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Title The Pressure Effect and Time Resolution of Helium Molecular Bands in a Pulsed Microwave Plasma

Abstract approved [Signature] (Major Professor)

The behavior of all of the observed molecular band emission as well as the prominent atomic line emission from a pulsed microwave helium plasma is given. Extensive efforts to purify the helium include the use of a uranium getter, a quartz diffusion tube, and a cataphoretic discharge. The plasma is produced in a cylindrical pyrex tube contained in a square waveguide section using pulsed $x$-band radiation from a 725A magnetron.

From the variations of the light intensity as a function of time, together with the pressure and power effects it is concluded that the molecular radiation follows from the same process both during and after the microwave excitation pulse. The dominant process is believed to be collisional recombination by the reaction $\text{He}_2^+ + 2e \rightarrow \text{He}_2^* + e$ with subsequent radiation from the excited molecules. This radiation appears at about the
same time as the atomic emission upon production of the plasma. The low molecular intensity during the microwave excitation pulse is probably due to quenching by the applied microwave field. While the initial degree of vibrational excitation of the helium molecular ion is not known, the observed molecular emission involved only the $0,0$ vibrational levels both during the pulse and in the afterglow.

Because of the extensive pressure variation, significant changes in the temporal light intensity of the atomic emission are also observed. These observations show that the excitation pulse period emission and the afterglow emission are significantly different. The dominant process during the excitation pulse is undoubtedly inelastic electron excitation of the helium atom. At higher pressures recombination at a reduced level is also present during the pulse and is dominant in the afterglow.

At least part of the decrease in atomic light intensity after the plasma formation and during the pulse is associated in time with increased transmission of the incident microwaves. There is, however, no observed change in the reflected microwave pulse at the time of this intensity decrease.
THE PRESSURE EFFECT AND TIME RESOLUTION OF HELIUM
MOLECULAR BANDS IN A PULSED MICROWAVE PLASMA

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THE PRESSURE EFFECT AND TIME RESOLUTION OF HELIUM MOLECULAR BANDS IN A PULSED MICROWAVE PLASMA

I. INTRODUCTION

The background presented in this first section is given with the justification that a review of this subject is not presently available and also that it is historically important by way of introducing the long series of the helium discharge studies which eventually led to the discovery of the helium bands.

The first reported observation of helium bands by Heuse (48, p. 13) in 1900 was followed for many years by disagreement and uncertainty as to the source of the optical radiation before the subject could be discussed with a degree of clarity. An understanding of the experimental observations was clouded by the fact that atomic and molecular theories were not clear at this time and consequently the interpretation of results was difficult. Before this and other early observations of the helium bands could be appreciated it was necessary to resolve first what will be called the helium-hydrogen problem.

1.1 The Helium-Hydrogen Problem

In 1913 a series of notes appeared in Nature concerning a helium discharge known to contain small contaminating and unavoidable amounts of hydrogen.
Evans (34, p. 5) directed attention to experimental work of Fowler and theoretical efforts of Bohr in connection with the resulting helium-hydrogen spectrum. Before Fowler's work the Balmer and Paschen series were the only ones known for hydrogen. But by passing a strongly condensed discharge through mixtures of helium and hydrogen, Fowler was able to photograph many more lines which he attempted to arrange into hydrogen-like series. Bohr on the other hand claimed that some of these lines were due to helium to which Evans agreed (34, p. 5) since his photographs of the 4686A line showed no evidence of either the H_\alpha or H_\beta lines. Furthermore, one of the lines present in the helium-hydrogen discharges and lying at 4686A could not be observed with either hydrogen-neon or hydrogen-argon mixtures.

Later Fowler (38, p. 232) reported experiments of enhanced radiation of the CO bands in helium mixtures in an effort to substantiate his idea that the helium in the helium-hydrogen experiments served to enhance the hydrogen lines. He called attention to the fact that Paschen had found the infrared series in hydrogen more intense in the hydrogen-helium mixtures than in hydrogen alone. According to Fowler the new atomic theory now proposed by Bohr was not conclusively demonstrated experimentally although he conceded that agreement was
becoming more satisfactory. In rechecking Evan's work Fowler satisfied himself that the 4686A line indeed was not observed in either hydrogen-neon or hydrogen-argon gas mixtures.

After another short interval Bohr (11, p. 231) re-emphasized his claim that the new lines previously observed by Fowler in the hydrogen-helium discharges were due to helium. He further called attention to the hydrogen-like series in helium to be expected at 6560.37A, 4859.53A, 4338.86A etc. In reply, Fowler (39, p. 232) agreed that with the more exact calculations now used by Bohr, the lines could possibly be due to helium but that the theory was still incomplete insofar as helium was concerned.

In the following year Fowler (39, p. 145) again commented upon the status of the 4686A line. After reviewing the work of the previous year and emphasizing the differences in arc and spark type spectra, he agreed with Bohr that the 4686A line was indeed due to a spark type discharge in helium and that in contrast the Lyman, Balmer, and Paschen hydrogen series appeared to be of the arc type. Another review of the helium-hydrogen problem was given by Evans (35, p. 284-297) in December 1914, where he also agreed that Bohr's calculations were correct and that when clean helium gas was used, the
hydrogen lines did not appear. In February 1915 Bohr (12, p. 6, 7) claimed that just as the hydrogen spectra wave numbers \( \nu \) could be represented by the well-known equation \( \nu = R \left( \frac{1}{n_1^2} - \frac{1}{n_2^2} \right) \), the wave numbers of the disputed lines could be derived from the equation

\[ \nu = 4R' \left( \frac{1}{n_1^2} - \frac{1}{n_2^2} \right) \text{ with } R'/R = 1.0041. \]

This last series of lines was assumed by Bohr to originate from the ionized helium atom since it was reported experimentally that the Balmer lines appeared at an excitation level of about 13eV, the normal helium lines at about 30eV, and the 4686A line at about 80eV while according to Bohr's atomic theory one could expect the hydrogen lines at an excitation potential of about the theoretical ionization energy of 13.6 eV, the normal helium lines at about 29eV, and the ionized helium lines at about 83eV. While the detailed quantitative aspects still left something to be desired, Bohr felt that qualitatively the relatively high energy required to produce the 4686A line together with the theory of the ionized helium atom was something more than fortuitous.

In 1916, Evans (36, p. 56, 57) reviewed the previous years efforts again with particular emphasis on the fact that he had been able to obtain the 4686A line in a helium discharge with no known hydrogen contamination. He also called attention to the fact that the hydrogen
interpretation of the 4686A line required a doublet structure with a separation of 0.0674A. From Bohr's atomic theory it was not certain what type of structure to expect, but Evans hoped to settle the problem once and for all by examining the structure of the 4686A line. Experiments showed that the line was a close doublet of separation 0.094A but with a sharpness comparable to helium lines rather than hydrogen. Upon closer examination a third faint line was observed on the high wavelength side as predicted in a generalization of Bohr's theory by Sommerfeld (98, p. 89). It is now an accepted fact that the 4686A line, a member of the Pickering series originally discovered in the star $\delta$-Puppis, arises from ionized helium atoms. This is a system which may be treated by Bohr's theory and the resulting wave numbers of the 4686A series may be found from the expression: \[ \nu = 4R_{He} \left( \frac{1}{3^2} - \frac{1}{n^2} \right) \] where the Rydberg constant \( R_{He} = 109,722 \text{cm}^{-1} \) and \( n = 4, 5, 6, \ldots \).

This interesting dispute as to the correct origin of the 4686A line, the development of a proper theory, the importance of the gas purity in the experiments, as well as the discovery of the helium bands, not yet discussed, all help give one an appreciation for the rather intense interest in the experimental helium discharge studies and those which were to continue.
Although the helium origin of some of the atomic lines seemed now beyond question, the difficulty in preparing pure gas samples continued to raise some doubt as to origin in the early discussion of the helium bands (17, p. 146).

1.2 The Helium Bands

The earliest reference on the band spectra in helium discharges by Heuse (48, p. 18) in the year 1900 contained no more than a passing remark that it appeared that one would have to be concerned with band as well as line spectra in the helium studies. In 1913, both Goldstein (44, p. 402) and Curtis (17, p. 146) published results giving considerable detail in their description of many of the helium bands. These papers really mark the beginning of the serious study of the helium bands. Persistent interest was to follow until 1930 with particular emphasis on the years 1928-1929, as indicated in the chronological summary in the appendix. A paper has yet to be written, however, which offers a complete summary of these helium band studies although many partial reviews are available to guide one to the original papers on the subject, for example: Dieke (30, p. 647), Herzberg (49, p. 535), Sponer (100, p. 9, 40-62), Weizel (111, p. 252), Landolt (59, p. 7), Jevons (54, p. 270),
and Pearse (89, p. 123).

Although higher vibrational transitions (the vibrational energy interval is of the order of 0.2eV) have been reported by Weizel (111, p. 255), most of the observed helium bands are (0-0) type transitions (30, p. 648). According to Weizel (111, p. 253) the most outstanding characteristics of the helium band system are that the bands themselves conform to a Rydberg type series as in atomic spectra and that two analogous singlet and triplet systems exist which evidently do not combine with one another. The fact that an atomic type series is observed is explained qualitatively by Mulliken (80, p. 60) as being due to the rather tight core type action of the two helium nuclei together with the three lower energy electrons on the fourth excited electron with a significantly larger radial expectation value. Such a system on the basis of Bohr's atomic theory would be expected to give a hydrogen type spectrum.

Of the many aspects of the helium bands, the one with which this study is mainly concerned is the manner in which the molecule is produced in pulsed discharges and its relation to the atomic excitation and ion production. This is a difficult area since there are several particle species which are affecting the discharge and transient effects may still be important. Consequently the results
are at this stage somewhat sketchy. It should be emphasized that most of the recent work with helium in the case of pulsed discharges has been restricted to times after 100 microseconds from the end of the discharge where sufficient assumptions can be made to simplify the problem. Even with these precautions it is still difficult to find agreement in the reported results. The very early characteristics of both the pulsed dc as well as the high frequency discharges have in the main been neglected because of their complicated nature. It is felt, however, that even if an initial study only includes the experimental facts these early characteristics cannot be overlooked if one is to understand the pulsed helium discharge completely. Furthermore, in view of the difficulties still existing (81, p. 1639; 37, p. A381) in the afterglow ionization studies, an additional examination of a closely related but neglected area may yield information that will prove helpful to the over-all view.

Table I summarizes some atomic and molecular processes which have been used to account for the results of various helium studies, including the more recent ones. The asterisks indicate excitation while the m superscript denotes a metastable level. The + sign represents either an ionized atom or molecule.
TABLE I

ATOMIC AND MOLECULAR REACTION PROCESSES IN HELIUM

1. \( \text{He}^* + \text{He} \rightarrow \text{He}_2^+ + e \) Arnot (3, p. 106), Bennett (8, p. 367), Helland (46, p. 81), Hornbeck (52, p. 621), Loeb (62, p. 975), Morris (74, p. 11), and Oskam (85, p. 390).

2. \( \text{He}^m + \text{He}^m \rightarrow \text{He}_2^+ + e \) Morris (74, p. 11), Myers (81, p. 1640), and Oskam (85, p. 392).

3. \( \text{He}^+ + 2\text{He} \rightarrow \text{He}_2^+ + \text{He} \) Bates (7, p. 718), Bennett (8, p. 367), Chen (14, p. 1393), Goldstein (45, p. 471), Helland (46, p. 81), Hornbeck (51, p. 621), Janin (53, p. 1073), Loeb (62, p. 567), Massey (64, p. 415), Morris (74, p. 11), Myers (81, p. 1640), Oskam (85, p. 390), Pahl (86, p. 239), Phelps (90, p. 103), and Rogers (94, p. 57).

