Normalized Plasma Enhanced Emission

\[ N_e = 5 \times 10^{11} \text{ cm}^{-3} \]
\[ T_e = 2500 \text{ °K} \]

Computed Emission for Laser Power \( I_0 = 10^6 \text{ w/cm}^2 \)

Computed Emission for Laser Power \( I_0 = 50 \text{ w/cm}^2 \)

Laser Pulse

FIG. 6

COMPUTED ENHANCED PLASMA EMISSION AT 6939A
FIG. 7

SCHEMATIC ILLUSTRATION OF SPATIAL AVERAGING EFFECT

Laser Spatial Profile

I(r=0,t)

J^*(r,t)

Plasma Enhanced Emission

Laser Temporal Profile

t=0

t
Laser and Associated Equipment

**FIG. 8a**

Laser Temperature Control System

**FIG. 8b**
FIG. 9a  View of the Laser Cavity

KEY TO MATERIALS

- LUCITE
- METAL
- GLASS WOOL
- BORON NITRIDE
- TEFOLON

FIG. 9b  DIAGRAM OF THE LASER CAVITY
LASER WAVELENGTH CALIBRATION

![Graph showing laser wavelength calibration with various curves representing different experiments and references.]

**FIG. 10**

5 Consecutive Ruby He-Ne Laser Shots (Reference) Interferometer Free Spectral Range 0.024 Å

RUBY LASER WAVELENGTH REPRODUCIBILITY

**FIG. 11**

Interferometer Free Spectral Range 0.024 Å

**FIG. 12**
$I(t)/I(t=0)$ Normalized Pulse Intensity

- Assumed in Analytical Model
  - $\tau_{\text{rise}} = 5$ nsec
  - $\tau_{\text{decay}} = 7$ nsec
- Experimental

**FIG. 13 LASER TEMPORAL INTENSITY PROFILE**
Normalized Laser Intensity

Gaussian Profile
($e^{-1}$ radius = 0.87 mm)

Assumed

FIG. 14 LASER SPATIAL INTENSITY PROFILE
RAZOR BLADE LIGHT TRAPS

SAPPHIRE WINDOWS

CERAMIC ELECTRICAL INSULATORS

FIGURE 15

POTASSIUM DISCHARGE CHAMBER
PLASMA DIODE CURRENT-VOLTAGE CHARACTERISTICS

Discharge Current (A)

[Graph showing discharge current as a function of applied potential]

PLASMA DIODE BREAKDOWN POTENTIAL CHARACTERISTICS

Breakdown Potential (Volts)

[Graph showing breakdown potential as a function of interelectrode gap]

Dependence of diode current on potassium vapour pressure

[Graph showing diode current as a function of potassium vapour pressure]

Dependence of diode discharge current on cathode temperature

[Graph showing discharge current as a function of cathode temperature]
FIG. 20a

FIG. 20b  EXPERIMENTAL ARRANGEMENT
FIG. 21  EFFECTS OF LASER TUNING ON THE PLASMA ENHANCED EMISSION AT 6911 A.
Plasma Emission at 5800 A

Plasma Emission at 6911 A

Diode current $I = 2.0$ amp.

5800 A

$I = 0.75$ amp.

6911 A

5800 A

$I = 0.30$ amp.

6911 A

20 nsec/div

FIG. 22 INTENSIFICATION OF RADIATION AT 5800 A AND 6911 A FOR DIFFERENT DISCHARGE CURRENTS.
FIG. 23 DEPENDENCE OF PLASMA ENHANCED EMISSION DECAY TIME ON ELECTRON DENSITY ($\lambda=6911\AA$)
FIG. 24 DEPENDENCE OF ENHANCED PLASMA EMISSION ON LASER POWER

Normalized Plasma Signal Peak

○ Experimental
△ Calculated

Laser Wavelength Mismatch $\Delta \lambda < 0.015 \text{Å}$

FIG. 25 DEPENDENCE OF PLASMA SIGNAL DECAY TIME ON LASER POWER

Signal Decay Time ($1/e$) (nsec)

○ Experimental

Wavelength Observed $\lambda = 6911 \text{Å}$

Constant Laser Wavelength ($\Delta \lambda \leq 0.01 \text{Å}$)
FIG. 26 DEPENDENCE OF ENHANCED PLASMA EMISSION ON LASER WAVELENGTH
FIG. 27
DEPENDENCE OF PLASMA SIGNAL DECAY TIME ON LASER TUNING