4. \( \text{He}^m + 2\text{He} \rightarrow \text{He}_2^+ + \text{He} + e \) Bennett (8, p. 367), Brigman (13, p. 958), and Phelps (91, p. 1307).

5. \( \text{He}^+ + \text{He} \rightarrow \text{He}_2^+ \) Arnot (3, p. 106), Morris (74, p. 11), and Myers (81, p. 1641).

6. \( \text{He}_2^m + e \rightarrow \text{He}_2^+ + 2e \) Morris (74, p. 11).

7. \( \text{He}_2^+ + e \rightarrow \text{He}_2^* \) Biondi (10, p. 157).

8. \( \text{He}_2^+ + e \rightarrow \text{He}^+ + \text{He} + e \) Chen (14, p. 1400).

9. \( \text{He}_2^+ + e \rightarrow \text{He}^* + \text{He} \) Bates (7, p. 718), Bennett (8, p. 367), Chen (14, p. 1393), Loeb (62, p. 567), Myers (81, p. 1640), Oskam (85, p. 391), and Rogers (94, p. 2).
10. \( \text{He}_2^+ \rightarrow \text{Wall} \) Myers (81, p. 1640).

11. \( \text{He}(1^1S) + \text{He}(n^1P) \rightarrow \text{He}(1^1S) + \text{He}(n^3S) \) Biondi (9, p. 453), and Meyerott (69, p. 671).

12. \( \text{He}^m + \text{He}^m \rightarrow \text{He}^+ + \text{He} + \text{e} \) Biondi (9, p. 453), Goldstein (45, p. 478), Kunkel (58, p. 220), Myers (81, p. 1640), Oskam (85, p. 392), Phelps (90, p. 103), and Rogers (94, p. 44).

13. \( \text{He}^m + \text{He} \rightarrow 2\text{He} \) Myers (81, p. 1640).

14. \( \text{He}^m \rightarrow \text{Wall} \) Myers (81, p. 1640).

15. \( \text{He}(2^1S) + \text{e} \rightarrow \text{He}(2^3S) + \text{e} \) Biondi (9, p. 453), Phelps (91, p. 1307), and Rogers (94, p. 52).

16. \( \text{He} + \text{e} \rightarrow \text{He}^+ + 2\text{e} \) Bennett (8, p. 367) and Morris (74, p. 11).

17. \( \text{He} + \text{e} \rightarrow \text{He}^* + \text{e} \) Bennett (8, p. 367), Holland (46, p. 81), Hornbeck (52, p. 621), and Loeb (62, p. 567).

18. \( \text{He}^+ + \text{e} \rightarrow \text{He} + \hbar \nu \) Bates (7, p. 718), and Myers (81, p. 1640).

19. \( \text{He}^+ \rightarrow \text{Wall} \) Myers (81, p. 1640).

20. \( \text{He}_2^+(\nu) + \text{He} \rightarrow \text{He}_2^+(\nu \nu) + \text{He} \) Ferguson et al. (37, p. A382).
It should be made clear once more that Table I represents attempts to describe the processes occurring at times usually greater than 100 microseconds after the plasma production. They will be examined, however, as to their applicability to the power pulse period and the early afterglow of this study. It is anticipated that the experimental results shown later will allow a qualitative discussion to be made concerning some of the more important reactions occurring early in the plasma formation for high purity helium gas.

Since the production of the molecules is pressure sensitive and these details are not known to be available, the experimental results of the pressure variation of the light intensity will also be summarized in this study. It is believed that the range of pressure variation shown in the results of this study are the most extensive of any of the reported helium studies.
II. EXPERIMENTAL ARRANGEMENT

2.1 Introduction

In the early stages of this experiment it was decided to emphasize the early optical behavior of a weak helium plasma produced in a containing enclosure by 3.2 cm microwave radiation. The manner in which this was accomplished can best be described in terms of the gas handling, microwave excitation, and optical detection system. Related details of a similar system are also given by Tynes (103, p. 11-45).

2.2 The Gas Handling System

A schematic of the gas handling system is shown in Figure 1. It consists of a 70 liter/second, three-stage mercury diffusion pump backed by a 1.6 liter/second mechanical pump which can be used to evacuate either the treated gas storage volume or the plasma tube. The pressure at the mechanical pump may be monitored by means of a removable thermocouple gauge which is not shown, while the lower pressures near the diffusion pump are determined with a Veeco ionization gauge. A second ionization gauge is also included near the plasma tube. It may be seen from Figure 1 that the tank helium undergoes three successive stages of purification by the uranium getter, the quartz diffusion tube, and the
Figure 1. Schematic of the gas handling system
cataphoresis tube. Appropriate liquid nitrogen traps and a bake-out oven complete the arrangement. The inclusion of an oven over all but the top of the discharge tube constituted a more reliable and convenient addition to the system over the previous method of wrapping heating tapes over certain sections and using hot air guns on the more accessible areas. The top and front side of the rectangular oven were formed in one piece and could be removed within minutes thus exposing the top and front side for valve adjustments, ionization gauge connections, and inspection. Multiple manual switch units on the two heating elements allowed one to control the warm-up and operating temperatures. Bake-outs were customarily over-night at about 400°C. Reflecting stands were used to protect the system close to the heating elements rather than including a driven fan to stir the air.

This modification of the apparatus reduced the evacuation time to hours rather than days as had been the case before. A heating tape was necessary on the tip of the discharge tube which extended out of the oven. However, since this was such a small section, generous insulation packed over the tapes allowed one to obtain slightly higher temperatures than in the oven itself. All temperatures were recorded from responses of
calibrated iron-constantan thermocouples placed at various locations.

The Uranium Getter

The properties that make uranium hydride useful in purifying gases have been known since 1949 when Spedding et al. (99, p. 4-5), Newton et al. (82, p. 21), and Warf et al. (105, p. 44) made extensive studies on the compound. Later Katz and Rabinowitch (55, p. 194-203) published data concerning the temperature and pressure effects on the uranium hydride compound. Three typical curves from their results are shown in Figure 2 and were used in determining the operating temperatures and pressures when forming and later decomposing the hydride. Other details in the reaction rates of hydrogen with uranium are furnished by Albrecht and Mallet (1, p. 404-409).

It should be made clear that in most applications of the uranium hydride for gas purification (72, p. 846) use is made of the dividing action as the hydride is formed. This has the effect of producing very finely divided uranium metal when the hydrogen is later driven off. Uranium metal in this condition is pyrophoric and must be isolated from the atmosphere (56, p. 34). The high chemical activity coupled with the large surface
Figure 2. Characteristic curves of uranium hydride formation and decomposition.
area presented after the hydride is formed are the major advantages utilized in the uranium treatment. In other applications one also finds the uranium hydride useful as a variable pressure hydrogen source (32, p. 187). The relative effectiveness of the uranium as opposed to other getters was not studied. Liquid helium has also been used to supply very pure helium gas.

**The Quartz Diffusion Tube**

The fused quartz diffusion tube was constructed as shown in Figure 3. The tube length was 53.3 cm with an outside diameter of 3.0 cm. The inner tube was expanded at periodic intervals to thin the walls to a few tenths of a millimeter. The tank gas was admitted to the larger cylinder and then upon heating the entire tube, the helium diffused into the smaller cylinder which formed part of what will be called the plasma tube volume. The action of the quartz diffusion tube in terms of cleaning the gas can best be appreciated by referring to past studies for a comparison of diffusion rates for several gases. Leiby and Chen (60, p. 271) give values of the permeation velocity $K$ through quartz in the equation

$$\frac{dp}{dt} = K\left(\frac{A}{Vd}\right) \Delta p$$

at a temperature of 400°C for helium, hydrogen, neon and nitrogen respectively of 470, 13, 5 and 0.06. In the preceding equation, $\frac{dp}{dt}$ is the time
rate of change of the pressure on the low pressure side of the diffusing membrane, A is the membrane area, d the membrane thickness, V the low pressure side volume, and \( \Delta p \) the pressure difference across the membrane. The units of K vary in the literature but for the numerical values given above K is dimensionally \((\text{cm}^3 \text{ of gas at NTP}) (\text{mm thickness})/(\text{sec})(\text{cm}^2 \text{ area})\). The variation in the values of the permeation velocity indicate a selective transfer through the membrane and in the case of helium an accompanying increase in helium gas purity. With the uranium getter reducing the effect of the active gases (principally hydrogen) the diffusion and cataphoresis tubes were used to reduce the concentration of the noble gases (principally neon). One could put the heating coil around only the inner cylinder as did Clark (16, p. 560), although no contamination problems were experienced with the present arrangement. Figure 4 shows the performance of the diffusion tube for several different tube temperatures.

**The Bakeable Metal Bellows Pressure Gauge**

In order to determine the higher operating pressures in the plasma tube after the purified helium gas was admitted, a metal bellows pressure gauge was used. This gauge, which is shown in Figure 5, functioned in a similar
To variac for temperature control

To plasma tube
To helium gas storage vessel and uranium getter

Figure 3. The fused quartz diffusion tube

![Graph showing plasma tube pressure as a function of time for different diffusion tube temperatures]

Figure 4. The plasma tube pressure as a function of time for different diffusion tube temperatures

To plasma tube
Kovar to pyrex seal

Metal clip
Wax seal

To LC meter
To auxiliary forepump and pressure gauge

Figure 5. Schematic of the sylphon pressure gauge
fashion to the more sensitive types in that the deformation of the bellows, as pressure differentials developed, was measured by the change in capacitance between the bellows and a fixed circular plate. The bellows assembly was enclosed and sealed in a brass housing so that variable external reference pressures of from $10^{-3}$ to 760mmHg could be used.

The ability to vary the external bellows pressure allowed the pressure measurements to be determined by the null method which involved first determining the null capacitance between the bellows and the fixed circular plate at some convenient reference pressure. This pressure was about $10^{-3}$mmHg on the outside and $10^{-8}$mmHg on the inside so that there was no measurable pressure difference deforming the bellows. Typically, this capacitance was about 150pf and as helium was admitted to the discharge tube the external pressure was varied in such a way as to preserve the 150pf capacitance value. An indication of the unusually good mechanical performance of the bellows is the fact that the 150pf capacitance was found to be stable to within $\pm 3$pf even after flexing the bellows with atmospheric changes in pressure. Now as the pressure in the gas discharge section increased the external pressure was increased to match it and then read from either the calibrated 0-100mmHg pressure gauge
or from a mercury manometer at the higher pressures.

The problem of obtaining initial parallel alignment of the fixed circular plate with the bellows was finally solved by enlarging the hole of the fixed probe and sealing it to the insulated lead with beeswax as shown in Figure 5. Parallel alignment was required to maximize the capacitance at any given separation. In this operation the heated circular plate was positioned in contact with the metal bellows and the upper plate of the external brass housing fastened. This involved lowering the Stupakoff insulator lead into the hot wax. As soon as the upper plate of the external brass housing was fastened with Allen screws and an O-ring seal, the external bellows pressure was reduced to about $10^{-3}\text{mmHg}$ while the wax was still hot in order to match the external pressure with the plasma tube which was always evacuated at this stage of the operation. If the hot wax was free from bubbles, it quickly cooled without spilling and the seal was made in a matter of minutes after which the upper plate of the external brass housing was removed and a metal clip used to bridge the wax seal to complete the electrical path to the L-C meter. Natural contraction of the cooling wax was sufficient to separate the circular plate attached to the insulator lead from the top of the metal bellows as shown in Figure 5. After
again replacing the upper plate of the external brass housing a reproducible null capacitance reading of about 150pf was indication that the gauge was ready for operation. Reproducibility of the gauge was found to be about $\pm 0.2\text{mmHg}$ so that percent errors in the pressure readings were less than 1% at pressures greater than 50mmHg. The relatively simple null calibration was all that was required for the operation of the gauge and could be determined just prior to admitting helium gas by diffusion into the plasma tube volume. It has been found that the bellows will withstand bake-out temperatures of 400°C for several hours under a pressure difference of approximately one atmosphere with no significant deterioration, providing a rigid support is attached to preserve its original position. However, in practice the assembly and calibration were accomplished after any bake-out procedures.

The Cataphoresis Tube

The cataphoretic effect involving a balance between diffusion and ion migration was discovered at least as early as 1893 by Baly (6, p. 200) in an experimental separation of rarified gases in an electric discharge. Later in 1925 (97, p. 284) it was again the general subject of an experimental investigation and in 1929 a
summary was made by Mierdel (70, p. 519-525). In 1935 Druyvesteyn (33, p. 255-266) presented some theoretical ideas of the phenomena which were later extended by Loeb (62, p. 1369). More recently Riesz and Dieke (93, p. 196-201) concluded that in noble gases and even with impurities of hydrogen and nitrogen, the minor constituent may be removed in each case.

The inclusion of the cataphoresis tube in the experimental arrangement was not considered to be as important in the purification of the helium as the uranium getter together with the diffusion tube action since separate spectrographic studies with and without the cataphoretic discharge showed no change in the spectral characteristics of the discharge.

**The Plasma Tube**

The actual discharge section or plasma tube was formed from pyrex test tubes having inner and outer diameters of 18 and 20mm respectively. The overall length was 49mm. A series of tubes with successively larger diameters had previously shown (103, p. 66) that longer afterglows were possible with the larger discharge vessel according to expected diffusion effects. In these earlier studies the tube diameter was limited by the dimensions of the x-band waveguide. In the present
arrangement the limitation came from the 22.9mm square waveguide transition section. The inner and outer diameters of the stem joining the plasma tube to the rest of the system were 3 and 5mm respectively.

A Typical Run

A typical run involved a procedure similar to the one described as follows: A three gram slug of uranium was sealed into a three liter spherical flask which is indicated in Figure 1 as the helium gas storage vessel. After checking for leaks, and having closed valve (No. 2) under high vacuum conditions, the pumps were used to evacuate the vessel to below $10^{-7}$mmHg. Three or more changes of hydrogen gas were used to flush this volume after which valve (No. 1) was closed and the vessel filled to slightly less than 760mmHg with hydrogen gas. The helium gas storage vessel was equipped with a separate oven which was then used to raise the temperature to 225°C where the reaction rate for the uranium hydride process has a maximum (see Figure 2). This temperature was held constant for several hours to assure a more complete conversion of uranium to uranium hydride.

Valve (No. 1) was then re-opened and the excess hydrogen removed through the pumps. The temperature of the helium gas storage vessel was then raised from 225°C
to about 400°C at which temperature the uranium hydride quickly decomposed leaving finely divided uranium. The evolved hydrogen was removed by pumping and the process continued for several hours until the pressure as read by ion gauge (No. 1) fell below $10^{-7}$mmHg. When this point was reached valve (No. 1) was closed and tank helium was admitted to the helium gas storage vessel. The temperature of the vessel was again cycled from 20°C to 400°C and back over a period of several hours to facilitate formation of chemical compounds with the non-rare gas impurities in the tank helium. This process has been found to be very effective in removing hydrogen and nitrogen. While the last cycling process was effected, valve (No. 2) was opened, the main oven temperature raised to about 400°C, and the plasma tube region evacuated to below $10^{-8}$mmHg as indicated by ion gauge (No. 2). A significant improvement in the operation of the gas handling system was accomplished by constructing a demountable oven which enclosed the two vacuum gauges, the two metal valves, the cataphoresis tube, the diffusion tube and part of the plasma tube. The section of the plasma tube which was enclosed within the waveguide transition section was simultaneously baked using special heating tape. After monitoring the pressure rise with ion gauge (No. 2) while valve (No. 2) was closed and
being assured that there were no leaks or significant outgassing, the quartz diffusion tube temperature was raised to about 500°C, allowing the treated tank gas to pass into the plasma tube. This produced a further purification of the helium by means of selective diffusion. Characteristic curves for several runs at different diffusion tube temperatures and different final pressures have been given in Figure 4. After the approximate desired pressure was attained the temperature of the diffusion tube was lowered to room temperature. After equilibrium with the room temperature a particular pressure was carefully approached from the high side by allowing helium to escape through valve (No. 2). At this time the plasma was produced, the pressure re-measured and the optical measurements made.

2.3 The Microwave Excitation

The production of the plasma was accomplished by means of a 725A magnetron which is shown in a schematic of the microwave excitation system in Figure 6. The 725A is a fixed frequency, pulsed type with peak pulse power capabilities of from 10 to 70 kilowatts which correspond to applied pulse voltages and currents of from 7 to 17 kilovolts and from 4 to 16 amperes respectively. The pulse duration is limited by the ability of the
Figure 6. Schematic of the microwave excitation system
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cathode to withstand ion bombardment. Two pulse lengths were available in the experimental arrangement. Their high-level durations were 0.6 and 2.1 microseconds at repetition frequencies of 1000 and 335 cycles/sec respectively. Variations in the characteristic frequency of this type of magnetron are indicated in the stated frequency band of from 9405 to 9345 megacycles/sec. The two magnetrons used in this experiment operated at an average frequency of 9374 megacycles/sec. The corresponding free space and guide wavelengths are 3.20 and 4.48 cm respectively. The average pulse power $P$ is related to the average power $P$, the average pulse duration $t$, and the repetition frequency $f$ by the relation $P = \int P dt / \int dt$ which becomes $P = P tf$ for rectangular pulses. This yields values for $P$ of 1215$P$ and 1330$P$ for the 0.6 microsecond, 1000 cycles/sec and the 2.1 microsecond, 335 cycles/sec pulses respectively, providing estimates of 0.823 and 2.24 microseconds are made for the average pulse durations rather than the previously mentioned 0.6 and 2.1 microsecond high-level values. Maximum values of the waveguide electric field in volts/cm for the two pulses are then given according to Moreno (73, p. 124) by

$$E_m = 1050P^{1/2}$$ and $$E_m = 1110P^{1/2}$$ respectively if $P$ is in watts.

The magnetrons were operated at a characteristic
fixed average power level of about 40 watts. The microwave radiation was directed to the power divider which allowed the desired fraction to pass towards the discharge region. The unused fraction was dissipated in the high-power termination. A general synchronizing trigger pulse, as shown in Figure 6, was obtained from the microwave pulse using a 1N23 crystal diode detector before the power division occurred. This pulse served as a time reference for the different observations both in the waveguide and through the photomultiplier tube. Directional couplers following the power divider, together with detecting mounts for Narda N-334 thermistors allowed measurements of incident, reflected, and transmitted power to be made. A ferrite isolator attenuated the reflected power pulses by 20db and prevented any reflections from disturbing the performance of the magnetron. The waveguide sections were RG-52/U type except for the plasma tube section which was a transition to a square cross-section to accommodate a larger plasma tube. The transition at each end was accomplished in 15cm by means of fixed inclined cuts finished with large-radius rounded ends. The internal finished square cross-section measured 2.287 by 2.287cm. Insertion power tests showed only 0.08% incident power reflected upon substitution of the transition section as compared with 0.05% for the
regular x-band guide. The waveguide was excited in the dominant $TE_{10}$ mode with all other modes experiencing exponential attenuation. A high-power termination following the plasma tube transition section absorbed any transmitted power and completed the excitation system.

2.4 The Detection System

The light emitted by the plasma was analyzed by either the Jarrell Ash 76-15m 480cm scanning spectograph with resolution and first order linear dispersion factors of 0.1 angstrom and 7 angstroms/mm respectively or the Bausch and Lomb DB548 grating monochromator with a linear dispersion of 66 angstroms/mm. The resolution of the monochromator was very sensitive to the slit width and for these observations varied from 20 to 100 angstroms depending upon the intensity of the band lines observed. No matter which dispersing system was used the basic action was the same in that the optical radiation from the plasma was incident on an entrance slit system, dispersed by the grating, focused on an exit slit system, and fell upon the photo-cathode of an RCA 1P21 photomultiplier tube. The signal developed across the coupling resistor was then directed either into the Tektronix 517A oscilloscope where the intensity (vs) time photographs were obtained or integrated
time-wise by the input circuit of a Gl1-A Varian recorder which then yielded the intensity (vs) wavelength charts. Figure 7 shows the schematic of the optical detection system while Figure 8 shows the 240KΩ photomultiplier tube dynode resistors and the 200KΩ coupling resistor all of which were connected directly to the pins of the tube. The capacitors on the last stages prevented pulse limiting for the more intense optical signals.

During the course of the investigation of the optical methods for observing various discharges the so-called "time-sampling" approach was considered. This method is usually reserved for very weak light sources and has been known to yield significant results with as few as 10 photons per second incident in the photomultiplier tube (10, p. 160). It was felt that such an approach in this work would extend the time during which data could be taken and consequently a circuit was devised similar to that used by Hendee and Brown (47, p. 52) and shown in Figure 9. Resistance units in this figure are ohms while capacitance units are microfarads except where picofarads are indicated.

The general synchronizing pulse from the LN23 crystal was not adequate for direct triggering of the 6SN7. Higher level pulses which were synchronized with the waveguide pulse were present, however, at both the
Figure 7. Schematic of the optical detecting systems
Figure 8. Conventional dc-operation circuit for the RCA 1P21 photomultiplier tube
Figure 9. Photomultiplier (PM) pulsing circuit - Synchronized and triggered with the magnetron pulse.
pulse forming network for firing the high level magnetron pulse and also the magnetron itself. Since the lack of synchronism between the trigger firing the spark gap and the spark gap discharge of the high level magnetron pulse was of the order of a few microseconds, the pulse used to trigger the 6SN7 in Figure 9 was obtained from the high level magnetron pulse. Very little delay or jitter was apparent in the magnetron firing. The trigger pulse initiated action in the variable delay whose pulse was inverted and sent to the univibrator where a delayed 60 volt positive pulse was sent to the 2D21 thyatron. The 2D21 thyatron was normally biased non-conducting. When this was overcome with the 60 volt positive pulse, the thyatron would fire for a time of twice that required to propagate an electrical signal the length of the coaxial cable in the plate circuit which served as a pulse forming network. The reason for this is that it takes half of this total time for the charge at the end of the cable to sense the cable is being discharged and then another equal time for it to travel out of the cable. The 36 kΩ charging resistor prevented the 2D21 thyatron from firing indefinitely while the positive pulse obtained from the cathode resistor was then used to fire the 829B tetrode which in turn delivered a 0.6 microsecond negative pulse of variable amplitude (0-5kilovolts) to
the cathode of the RCA 1P21 photomultiplier. Figure 10 shows intensity (vs) time for the 3889A line using the circuit of Figure 9. The degree of reproducibility of several attempts was used to estimate the uncertainty in the intensity readings. Although some data were obtained from this arrangement for the intense lines, the photomultiplier was found to oscillate at a frequency of about 5 megacycles/sec for pulse amplitudes high enough to give reasonable sensitivity for ordinary lines. This resulted in abnormally high "dark current" readings from the photomultiplier tube. Singer et al. (96, p. 42) also reported the possibility of oscillation in the case of a pulsed photomultiplier tube. Even though this extended time observation was not completely successful, evidence does seem to exist as shown in Figure 10 that in the present discharge arrangement a weak afterglow exists for times much longer than can be detected using the conventional photomultiplier circuit.