Signal Decay Time (1/e) (nsec)

0  50  100

0.01 0.1 0.5

Laser Wavelength Mismatch (Å)

Wavelength Observed $\lambda = 6911$ Å
Constant Laser Power $\sim 10^6$ W/cm²

○ Experimental

FIG. 28
DEPENDENCE OF PLASMA SIGNAL DECAY TIME ON LASER TUNING

Signal Decay Time (1/e) (nsec)

0  10  15  20

0  0.05  0.10  0.15

Laser Wavelength Mismatch (Å)

Wavelength Observed $\lambda = 6911$ Å
Constant Laser Power $\sim 10^6$ W/cm²

○ Experimental
FIG. 29  EFFECTS OF LASER PULSE WIDTH ON PLASMA ENHANCED EMISSION (for constant plasma conditions)
FIG. 30
SIGNAL DECAY TIME vs FREE ELECTRON DENSITY
FIG. 31 TEMPORAL VARIATION OF THE ENHANCED PLASMA EMISSION AT 6939Å FOR DIFFERENT RADII

Correlation Based on Computer Results for $N_e = 2 \times 10^{12}$, $T_e = 2500\, ^oK$
Laser Peak Power $I_0 = 10^7\, \text{w/cm}^2$
**FIG. 32**

Correlation Based on Computer Results for $N_e = 5 \times 10^{11}$ cm$^{-3}$, $T_e = 2500$°K

Laser Peak Power $I_p = 10^7$ W/cm$^2$

**FIG. 33**

Correlation Based on Computer Results for $N_e = 10^{13}$ cm$^{-3}$, $T_e = 2500$°K

Laser Peak Power $I_p = 10^7$ W/cm$^2$

TEMPORAL VARIATION OF THE ENHANCED PLASMA EMISSION AT 6939Å FOR DIFFERENT RADII
FIG. 34
CALCULATED DYNAMICS OF THE PLASMA ENHANCED EMISSION BUILD-UP PERIOD

FIG. 35
CALCULATED DYNAMICS OF THE PLASMA ENHANCED EMISSION DECAY PERIOD
**FIG. 36**

**GEOMETRICAL CONSIDERATIONS ABOUT THE EXCITED PLASMA VOLUME.**

\[ I(x) = 2 \int_{x}^{R} \frac{J^*(r)r}{\sqrt{r^2-x^2}} \, dr \]

\[ R > x_0 \]

- **observed plasma volume**
- **photomultiplier**
- **stop**
- **lens**
- **laser beam perpendicular to paper**
- **full cylinder observed**
- **truncated** cylinder observed
- \( x_0 > R \)
FIG. 37
CALCULATED PLASMA ENHANCED EMISSION SIGNAL FOR DIFFERENT LASER POWERS
FIG. 38

CALCULATED DEPENDENCE OF PLASMA PEAK SIGNAL ON EFFECTIVE LASER POWER

Normalized Plasma Peak Signal

Peak Plasma Signal Normalized to Signal for \( I_{\text{eff}} = 6 \times 10^6 \text{ w/cm}^2 \)

Laser Effective Peak Power Density

\[
I_{\text{eff}} = \frac{L_v}{L_{v_0}} I_0 \quad (\text{w/cm}^2)
\]
FIG. 39 NORMALIZED PLASMA ENHANCED EMISSION SIGNAL

Calculated for
Ne = 2 \times 10^{12} \text{ cm}^{-3}, \ Te = 2500^\circ \text{K}

FIG. 40 NORMALIZED PLASMA ENHANCED EMISSION SIGNAL

Calculated for
Ne = 2 \times 10^{12} \text{ cm}^{-3}, \ Te = 2500^\circ \text{K}
Normalized Line Profile

\[ \frac{L_{\nu}}{L_{\nu_0}} \]

(\( \lambda_0 = 6939 \text{A} \))

Wavelength Difference \( \lambda - \lambda_0 (\text{A}) \)

FIG. B1-CALCULATED ABSORPTION LINE PROFILE
LANGMUIR PROBE GEOMETRY AND MOUNTING DETAILS

FIG. D-1

LANGMUIR PROBE CIRCUIT

FIG. D-2
probe trace taken with contaminated insulator

probe trace for identical plasma conditions, after cleaning.