Since the more moderate positive pulse of the 2D21 thyatron was available a simple circuit was arranged for pulsing only the first dynode. In this modification of Figure 8 the 2D21 thyatron pulse was applied through two 1μF, 1000 volt capacitors to a 66 ohm terminating resistor which connected the cathode to dynode 1.
Figure 10. Intensity (vs) time for the 3889A line. The dashed horizontal line indicates the level of the "no-light background."
All other dynode resistors in Figure 8 were changed to 27kΩ and the normal 900 volt dc photomultiplier potential applied from the cathode to ground. The 66 ohm resistor resulted in a potential drop of only about 3 millivolts between the cathode and D1 with 100 volts being normal for the other dynodes. A normal voltage across the cathode and D1 appeared when the 100 volt 2D21 thyatron pulse was applied. Now the normal use of the circuit in Figure 8 resulted in a pulse voltage appearing across the 200kΩ signal resistor when photomultiplier action was occurring. However, in this modification the 200kΩ resistor was also removed and a capacitor inserted. The resulting voltage appearing across this capacitor in the anode circuit was measured after a given time interval with the Keithley electrometer. This modification of Figure 8 did not result in electrical oscillation of the photomultiplier but the shut-off action was not complete during the pulse off period even though only about 3 millivolts existed between the cathode and D1. This meant that for the complete period of an optical event there was some "leakage current" in the photomultiplier even though it was only to be pulsed on for some small fraction of the period of the event. In addition to this leakage current from the cathode to D1, the dark current would
contribute to the charge detected by the integrating circuit during that part of the period when little or no light was being emitted. In an effort to reduce the dark current the photomultiplier was cooled to 77°K with liquid nitrogen and normal dc potentials applied to the tube only from D2 to ground. The cooling reduced the dark current by a factor of nearly 50 with normal 100 volt dynode operating potentials for all dynodes from D2 to ground. However, when the 66 ohm pulsing resistor was used from the cathode to D1 and 900 volts placed between the cathode and ground similar to Figure 8, a significant over-all current reduction was not obtained by reducing the temperature.

It had originally been planned that the Jarrell-Ash spectrograph would be used together with a highly sensitive variable delay pulsed photomultiplier tube to yield time resolved intensity (vs) wavelength spectral charts of the helium band radiation. However, the poor results finally obtained required that this approach be modified and consequently the time resolved band results were obtained with the oscilloscope-monochromator combination and are shown in Section IV. It would seem that further efforts could still be made in pulsing the entire dynode structure in such a way as to minimize the effect of the oscillations. Also, a particular arrangement of the partial pulsing might be devised which would be more satisfactory.
III. THE INTENSITY VARIATION WITH POWER AND PRESSURE

3.1 Introduction

The available methods of collecting the band behavior data of the discharge were either visual, photographic, by chart recording, or with the Tektronix 517A oscilloscope. In the last two methods RCA 1P21 photomultiplier tube was used to detect the light emission. In the visual observations the principal advantage was the speed with which the observation could be made. This type of information eventually became very useful in connection with the question of hydrogen contamination. With practice one could easily see the Hα, Hγ, or Hη atomic lines when commercial tank helium was used in the discharge. With very weak hydrogen contamination the Hγ line was the most sensitive visual indicator.

The spectrum was photographed over a series of pressure ranges but again this was convenient mainly in fixing the line spacing and identifying the spectra. In order to obtain intensity information from the photographs they had to be analysed with the densitometer which added more time to the analysis. This approach is valuable, however, when very weak lines are being investigated since exposures for times of the order of hours may be used. In this respect the photographic
method surpasses the visual checks for impurities. Also it allows the most precise comparison with reference spectra for wavelength determinations.

The utility of the chart recording method lies in the simultaneous presentation of not only the line spacing but also the relative intensity values of each of the lines. Direct relative intensities of lines widely separated includes the effect of the variable response of the photomultiplier tube with the wavelength of the incident light. The chart recording proved to be very advantageous in terms of the time requirements in obtaining data. One limitation of this method in comparison to careful film work is the fact that the uncertainty in the line peak position on the direct chart recording is usually larger. This reflects the small irregularities in both the grating and chart motor drives and is discussed later in connection with Figure 13.

In pulsed type discharges the chart recording approach must effectively time integrate many nearly identical events so that the intensity resulting from this type of data reflects an average response during the duration of the optical event. For example, Figure 11 shows several sections of the 60mmHg, 39 watt chart recording (0.1 volt scale) with the helium bands marked with asterisks. The excitation pulse duration was 2.1
Figure 11. Intensity (vs) wavelength for helium at 60 mmHg, 39w. Peak band lines are marked with asterisks.
microseconds with a repetition rate of 335/sec. The atomic lines are also present and in many cases the stronger ones were allowed to go off-scale in order to more carefully observe the behavior of the bands. This is even more emphasized in Figure 11 since the upper 55% of the chart has been removed for convenience in the mounting. The wavelength region between 4400A and 4500A will not be discussed since the 4402A, 4436A, 4440A, and 4454A bands are mixed together here and the sacrifice of resolution in favor of intensity makes it difficult to properly identify them. The weaker 4388A, 4439A, and 4472A atomic lines were also identified in that region by comparing the charts at different pressures. However, the easily identified atomic line at 4472A is marked and at 4545A one observes the 4s$^2\Sigma_u^+ - 2p\pi^2\Pi_g$ band with the much stronger 3p$^2\Pi_g - 2s\pi^2\Sigma_u^+$ band at 4648A. Four atomic lines are marked as one increases in wavelength at 4713A, 4922A, 5016A, and 5047A. The only singlet series band observed is the $3\pi'\Pi_g - 2s\pi'\Sigma_u^+$ at 5133A. Next, one observes the rather intense $3d^5\Delta_u - 2p\pi^3\Pi_g$ band at 5733A. This band is unique in that it degrades to shorter wavelengths in contrast to the other helium bands. This is a manifestation of the fact that the internuclear distance is smaller in the upper state than it is in the lower one. The very intense 5876A atomic
line appears in the midst of the 5885A $3d^2 \frac{3}{2}_{u} - 2p^3 \frac{5}{2}_{g}$ band. At about 5959A the $3d^2 \frac{5}{2}_{u} - 2p^3 \frac{5}{2}_{g}$ band is observed. The first 2nd order atomic line (3187A) appears at 6374A while next to it one observes the $3s \frac{5}{2}_{u} - 2p^3 \frac{5}{2}_{g}$ 6400A band. The lower spectral chart in Figure 11 shows the 2nd order $4p^3 \frac{3}{2}_{g} - 2s \frac{5}{2}_{u}$ 3677A band at 7354A. Several additional 2nd order atomic lines are also indicated.

The molecular notation is discussed in almost any book on the subject and briefly consists of first of all three symbols, for example $4p_{\pi}$, representing the valence electron quantum numbers designating the shell, the orbital angular momentum, and the component of this momentum along the internuclear molecular axis respectively. Next, the molecular symbol $3 \Pi_g$, which follows, contains the information on the multiplicity $2S+1$ in the usual place as a left superscript thereby also designating the spin $S$ while the algebraic sum of the electron orbital angular momenta along the molecular axis is designated by $\Pi$. Also the symmetry with respect to reflection through the center of symmetry of the molecule is given as a right subscript being $g$ for even and $u$ for odd. In addition, for $\Sigma$ states, an upper right superscript of $+$ or $-$ represents even or odd symmetry with respect to reflections in a plane containing the internuclear axis.
Figure 12 shows a schematic of the reported helium 0,0 molecular bands by Weizel (111, p. 262), Dieke (30, p. 648), and Landolt-Bornstein (59, p. 7, 8) with wavelength indications for the transitions observed in the pulsed microwave plasma of this study. The vertical separation between energy levels is only qualitative in this Figure.

Although thermal equilibrium is not probable at early times in this study, nevertheless, rough effective temperatures can be calculated from the positions of the maximum intensity distributions of the P and R branches. According to Herzberg (49, p. 127) $T = \frac{(\Delta \nu_{PR})_{\text{max}}^2}{5.53B}$. Here the factor $(\Delta \nu_{PR})_{\text{max}}^2$ refers to the square of the wave number interval between the intensity maxima of the P and R branches and B is the rotation constant $\frac{h}{8\pi^2cI}$ involving Planck's constant $h$, the speed of light $c$, and the moment of inertia $I$ of the molecule. The temperature $T$ is in °K when $\Delta \nu$ and $B$ are given in cm$^{-1}$. Application of this result to the 6400Å band yields a temperature of 597°C while a similar treatment of the 5733Å band yields 833°C. Average B values of about 7.4 cm$^{-1}$ were taken from Herzberg (49, p. 536). These results may be compared with the rather recent values from Dieke (31, p. 5) of from 580 to 820°C using the 4648Å, $3\pi^3\Pi_g - 2\sigma^3\Sigma_u^+$ band in a capillary type low
Figure 12. Schematic of the 0,0 bands of the helium molecule
frequency helium discharge at 25mmHg. Earlier estimates of from 750 to 1000°K were given by Childs (15, p. 317) while Mulliken (76, p. 403) gives about 700°K. It is generally accepted that such effective temperatures exceed those which would be manifest if thermal equilibrium were to exist (15, p. 309).

Figure 13 shows two scans at 200mmHg with the 2.1 microsecond pulse at a repetition rate of 335 cycles per second and with an average power of 38 watts. This Figure is included to show that intensity levels are reproduced usually to 1 or 2 percent which reflects the stability of the recording arrangement as well as the stability of the pulsed discharge. The prominent lines are identified by wavelength while the bands are marked with asterisks as before. The two scans were made in opposite directions and indicate the largest variation in scanning speed that was encountered. This variation is due mainly to backlash in the grating drive assembly. Notice for example the difference in position of the 7065A line in the two scans. By scanning in the same direction however these effects were significantly reduced so that to the unaided eye duplicated scans could be made to match not only in intensity but also in the position of the lines. However, because of this effect the wavelength determinations were always checked with
Figure 13. Intensity (vs) wavelength for helium at 200mmHg and 38w. This scan shows the intensity reproduction and the maximum observed scanning speed variation due to backlash.
photographic plates where reference could be made with the iron spectral lines.

Figure 14 shows two sections from one of the photographs. The helium spectrum is shown both above and below an iron arc reference spectrum. In the helium spectrum, three exposures were used to observe more clearly both the strong and weak lines. The top line of each section of the film is helium at about 100mmHg and taken with an exposure of 60 minutes while the iron arc exposure on the second line was 0.6 minutes. The third and fourth lines are helium and were exposed at 6 and 0.6 minutes respectively. The third line shows the atomic lines and the stronger band lines while the fourth line shows only the atomic lines. Several of the iron arc lines are indicated by wavelength. The linear dispersion of this Figure is 2.45 angstroms/mm so that the spectral region of the upper half of Figure 14 is from 4730 to 4360A going from left to right while the lower section is from 4030 to 3660A again going from left to right.

Since many times it is convenient to speak in terms of the wave number $\nu$, one can use the relation $d\nu = -d\lambda / \lambda^2$ to find the approximate range in wave numbers represented by 1mm. For the left, middle, and right of the top section of Figure 14, 1mm corresponds to
Figure 14. Typical film record of the helium atomic line and molecular band spectra at several different exposures. The iron arc spectrum is used as a wavelength reference.
intervals of 11, 12, and 13 cm\(^{-1}\) respectively. For the
lower section the corresponding intervals are 15, 17,
and 19 cm\(^{-1}\). This allows one to compare the fundamental
vibrational term interval of approximately 0.2 eV
or 1628 cm\(^{-1}\) (93, p. 70) with the rotational intervals
shown in Figure 14 which are less than the vibrational
intervals by a factor of about 1/100.

3.2 The Intensity Variation with Power

The method of analysing the plasma proceeded along
the following lines. First, the gas was admitted to the
plasma tube at pressures slightly in excess of the pro-
posed maximum values by diffusing the helium through the
hot quartz tube. When thermal equilibrium had been
attained and the desired pressure more carefully approach-
ed, a series of chart recordings were taken at different
power levels after which the pressure was lowered to the
next value and the process repeated. From the charts
thus taken it was possible to plot the power variation of
the intensity for any of the band lines shown in Figure
11. Figures 15, 16, and 17 show the results of these
plots for the most prominent line in the 3677A, 4648A,
5133A, 5733A, 5959A, and 6400A bands respectively at
pressures ranging from 1 to 200 mmHg. One of the most
obvious results of these plots is that at the lowest
Figure 15. Intensity (vs) power curves at indicated pressures for the 3677A band (TOP) and the 4648A band (BOTTOM) with the 2.1 microsecond excitation pulse at 335 pulses per second.
Figure 16. Intensity (vs) power curves at indicated pressures for the 5133A band (TOP) and the 5733A band (BOTTOM) with the 2.1 microsecond excitation pulse at 335 Pulses per second.
Figure 17. Intensity (vs) power curves at indicated pressures for the 5958A band (TOP) and the 6400A band (BOTTOM) with the 2.1 microsecond excitation pulse at 335 pulses per second.
pressures of about 1mmHg, for which it was possible to maintain the discharge, the intensity is nearly independent of the power for all of the observed bands. It is also evident that for the 3677A, 5733A, and 5959A bands at the maximum pressure of 200mmHg the intensity is directly proportional to the power. Again at 200mmHg for the 4648A and 5133A bands the intensity is increasing with power but is leveling off at higher power. It seems that in the case of the 6400A band at 200mmHg the intensity has leveled off when the microwave average power reached 12 watts and the intensity remains constant up to 40 watts.

The slopes of the 3677A, 5733A, and 5959A bands of 0.21, 0.38, and 0.19 taken at 200mmHg indicate a slight tendency for excitation of these three bands with increasing power in the order 5733A, 3677A, and 5959A. It is very possible that were the power range extended, the intensity of these bands would also level off and even perhaps become independent of the power as the 6400A band does.

The fact that the band intensity is independent of the power at low pressures suggests that excited or ionized atom lifetimes are less than the appropriate inverse collision frequency so that the energy is not effectively transferred to the molecular states.
3.3 The Intensity Variation with Pressure

The nature of the pressure variation of the intensity for these same bands for different power levels is shown in Figures 18, 19, and 20. These results each show a characteristic broad maximum which is attained at higher pressures as the power is increased. On the low pressure side, a sharp decrease is observed which is generally in the neighborhood of 10mmHg or less.

It is interesting to see that at the low power of 10 watts the intensity maxima occur at the increasing pressures of about 15, 30, 40, and 60mmHg for the 5733A 4648A, 3677A, and 6400A bands while the 5133A band intensity is so broad that there is only a very slight tendency to decline in intensity as the pressure increases from 50 to 200mmHg.

If one examines Figure 12, the upper levels of these first transitions are all triplets and found to be in the order of $3^3\Delta_u$, $3^3\Pi_g$, $4^3\Pi_g$, and $3^3\Sigma_u^+$. The reason for the rather insensitive intensity response with increasing pressure of the singlet 5133A band which arises from a $3^1\Pi_g - 2^1\Sigma_u^+$ transition is not immediately apparent.

Figures 21, 22, 23, and 24 show a combination of both power (dashed) and pressure (full) variation of intensity for some of the less prominent atomic lines.
Figure 18. Intensity (vs) pressure curves at indicated average power levels for the 3677A band (TOP) and the 4648A band (BOTTOM) with the 2.1 microsecond excitation pulse at 335 pulses per second.
Figure 19. Intensity (vs) pressure curves at indicated average power levels for the 5133A band (TOP) and the 5733A band (BOTTOM) with the 2.1 microsecond excitation pulse at 335 pulses per second.
Figure 20. Intensity (vs) pressure curves at indicated average power levels for the 5958A band (TOP) and the 6400A band (BOTTOM) with the 2.1 microsecond excitation pulse at 335 pulses per second.
Figure 21. Intensity (vs) power curves (dashed) at indicated pressures and intensity (vs) pressure curves (full) at indicated average power levels for the singlet 4922A (4D-2P) atomic line.

Figure 22. Intensity (vs) power curves (dashed) at indicated pressures and intensity (vs) pressure curves (full) at indicated average power levels for the singlet 5047A (4S-2P) atomic line.
Figure 23. Intensity (vs) power curves (dashed) at indicated pressures and intensity (vs) pressure curves (full) at indicated average power levels for the triplet 4026A (5D-2P) atomic line.

Figure 24. Intensity (vs) power curves (dashed) at indicated pressures and intensity (vs) pressure curves (full) at indicated average power levels for the triplet 3889A (3P-2S) atomic line.
This study is not specifically concerned with the atomic lines since their behavior has been reported by Tynes (103, p. 54-121). For this reason and also since the atomic lines are much more intense than the bands, the more prominent atomic lines were allowed to go off-scale in order to use a chart recording range sensitive enough to show the weaker band detail. However, for reasons of contrast and for comparison with the bands, the response of some atomic lines are included in these Figures. A complete pressure range of observations was not possible in the case of the 3889A and 4922A lines since they exceeded the maximum scale reading at the lower pressures where the atomic lines are very intense in comparison with the molecular bands.

Again from Figures 21, 22, 23, and 24 for the atomic lines, as was shown for the molecular bands, it is evident that the intensity is directly proportional to the microwave power at the higher pressures while on the other hand the pressure variation of the intensity indicates an inverse type dependence with increasing pressure at all power levels.

An examination of the pressure variation of the relative intensity of the 4922A (4D-2P) atomic line to the 5047A (4S-2P) atomic line at given power levels shows the former to be more intense by about a factor of
seven. Slight variations of this factor do occur with pressure but this is one indication of the tendency to populate the higher angular momentum states under the conditions of this experiment.

While the intensity information on the preceding pages is subject to some criticism since there is no time resolved presentation of a very time dependent event, nevertheless, one should be able to draw some conclusions concerning major effects connected with the afterglow. This is perhaps more true of the bands than of the atomic lines since in the latter case a large fraction of the total light emitted appears during the incident pulse period while for the molecular bands the preceding figures deal principally with the afterglow since the molecular emission during the pulse period tends to be quenched as discussed in section 4.2. It should be mentioned that although the line intensity is generally high at low pressure, it does drop sharply in the 1mmHg region where it becomes difficult to sustain the discharge. The marked pressure difference between the atomic line behavior as opposed to the bands makes it very easy to separate the two on the chart recordings providing several chart recordings are available at different pressures. While a given atomic line may be the more intense in the low pressure region, they decrease
in intensity as the pressure is increased while at the same time the intensity of the band lines increases. Consider for example the 2nd order 3187A atomic line and the most intense line in the 6400A band (39 watts) as shown in Table II. One may check the arbitrary intensities of the line and the band at 60mmHg as shown in Figure 11. Due to the mounting of the trimmed chart recording in Figure 25 the atomic line intensity value of 74 cannot be read, but the band intensity of 13 is observable.

**TABLE II**

**COMPARISON OF THE 3187A ATOMIC LINE AND THE 6400A BAND**

<table>
<thead>
<tr>
<th>Pressure (mmHg)</th>
<th>3187A Line Intensity (arbitrary)</th>
<th>6400A Band Intensity (arbitrary)</th>
<th>Relative Intensity (Line per Band)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>off-scale</td>
<td>6</td>
<td>----</td>
</tr>
<tr>
<td>10</td>
<td>off-scale</td>
<td>12</td>
<td>----</td>
</tr>
<tr>
<td>20</td>
<td>74</td>
<td>13</td>
<td>5.69</td>
</tr>
<tr>
<td>40</td>
<td>50</td>
<td>20</td>
<td>2.50</td>
</tr>
<tr>
<td>60</td>
<td>29</td>
<td>21</td>
<td>1.38</td>
</tr>
<tr>
<td>80</td>
<td>27</td>
<td>26</td>
<td>1.04</td>
</tr>
<tr>
<td>100</td>
<td>20</td>
<td>22</td>
<td>0.91</td>
</tr>
<tr>
<td>150</td>
<td>14</td>
<td>20</td>
<td>0.70</td>
</tr>
<tr>
<td>200</td>
<td>10</td>
<td>17</td>
<td>0.59</td>
</tr>
</tbody>
</table>

Figures 25 and 26 are similar to Figure 11. They are included to show the behavior of the bands and lines at the lower pressures of 20 and 5mmHg respectively. All of the previously named bands are marked as before and
Figure 25. Intensity (vs) wavelength for helium at 20 mmHg, 38w. Peak band lines are marked with asterisks.
Figure 26. Intensity (vs) wavelength for helium at 5 mmHg, 40w. Peak band lines are marked with asterisks.
in addition the weaker atomic lines are added as they appear at the lower pressures. One example of this is the singlet 5D--2P atomic line at 4388A. In Figure 11 at 60mmHg it has the appearance of one of the band lines to the left of the 4472A line. In Figure 25 at 20mmHg it has an arbitrary intensity of 31 and is off-scale in Figure 26 at 5mmHg. Notice that the band lines which surround it steadily decrease in this sequence of figures. The inclusion of these figures is sufficient to illustrate the source of the data in Table II and also the intensity variation with pressure plots shown in Figures 18-24.
IV. THE INTENSITY VARIATION WITH TIME

4.1 Introduction

With the discarding of the more elaborate time resolved chart scanning approach the only remaining method available was a direct photomultiplier tube response of the optical events. This response was displayed, observed, and photographed from the screen of the oscilloscope. This is essentially the same approach previously used by Tynes (103, p. 35-45) to study the atomic lines with the exception that in the case of the molecular radiation it was necessary to resort to the use of the monochromator with a reduced linear dispersion. In this way one could easily obtain sufficiently large signals from the photomultiplier tube by including the entire band system instead of a single band line.

As previously mentioned this approach does not supply the detail which was initially desired and in some cases it was difficult to exclude weak atomic lines which happened to be near or mixed with the bands. In spite of the experimental difficulties, however, the information on the time resolve band behavior is more specific and complete than any available thus far for the pulsed microwave plasma. It should be remembered that the band detail shown in Figures 11, 13, 25, and 26
was obtained not with the monochromator but with the Jarrell-Ash spectrograph together with the Varian chart recorder which was sensitive enough to respond to single band lines for intensity (vs) wavelength data. With the monochromator and the time presentation on the oscilloscope this wavelength resolution detail was lost with the gain in intensity as mentioned above.

In addition to the time response of the bands themselves, the time behavior of the excitation pulse together with the reflected and transmitted pulses are included in section 4.4 where the correlation of these pulses with the light intensity is also discussed. Typical values of the delay in breakdown in the plasma tube after arrival of the excitation pulse range from 50 to 70 nanoseconds when measured from the 10% peak power incident pulse time mark to the 10% light response time mark. The light response rise time ranged from 150 to 250 nanoseconds in the 5 to 100mmHg pressure range when an average incident microwave power of 40 watts was used.