discharge to insulator

proper discharge to probe tip

ALKALI CONTAMINATION EFFECTS ON LANGMUIR PROBE PERFORMANCE

FIG. D-3
TYPICAL CONTINUUM EMISSION SPECTRUM OF POTASSIUM DUE TO RADIATIVE RECOMBINATION. 
NOTE: the results have not been corrected for spectral response variation of the PMT.
<table>
<thead>
<tr>
<th>$\xi$</th>
<th>$\Delta \lambda [\text{A}^\circ]$</th>
<th>$e^{-\xi^2}$</th>
<th>$-2 \sqrt{\pi} \xi^{-1/2} [1 - 2 F(\xi)]$</th>
<th>$L_\nu/L'_{\nu_0}$</th>
<th>Normalized Line Profile $L_\nu/L_{\nu_0}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>1</td>
<td>$-1.47 \times 10^{-2}$</td>
<td>0.985</td>
<td>1</td>
</tr>
<tr>
<td>0.5</td>
<td>$6.33 \times 10^{-3}$</td>
<td>0.78</td>
<td>$-8.45 \times 10^{-3}$</td>
<td>0.771</td>
<td>$7.89 \times 10^{-1}$</td>
</tr>
<tr>
<td>1</td>
<td>$1.27 \times 10^{-2}$</td>
<td>0.37</td>
<td>$1.105 \times 10^{-3}$</td>
<td>0.371</td>
<td>$3.76 \times 10^{-1}$</td>
</tr>
<tr>
<td>2</td>
<td>$2.53 \times 10^{-2}$</td>
<td>1.83$\times 10^{-2}$</td>
<td>$3.00 \times 10^{-3}$</td>
<td>$2.13 \times 10^{-2}$</td>
<td>$2.16 \times 10^{-2}$</td>
</tr>
<tr>
<td>3</td>
<td>$3.80 \times 10^{-2}$</td>
<td>1.23$\times 10^{-4}$</td>
<td>$1.01 \times 10^{-3}$</td>
<td>$1.13 \times 10^{-3}$</td>
<td>$1.15 \times 10^{-3}$</td>
</tr>
<tr>
<td>4</td>
<td>$5.06 \times 10^{-2}$</td>
<td>$10^{-7}$</td>
<td>$5.07 \times 10^{-4}$</td>
<td>$5.07 \times 10^{-4}$</td>
<td>$5.15 \times 10^{-4}$</td>
</tr>
<tr>
<td>6</td>
<td>$7.60 \times 10^{-2}$</td>
<td>$10^{-7}$</td>
<td>$2.08 \times 10^{-4}$</td>
<td>$2.08 \times 10^{-4}$</td>
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<td>$3.90 \times 10^{-5}$</td>
<td>$3.96 \times 10^{-5}$</td>
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**TABLE B-1. CALCULATION OF ABSORPTION LINE PROFILE**

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<tr>
<th>$\xi$</th>
<th>$\Delta \lambda [\text{A}^\circ]$</th>
<th>Normalized Wing Profile $L_\nu/L_{\nu_0} = \frac{7.35 \times 10^{-3}}{\xi^2}$</th>
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**TABLE B-2. CALCULATION OF ABSORPTION LINE WING PROFILE**
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<th>λ_0n [mm]</th>
<th>λ_1n [mm]</th>
<th>λ_0n/λ_p</th>
<th>λ_1n/λ_p</th>
<th>T_e [eV]</th>
<th>N_e (1) [cm⁻³]</th>
<th>λ_p (1) [mm]</th>
<th>r_p/λ (1)</th>
<th>r_e/λ (1)</th>
<th>N_e (2) [cm⁻³]</th>
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**Table D-I: EXPERIMENTAL CONDITIONS**

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<th>N_e (1) [cm⁻³]</th>
<th>λ_p (1) [mm]</th>
<th>r_p/λ (1)</th>
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<th>N_e (1) [cm⁻³]</th>
<th>λ_p (1) [mm]</th>
<th>r_p/λ (1)</th>
<th>λ_1n/λ_p</th>
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<th>N_e (1) [cm⁻³]</th>
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**TABLE D-II.** CALCULATION OF N_e FROM ION COLLECTION DATA
<table>
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<tr>
<th>NO.</th>
<th>$T_e$ [°K]</th>
<th>$N_e$ [cm$^{-3}$] from i-</th>
<th>$N_e$ [cm$^{-3}$] from i+</th>
<th>$N_e(i+)$</th>
<th>$T_e$ (°K) (assumed)</th>
<th>$N_e$ [cm$^{-3}$] (reduced to the assumed $T_e$)</th>
<th>$\tau_d$ (nsec) decay time of plasma enhanced emission signal</th>
</tr>
</thead>
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(*) Values not corrected for the finite resolution time of the measuring system ($\tau_{resolution} = 10$ nsec).

Table D-III. Values Obtained
A thermally tuned Q-switched ruby laser of moderate power, with emission wavelength at 6939A, has been used to selectively excite the \( \frac{4}{2}P_{3/2} \rightarrow \frac{5}{2}D_{1/2} \) transition in the atoms of a pure potassium plasma of low degree of ionization produced in a hot cathode diode. A pulse of intensified spontaneous emission has been produced at 6939A, 6911A and at a number of other wavelengths, as a result of the laser selective excitation. The characteristics of the plasma intensified emission pulse have been observed to depend on the local conditions of the plasma, at the point of intersection of the laser beam and the solid angle of observation, estimated by a Langmuir probe. In particular, it has been observed that the signal decay time decreases as the free electron density increases. The possibility of using this type of interaction for the purpose of diagnosing the plasma has been studied experimentally. The results obtained indicate that using an adequately tailored laser pulse it should be possible to measure the electron density and the excitation temperature in a potassium plasma in the range \( 10^{12} \text{ to } 10^{14} \text{ cm}^{-3} \) and \( 2000 \text{ to } 3500 \text{ K} \) respectively. The spatial resolution associated with the measurement should be of the order of a few cubic millimeters, with a temporal resolution of better than \( 10^{-7} \text{ sec} \). Furthermore, because of the low power required for excitation, the plasma perturbation effects associated with the measuring technique are likely to be negligible.
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<th>LINK C</th>
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<td>LASER-PLASMA RESONANT INTERACTION</td>
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A thermally tuned Q-switched ruby laser of moderate power, with emission wavelength at 6939 Å, has been used to selectively excite the $4^2P_{3/2} - 4^2S_{1/2}$ transition in the atoms of a pure potassium plasma of low degree of ionization produced in a hot cathode diode. A pulse of intensified spontaneous emission has been observed at 6939 Å, 6911 Å and at a number of other wavelengths, as a result of the laser selective excitation. The characteristics of the plasma intensified emission pulse have been observed to depend on the local conditions of the plasma, at the point of intersection of the laser beam and the solid angle of observation, estimated by a Langmuir probe. In particular, it has been observed that the signal decay time decreases as the free electron density increases.

The possibility of using this type of interaction for the purpose of diagnosing the plasma has been studied experimentally. The results obtained indicate that using an adequately tailored laser pulse it should be possible to measure the electron density and the excitation temperature in a potassium plasma in the range $10^{12} - 10^{14}$ cm$^{-3}$ and 2000-3500 K respectively. The spatial resolution associated with the measurement should be of the order of a few cubic millimeters, with a temporal resolution of better than 10$^{-7}$ sec. Furthermore, because of the low power required for excitation, the plasma perturbation effects associated with the measuring technique are likely to be negligible.

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A thermally tuned Q-switched ruby laser of moderate power, with emission wavelength at 6939 Å, has been used to selectively excite the $4^2P_{3/2} - 4^2S_{1/2}$ transition in the atoms of a pure potassium plasma of low degree of ionization produced in a hot cathode diode. A pulse of intensified spontaneous emission has been observed at 6939 Å, 6911 Å and at a number of other wavelengths, as a result of the laser selective excitation. The characteristics of the plasma intensified emission pulse have been observed to depend on the local conditions of the plasma, at the point of intersection of the laser beam and the solid angle of observation, estimated by a Langmuir probe. In particular, it has been observed that the signal decay time decreases as the free electron density increases.

The possibility of using this type of interaction for the purpose of diagnosing the plasma has been studied experimentally. The results obtained indicate that using an adequately tailored laser pulse it should be possible to measure the electron density and the excitation temperature in a potassium plasma in the range $10^{12} - 10^{14}$ cm$^{-3}$ and 2000-3500 K respectively. The spatial resolution associated with the measurement should be of the order of a few cubic millimeters, with a temporal resolution of better than 10$^{-7}$ sec. Furthermore, because of the low power required for excitation, the plasma perturbation effects associated with the measuring technique are likely to be negligible.

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