Measurements of the incident, reflected, transmitted, and loss power for a plasma tube pressure of 88mmHg are shown in Table III. Deviations from the average values are + 5.3 and - 7.7% for the reflected, + 63 and - 47% for the transmitted, and + 1.4 and - 1.7% for the loss.
TABLE III
POWER MEASUREMENTS OF THE AVERAGE INCIDENT, REFLECTED
TRANSMITTED, AND LOSS

<table>
<thead>
<tr>
<th>Incident level (watts)</th>
<th>Reflected level (watts)</th>
<th>% of Incident</th>
<th>Transmitted level (watts)</th>
<th>% of Incident</th>
<th>Loss level (watts)</th>
<th>% of Incident</th>
</tr>
</thead>
<tbody>
<tr>
<td>45.2</td>
<td>11.0</td>
<td>24.3</td>
<td>2.20</td>
<td>4.86</td>
<td>32.0</td>
<td>71.1</td>
</tr>
<tr>
<td>39.2</td>
<td>9.4</td>
<td>24.0</td>
<td>1.51</td>
<td>3.85</td>
<td>28.3</td>
<td>72.2</td>
</tr>
<tr>
<td>32.3</td>
<td>8.1</td>
<td>25.1</td>
<td>1.07</td>
<td>3.32</td>
<td>23.0</td>
<td>72.0</td>
</tr>
<tr>
<td>27.0</td>
<td>6.6</td>
<td>24.4</td>
<td>0.66</td>
<td>2.44</td>
<td>19.7</td>
<td>72.9</td>
</tr>
<tr>
<td>21.4</td>
<td>5.2</td>
<td>24.3</td>
<td>0.40</td>
<td>1.87</td>
<td>15.7</td>
<td>73.3</td>
</tr>
<tr>
<td>15.4</td>
<td>4.0</td>
<td>26.0</td>
<td>0.26</td>
<td>1.68</td>
<td>11.1</td>
<td>72.1</td>
</tr>
<tr>
<td>10.0</td>
<td>2.6</td>
<td>26.0</td>
<td>0.24</td>
<td>2.40</td>
<td>7.2</td>
<td>72.0</td>
</tr>
<tr>
<td>4.4</td>
<td>1.0</td>
<td>22.7</td>
<td>0.23</td>
<td>5.22</td>
<td>3.2</td>
<td>72.6</td>
</tr>
</tbody>
</table>

Average Values 24.6 3.20 72.3

The size of these deviations from the average suggest that one look at plots of the reflected, transmitted, and loss percent power levels against the incident power level as shown in Figure 27. It is observed that the reflected and loss levels oscillate 180° out of phase while the transmitted level first drops and then rises with increasing incident power. The transmitted power has a broad minimum for an incident level of about 18 watts.

Using the 40 watt average incident power as an upper limit we can estimate a corresponding limit on the ionization rate of the gas. Suppose we use the ionization potential of 24.6 volts for helium and assume that all of the power produces ionization. The 24.6 electron
Figure 27. % of the incident power lost, reflected, and transmitted as a function of the average incident power.
volts is equivalent to $39.4 \times 10^{-12}$ ergs while 40 watts is equivalent to $40 \times 10^7$ erg/sec so that about $10^{19}$ ions/sec could be produced. Table III shows that a more realistic power level would be about 72% of the 40 watt average incident power level or about 29 watts. This leads to a maximum ion production rate of about $7.4 \times 10^{18}$ ions/sec. On the other hand, excitation is also occurring. A reasonable estimate is that the power is roughly divided in exciting and ionizing the gas. We therefore arrive at about $3.7 \times 10^{18}$ ions/sec for an estimate of the maximum time average rate of production of primary helium ions. In section 2.3 the pulse power level is given as 1215 times the average level in the case of the 0.6 microsecond pulse and 1330 times the average level for the 2.1 microsecond pulse. This yields pulse ionization rates of about $4.5 \times 10^{21}$ ions/sec for the duration of the short pulse and $4.9 \times 10^{21}$ ions/sec for the duration of the 2.1 microsecond pulse. Total probable primary ion production per pulse then becomes about $3.7 \times 10^{15}$ ions in the case of the shorter pulse and $11 \times 10^{15}$ ions for the longer one.

4.2 The More Intense Bands

Typical band response is shown in Figure 28 where one observes from top to bottom the 6400A, 5958A, 5733A,
Figure 28. Intensity (vs) time characteristics for the 6400A, 5958A, 5733A, and 4648A bands.
5133A, and 4648A band intensity (vs) time characteristics. The average power is 39 watts with the 2.1 microsecond pulse. The sweep speed is 2 microseconds per division while the oscilloscope sensitivity is 100 millivolts per division with 20db external attenuation. For comparison, Figure 29 shows several atomic lines with the same excitation pulse and oscilloscope sweep speed as before and at an average power of 30 watts. Additional remarks concerning the atomic lines are found in section 4.3.

Since the oscilloscope sweep was triggered by the arriving microwave pulse, Figure 28 shows that relative to a microsecond time scale the molecular radiation begins immediately with about 1/5 of one division or 0.4 microseconds from breakdown being required for an equilibrium condition to be attained. This delay is assumed to reflect the time required to produce an equilibrium concentration of radiating molecules. Notice that in contrast to the atomic line emission in Figures 29 and 31 the molecular radiation does not exhibit the extreme shoulder behavior, i.e. the fast rise and slower decay in light intensity during the microwave pulse. The molecular radiation is, however, distinctly present not only in the afterglow as reported by Tynes (103, p. 92, 203) but also at reduced levels during the incident pulse period itself. The time required for the molecular
Figure 29. From top to bottom the light intensity as a Function of time for the 5016A, 5876A, 6678A, 6563A, and 4472A atomic lines at an average power of 30 watts.
emission to reach equilibrium is very nearly equal to the time required for the atomic emission to decay from the initial light intensity spike.

Several possible schemes for obtaining the molecules can be considered as shown in Table I. The actual production rates must be rather fast, however, on the micro-second time scale since the molecular production and radiation are certainly not primary processes. For example, upon excitation the helium atoms can produce molecular ions either through collisions with normal atoms or with collisions with other excited atoms as indicated in processes 1 and 2 of Table I. In this experiment, the fast production of molecular radiation as shown in Figure 28 is in agreement with results of Hornbeck and Molnar who have reported (52, p. 625) from electron beam studies together with mass spectrographic measurements that molecular ions can appear in less than one microsecond after atomic excitation.

The possibility of molecular ion production through the interaction of a normal and an excited atom not necessarily a metastable state does allow molecular ion formation with the excitation of the helium atoms at the very beginning of the microwave pulse. However, because of the short lifetime of the atomic levels as listed in Appendix C, this contribution to the molecular radiation
would require high concentrations of excited atoms. The behavior of the molecular radiation at the end of the excitation pulse should give some indication of the importance or lack of it for this reaction. At any rate, the molecular ions when produced could then recombine with electrons into different radiating molecular states. The lifetimes of such states with the exception of the reported (91, p. 1307) 0.05 seconds of the $^3\Sigma_u^+$ metastable level will be assumed to be negligible with respect to a microsecond time scale so that subsequent radiation would be observed almost immediately.

It is well known (14, p. 1391) that recombination is greatly inhibited by the exciting field as a result of the increased electron energy. This could explain the relatively low intensity of the bands during the pulse as compared with the afterglow level since Severin (95, p. 129) has reported that even 100 microwatts of microwave power incident upon a helium glow plasma produced intensity quenching of from 20 to 50% for most helium atomic lines and between 50 and 80% for all helium molecular bands. The afterglow molecular radiation should also be attributed to recombination of the molecular ions and the electrons with the approximately 0.2 microseconds delay between the end of the microwave pulse and the maximum afterglow light intensity peak.
reflecting the electron cooling time. Aside from the
lack of a sharp initial light spike, in the case of the
bands, another significant difference between the band
behavior in Figure 28 as compared with the atomic lines
in Figures 29 and 31 can be made by noticing that the
intensity of the bands does not drop to a near zero inten-
sity at the end of the excitation pulse as is the tendency
for the atomic radiation. That there is such a difference
between the two cases just at the end of the excitation
pulse suggests that there is also a difference in the
processes which eventually produce the excited atoms and
molecules. Since the atomic light intensity drops sharply
at the end of the incident excitation pulse, this part of
the helium atomic radiation during the latter part of the
incident pulse must derive its energy directly from the
electrons which are energized by the microwave electric
field. Evidently in the case of the molecules the
cessation of the electric field as the incident pulse
ends enhances the light emission rather than supresses it.
Thus the molecular radiating processes do not arise
directly from the energetic electrons but are expected to
be essentially the same in both the incident pulse
period and the afterglow while the opposite is expected
in the atomic emission.

The fact that the molecular radiation does not
drop significantly at the end of the excitation pulse indicates that the molecular ion density is saturated or that the production from short lifetime excited atomic states as in process 1 of Table I is not important.

The atomic emission is more involved than the molecular emission not only for drop in light intensity at the end of the pulse, but also because of the light spike in the early part of the excitation pulse period.

Thus one is concerned in the case of the helium atoms with the initial rise in the light intensity to the sharp spike with its subsequent decay to the shoulder for the duration of the incident excitation pulse. Also, after the decay to the shoulder, the atomic light emission seems to be strongly dependent upon the shape of the power pulse since both the light and the excitation pulses decay at the same time. In addition to this, the atomic afterglow is certainly not due directly to any incident power but must come from energy sources produced during the power pulse and so must also be considered separately. These points are discussed further in connection with experimental results shown in following sections.

Still another possibility to consider in the explanation of the molecular radiation is that excited molecules rather than molecular ions could be produced in the interaction of the excited and normal helium
atoms. This would eliminate the time required for secondary production processes and thus speed the appearance of the molecular radiation in accordance with the observations of Figure 28. The results obtained at this time do not allow a determination of this possibility. In the late afterglow the emphasis has been placed on the molecular ion production and subsequent recombination.

Figure 30 shows the results of plotting both log \( I \) (horizontal marks) and then \( I^{-1/2} \) (vertical marks) as a function of the time into the afterglow for the bands shown in Figure 28. The log \( I \) data should yield a straight line when plotted against the time if diffusion losses predominate while on the other hand the plot of \( I^{-1/2} \) against the time is used to identify losses controlled by recombination (103, p. 69, 76).

At this early stage of the afterglow and where the time range is so limited, neither of the plots should be judged to be decisive. However, some trends do appear which cannot be overlooked. First, in the log \( I \) plot for all bands, the early initial afterglow decay does not even suggest linearity while for times after 5 microseconds from the end of the exciting power pulse the points do begin to lie on a straight line. For the \( I^{-1/2} \) plots the reverse is true. That is the linearity is very good up to about 5 microseconds from the end of
Figure 30. Log I (vs) t for the bands listed (horizontal) and $1/(I)^{1/2}$ (vs) t for the same bands (vertical).
the excitation pulse and sometimes longer. This is similar to the afterglow results of Tynes (103, p. 85) for the atomic lines with the exception that he does not plot \( \log I \) as a function of time.

Notice in Figure 30 the distinction in the \( I^{-1/2} \) plots of the 5733A band as compared with the other bands shown. This \( 3d^2\Delta_u \rightarrow 2p^3\Pi_g \) transition shows a distinct tendency to exhibit non-linearity of the \( I^{-1/2} \) plot before any of the other bands. The reason for this response is not known at this time.

These observations suggest that one look for heavy recombination type losses at the beginning of the afterglow but also consider the possibility of diffusion losses at later times. The nonlinearity of the diffusion plots at the beginning of the afterglow is almost a necessity since at rms speeds of the order of \( 2 \times 10^5 \text{cm/sec} \), typical for helium at the temperatures in this experiment, about 5 microseconds are required for the particles to travel 1 cm where wall losses can begin.

This suggestion in Figure 30 of diffusion losses early in the afterglow together with the corresponding early decay of the light intensity is the afterglow could easily mean that wall reactions are important in this particular experimental arrangement, especially as
a third body in some of the reactions listed in Table I.

4.3 Intensity Behavior for Several Atomic Lines

Figure 31 shows a very important series of plots of the light intensity as a function of the time for the triplet 4472A (4D-2P), singlet 4922A (4D-2P), singlet 5016A (3P-2S), triplet 5876A (3D-2P), and the triplet 3889A (3P-2S) atomic lines from top to bottom and at pressures of 100, 50, 25, and 5mmHg from left to right. The intensity units are arbitrary and different for each individual line but are constant for each line as the pressure is varied. The sweep speed is two microseconds per division.

One observes a distinct difference upon comparing the initial light intensity peak occurring during the power pulse with the afterglow peak and its subsequent decay in the D to P transitions as exhibited by the 4472A, 4922A, and 5876A lines as opposed to the P to S transitions of the 5016A and 3889A lines. At the higher pressures the afterglow peak height is of the same order as the initial peak and occasionally higher for the first group while for the latter two lines the afterglow is much weaker by comparison. The behavior is qualitatively the same at the lower pressures. Thus one concludes that the number of P to S transitions in the
Figure 31. Intensity (vs) time for the triplet 4472A (4D-2P), singlet 4922A (4D-2P), singlet 5016A (3P-2S), triplet 5876A (3D-2P), and triplet 3889A (3P-2S) atomic lines from top to bottom respectively and at pressures of 100, 50, 25, and 5mmHg from left to right.
afterglow is less than the number of D to P transitions. Any theoretical discussion of the afterglow should on the basis of these results indicate preferential population of the D over the P states in the n equal 3 and 4 levels.

The observations of Figure 31 experimentally confirm the theoretical studies of collision transfer by St. John and Fowler (101, p. 1813) and others. This collisional transfer of helium excitation levels which is also reported by Lin and Fowler (61, p. 468) assumes that n^1P states previously excited through electron-atom collisions are transferred to nF states upon collision with normal helium atoms. They find by approximate theoretical methods that for the F states the total spin S is no longer a constant of the motion and therefore the distinction between singlet and triplet states is no longer meaningful. Population of particularly the triplet levels can therefore be accomplished without violating Wigner's spin conservation rule providing the transfer occurs where the electron orbital angular momentum is greater than or equal to 3\(^s\). Now if the n^1P excitation is transferred by collision processes to n^3F states which cascade to the 3^3D level, one would expect to observe a preponderance of D to P transitions as opposed to P to S transitions
just as shown in Figure 31. Since this is a collision transfer process it should also have some pressure dependence so that one would expect the distinction between the D to P and the P to S transitions to be less important at some lower pressure. The results of Figure 31 do show a reduction in the afterglow light intensity maxima relative to the excitation pulse period light intensity maxima as the pressure is reduced. They do not, however, show much distinction in this effect between the D to P transitions as opposed to the P to S transitions.

Another observation which should be made from Figure 31 concerns the intensity of the shoulder immediately following the first peak and which lasts for the duration of the excitation pulse. At high pressures the shoulder is low, well formed, and nearly horizontal for all of the atomic lines while as the pressure is decreased the shoulder intensity rises with respect to the initial peak occurring during the power pulse and at the same time begins to slope down more until at the end of the power pulse the light intensity decays sharply.

This description is best illustrated by the 5876A line but is generally true for all of the lines in the Figure 31. Thus while a distinct difference exists in the afterglow between the D to P and the P to S transitions, during the excitation pulse the general
behavior of the shoulder is very much the same, yielding increased intensity at the lower pressures. This of course indicates a significant difference in the excitation mechanisms for the two periods and will be discussed again later. At the lower pressure the relation between the shoulder and the afterglow is very similar for all the transitions shown.

During the pulse itself the electron impact excitation seems to populate the low lying states in roughly the same manner while the intensity increase of the shoulder with decreasing pressure shows that not only is there a possibility of collisional shift from P to perhaps F states but that collisions actually are depleting the source of the atomic line emission during the later part of the incident pulse period.

The cause of the light spiking and subsequent shoulder behavior seems to be rather non-selective in terms of the energy levels in its action. Bakakina (5, p. 413) in a preliminary report of a high frequency coaxial discharge in helium observed spiking at pressures above 10mmHg and suggested that the light intensity decrease to the shoulder was due to an increase in the electron temperature or a change in distribution.

Tynes (103, p. 134) discounts the possibility of decreased recombination of the ionized helium atoms
with electrons as a result of increased electron temperature with time during the power pulse and attributes (103, p. 166) the initial spiking of the light intensity to microwave reflection and consequent isolation of the plasma as the electron density builds up. He also considered the possibility of the production of helium molecule ions from excited helium atoms (103, p. 61, 165) thus depleting the density of excited atomic states and consequently reducing the atomic emission and concluded that this was not the reason for the drop in light intensity to form the shoulder during the early part of the power pulse. Another interesting result of Tynes' study is (103, p. 190) that the theoretical electron temperatures at the end of the pulse are higher at the low incident power levels than at the higher ones. This was attributed to more complete plasma isolation with increased reflection at higher power levels. Another possibility is that at higher powers more metastables are produced which are then ionized by lower energy electrons. If the metastable density is high, this could prevent the electron energy from increasing as much as at lower power levels where the metastable density is lower.

Before the results of this experiment can be used to discuss these problems it is necessary to refer to
some additional photographic measurements. In the next section detailed oscillographic traces of the time behavior of the incident, reflected, and transmitted pulses are shown.

4.4 Light Intensity Correlation with the Excitation, Reflected, and Transmitted Pulses.

Figure 32 (LEFT) taken at 40.5mmHg is typical of an entire series of plates which are shown later in section 4.5 of the inverted pulse height as a function of time for pressures ranging from 220 to 10mmHg and includes the incident, reflected, and transmitted pulse shapes in A, B, and C for the 2.1 microsecond pulse. D, E, and F refer to the incident, reflected, and transmitted pulse shapes for the shorter 0.6 microsecond excitation pulse.

The normal sweep speed is 0.5 microseconds per division but in faster second exposures the leading edge of the pulse is expanded at 0.02 microseconds per division for A and D and at 0.05 microseconds per division for the rest. Since, for example, the transmitted spike is so sharp, one needs these fast second exposures to explain the pulse profile. Average power levels are 37 watts for the 2.1 microsecond pulse and 41 watts for the shorter 0.6 microsecond excitation pulse.

The incident pulse was obtained from the directional
Figure 32. (LEFT) Inverted pulse height (vs) time at 0.5 microseconds/division for the incident, reflected, and transmitted 2.1 microsecond pulse for A, B, and C. Similarly with the 0.6 microsecond pulse for D, E, and F. The fast second exposure sweep speeds are 0.02 microseconds/division for A and D with 0.05 microseconds/division for the others. (RIGHT) Light intensity (5876A) and inverted transmitted pulse (vs) time at 0.5 microseconds/division and 37, 20, and 10 watts with the 2.1 microsecond pulse for A, B, and C. Similarly at 41, 20, and 10 watts with the 0.6 microsecond pulse for D, E, and F.
coupler following the power divider as shown in Figure 6 with a total attenuation of 60db and an oscilloscope sensitivity of 100 millivolts per division. The reflected pulse was obtained from the directional coupler following the ferrite isolator with a total attenuation of 70db and an oscilloscope sensitivity of 100 millivolts per division. The transmitted pulse was obtained from the directional coupler following the plasma tube section with the same attenuation and oscilloscope sensitivity as the reflected pulse.

Both the 2.1 microsecond and 0.6 microsecond incident pulses from the 725-A magnetron exhibit 10% to 90% rise times of about 10 nanoseconds as shown in the fast second exposures of traces A and D on the left of Figure 32. The longer incident pulse in A maintains a level of about 80% of its maximum value for a time of 2.1 microsecond pulse. In contrast to this the shorter pulse in D maintains a level of 90% of its maximum value for a time of 0.6 microseconds and is most often referred to as the 0.6 microsecond pulse. It is estimated that an equivalent rectangular power pulse would have a duration of 2.3 microseconds in the case of the longer pulse, and 0.85 microseconds for the shorter one.

Practically every detail of the incident pulse is preserved in the reflected part while quite the opposite
is true for the transmitted pulse. This is quite contrary to the assumption of Tynes (103, p. 166) that the decay of the initial light spike to form the shoulder during the power pulse is a result of microwave power reflection from the plasma as the electron density builds up. If this were the case then one would expect to see a correlation of the light intensity and the reflected pulse which is not observed. Instead, what is observed is a very good correlation of the light intensity and the transmitted pulse but in an opposite sense.

In the latter case and for the longer incident power pulse on the left of Figure 32 in C, a 10.5 nanosecond (halfwidth) pulse penetrates the helium gas as the discharge occurs or while the plasma is being produced. Immediately after this pulse penetration, the plasma becomes practically opaque to the microwaves for a period of about 0.3 of a microsecond after which it tends to relax slightly and some transmission is observed for the duration of the power pulse. Notice that essentially the same action is exhibited by the transmitted pulse in F for the shorter 0.6 microsecond power pulse on the left of Figure 32.

This observation lends more support to the idea that as the plasma is being produced, the initial breakdown does not represent the steady-state condition. As a
steady state condition is achieved in the discharge tube, the shoulder effect is observed in the light intensity and the plasma becomes partially transmitting for the duration of the power pulse.

Figure 32 (RIGHT) was taken at 20mmHg and emphasizes the correlation of the light intensity and transmitted pulses mentioned in the preceding paragraph. The light pulse is from the 5876A atomic line and exhibits behavior typical of the other atomic lines while the transmitted portion of the microwave excitation pulse is inverted and due to improper exposure is missing the initial sharp transmitted spike in A and B. A, B, and C represent the 2.1 microsecond pulse at average incident powers of 37, 20 and 10 watts respectively. D, E, and F represent the 0.6 microsecond pulse at average incident power of 41, 20 and 16 watts respectively. The sweep speed is 0.50 microseconds per division in all cases.

Notice in A, B, D, and E that reducing the power to one half of its original value does little to the light intensity of either the 2.1 or 0.6 microsecond pulses during the discharge period itself. At the same time with both pulse lengths the transmitted power is reduced to much less than half of its initial value. At the lowest power setting in C and F the light intensity during the discharge period begins to fall as one would
expect, however, more significantly in the case of the longer pulse than the shorter one. The transmitted power for C and F seems limited to the early transmitted spike for both pulse lengths at these low powers. These results seem to indicate that there is an adjustment or relaxation of the plasma so that during the power pulse the optical radiation does not always follow the approximately 72% loss to the plasma of the incident microwave power as has been shown in Figure 27. This could very well be a result peculiar to this particular experimental arrangement, although the shape of the light intensity curves are practically the same as reported by Bakakina (5, p. 413) and Janin and Eyraud (53, p. 1073) under two additional different experimental arrangements of the plasma tube.

A semi-quantitative picture of the electron density may be obtained from the transmitted pulse as follows. When the excitation pulse reaches the plasma tube very little attenuation in the transmitted pulse is observed until breakdown. At this time the transmitted pulse goes essentially to zero. Now if one assumes that the transmission and reflection effects can be discussed on the basis of the plasma frequency \( \omega_p \) given by \( \omega_p = \left(\frac{ne^2}{m}\epsilon_0\right)^{1/2} \) where \( n \), \( e \), and \( m \) are the electron density, charge, and mass respectively, and \( \epsilon_0 \) is the permittivity
of free space then the microwave power can be expected to be transmitted until the electron density reaches the critical value of \( w^2 n \varepsilon_\omega / \varepsilon_0^2 \) with \( n, \varepsilon_\omega, \) and \( \varepsilon \) defined as before and where \( w \) is the angular frequency of the incident microwaves. This relation for the electron density may be put into the form of \( f = 8980 n^{1/2} \) when cgs units are used and \( f \) is the frequency of the incident microwaves related to \( w \) by the relation \( w = 2\pi f \).

In this experiment for the critical case where the plasma frequency \( w_p \) is numerically equal to the microwave angular frequency \( w \), then \( n = n_c = 1.09 \times 10^{12} / \text{cm}^3 \). After about 0.25 microseconds the electron density appears to decrease below this critical value as the plasma relaxes and remains essentially constant during the duration of the pulse at a value of about \( 10^{11} / \text{cm}^3 \) or at least low enough to allow the observed low level transmission of power through the plasma.

Another significant result of Figure 32 (RIGHT) is evident in the afterglow period of the oscillographic traces. Here in contrast to the pulse period the reduction of the afterglow intensity seems to follow the incident power reductions quite evenly for both the longer and shorter incident pulses. In other words, even though the light intensity during the pulse does not always decrease with the incident power level the light
intensity during the afterglow which is dependent on the pulse period excitation processes does decrease with the incident power level. This is a very interesting effect since for a given atomic transition an increase of power during the pulse period has little effect on the light intensity during that pulse period once one exceeds a certain power value but the afterglow which depends upon what has happened during that pulse period does increase with an increase of power over the entire power range. This really is the result of several energetic processes occurring at once where the storage of energy during the pulse period, which results in the afterglow light intensity, builds up with an increase of power while the actual light emission or excited atomic state density during the pulse seems limited after a certain power level is reached. This of course also indicates that the main mechanism of light emission is different for the two time periods. It is generally believed that the afterglow is connected with the production of metastable atoms during the power pulse. If this is true, then Figure 32 (RIGHT) would seem to indicate that during the power pulse, there is a shift from excited atom production to metastable atom production as the power is increased.

It should be mentioned that Figure 32 also seems to
offer evidence of better correlation of the initial light spiking followed by the shoulder effect with the transmitted pulse rather than with the reflected pulse as given by Tynes (103, p. 166). Notice that in say B and E on the left which are reflected pulses which have been synchronized with the incident pulse that on the microwave time scale at least there is no significant qualitative difference between the reflected and incident pulses. On the other hand if one observes A, B, D, or E on the right where it is possible to observe the light intensity and the corresponding transmitted pulse at two different power levels and for two different pulse lengths, there is the indication that during the initial sharp spiking of the light intensity the microwave power is not being transmitted and, if one remembers again the left side of the same Figure, it is not being reflected any differently during the shoulder formation of the light intensity immediately following the sharp spiking either. As the light intensity decays from the spike to the shoulder one observes again in A, B, D, or E on the right that the microwave power is correspondingly being partially transmitted for the duration of the pulse. This indicates that the explanation of the light spiking in terms of microwave reflection as given by Tynes and referred to above is not consistent with a close
correlation of the incident, reflected, and transmitted microwave pulses. It seems more likely that there is a relaxation or change of distribution of the plasma itself which presents on the other hand no significant change in the condition of the plasma with time to the incident microwave pulse but allows less total power absorption by the plasma with time so that some power is transmitted by the plasma at about the same time the light intensity decays and the shoulder forms. It should also be remembered that Figure 31 shows the change with pressure of the shoulder indicating the importance of collisions in depleting the population of the atomic states.

Finally, Figure 32 (LEFT), lines C and F show an effect accentuated by the larger plasma tube of this study as compared with that used by Tynes (103, p. 66) At low power (10 watts average) one observes a sharp light intensity spike synchronized with the transmitted spike. It is very probable that this results from metastable atoms still in existence from the previous pulse occurring at times of the order of milliseconds earlier. This early light spike is present in A, B, D, and E also, but the main pulse light intensity occurs so fast that it is not easy to separate the two. At lower powers in C and F the main pulse light intensity is delayed and separation is good. There is also a pressure effect of
this early light intensity spike which is reported in connection with Figure 33 and 34.

Figures 33 and 34 show the time resolved intensity of the integrated light and the incident inverted microwave excitation pulse in A together with an assortment of lines and bands in B through F over a wide range of gas pressures for the 2.1 microsecond excitation pulse length. The integrated light in A is the photomultiplier response for the entire light output including bands as well as atomic lines as a function of time. The double exposure of this integrated light and the inverted incident microwave excitation pulse allows a time correlation of the events which are recorded. Notice the inclusion of both the more intense incident as well as the lower level reflected microwave pulses in trace A at 99mmHg in Figure 33. Once again it is obvious that except for the lower level of the pulse, the reflected and incident pulses behave qualitatively the same.

For the atomic line emission as well as the integrated light there is a very sharp initial light spike corresponding in time to the arrival of the microwave excitation pulse as shown in A, B, and E at 99mmHg in Figure 33. Figure 32 shows a very remarkable time coincidence of this initial sharp light spike with the sharp transmitted pulse of microwave power which
Figure 33. Integrated light intensity and inverted 2.1 microsecond excitation pulse (A) and selected lines and bands at the indicated pressures.
Figure 34. Integrated light intensity and inverted 2.1 micro-second excitation pulse (A) and selected lines and bands at the indicated pressures.
evidently means that at the beginning and for a very small fraction of the incident pulse, the resulting increased temperature of the residual electrons increases the optical radiation of the atomic lines. Tynes (103, p. 203) mentions a light spike similar to the ones shown in Figures 33 and 34 and attributes it to the molecular radiation. Figure 33 shows that this is not the case and that it is associated with every atomic line shown instead. It is possible that in an attempt to increase the light received by the photomultiplier the slits were opened wide enough to allow atomic as well as molecular radiation in Tynes' observations. He suggests, in explaining this very early light spike, the possibility of an optimum recombination electron temperature for the molecule ion which is slightly above the electron temperature between pulses. Whatever the process is, it is one which is dependent upon the incident microwave pulse and coincides almost exactly with the part of the incident pulse which is transmitted through the gas sample as shown in Figure 32.

Notice in A of Figures 33 and 34 the changing characteristic of this early initial light spike. At 221mmHg it is just visible if you know where to look for it and becomes more and more prominent as the pressure is reduced until at 99mmHg it is more than half as high
as the secondary light intensity peak which follows it during the incident power pulse period. As the pressure is reduced still further the initial light intensity peak increases in intensity while the secondary peak which follows it is advanced in time so that at the lowest pressures shown the two intensity peaks are hardly resolved in the Figures. Close study of A at 40.5 and 19mmHg show that the effect is still present even though it is difficult to resolve the two. It is possible to measure the time between the two peaks from these Figures so that at 221mmHg they are separated by about 0.47 microseconds while at 40.5mmHg the separation is about 59 nanoseconds and still less at 19mmHg.

The decay of the secondary light intensity peak during the excitation pulse period to form the shoulder is evident at all pressures for the integrated light in A, the 5876A atomic line in B and the other atomic lines in D, E, and F. At all pressures lower than about 200mmHg this shoulder or relatively stable pulse period radiation never has a completely horizontal or fixed intensity but decays very slowly until the end of the microwave incident power pulse where it then essentially decreases with the latter as the power pulse ends.

One observes a further correlation in time if a comparison is made of the line and band intensity during
the excitation pulse. At 221mmHg, especially in D of Figure 33 one sees the band intensity rise slowly and reach a relatively flat region after about one division or 0.5 microseconds. This corresponds to the time during the excitation pulse when the atomic line intensity has just reached its secondary maximum and is beginning to decay to the shoulder. As the pressure is decreased the band intensity equilibrium during the power pulse is advanced in time just as was the case of the initial atomic light intensity spike in a preceding paragraph. This is evident in Figure 34 except that here as the pressure is reduced the band intensity decreases with the consequence that the traces especially in C become very weak. The bands shown in E do show this advance in time very clearly however. Again a very significant relation is established using the pressure effect between the behavior of the atomic lines and the molecular bands.

Before leaving Figures 33 and 34 it is necessary to look at the afterglow and compare the atomic lines with the molecular bands. At all pressures the molecular band afterglow peaks in intensity near but just after the atomic line radiation reaches its maximum. This effect is most noticeable at the lower pressures. At the higher pressures the afterglow maxima are reached at about the same time. Also the decay in intensity of the
bands and atomic lines is very different. The atomic line radiation decays much faster than the molecular bands do. Again this seems to be more noticeable at the lower pressures than for the higher ones. Trace E of Figure 34 shows this effect very well for pressures from about 80 down to 20 mmHg since in this case a molecular band and an atomic line are included together.

Figure 35 is similar to Figures 33 and 34 except that here the 0.6 microsecond excitation pulse is used and the pressure variation is not so great. Notice again in the integrated light of A at each of the indicated pressures that the initial light intensity spike is present as in the case of the longer excitation pulse shown in Figures 33 and 34 and is more clearly separated at 98.5 mmHg from the main light intensity peak which follows. Observation of trace B in this same Figure again shows that the very early sharp light intensity spike is associated with the atomic lines and not the bands. As the pressure is reduced the main light intensity peak is advanced in time as described before for Figures 33 and 34 so that at the lower pressures the initial light intensity spike and the main light intensity peak almost merge into one as shown in trace A at 40.5 mmHg. By comparing this advance in time of the main light intensity peak with the incident pulse one can
Figure 35. Integrated light intensity and inverted 0.6 micro-second excitation pulse (A) and selected lines and bands at the indicated pressures.
see that the shift is really an advance of the main second peak and not a delay of the initial light intensity spike. Careful examination of trace A at 40.5mmHg shows that even though these intensity maxima are very close they are still distinct events in the discharge.

Again in trace A at the different pressures in Figure 35 one sees evidence of a slight correlation of the light intensity decay from the second intensity peak during the power pulse and the decay of the microwave pulse itself. This is more evident at 98.5 and 81.5mmHg than at the two lower pressures. Ordinarily with the longer excitation pulse the light intensity decrease of the second peak levels off to a shoulder region where the light intensity holds almost constant until the microwave pulse begins to decrease at which time the light intensity follows this decrease. In this case with the shorter excitation pulse the evidence seems to indicate from the slight change in the slope of the decay that the shoulder is on the verge of formation at the time the short power pulse begins to decrease. This of course means that the light intensity continues to decrease but at a different rate and that the equilibrium condition so typical of the longer pulse never has time to form.

As was described in the case of Figures 33 and 34 for the 2.1 microsecond excitation pulse, Figure 35 shows
a tendency for the much weaker molecular radiation to follow the level of intensity of the atomic line radiation during the excitation pulse itself while in the after-glow the molecular radiation, while weaker than the atomic lines, remains at a given intensity level for a much longer time. Again since the pulses are synchronized in time one can observe in E and F of Figure 35, as was the case with the longer pulse in Figures 33 and 34, that the afterglow atomic light intensity always peaks just before the molecular radiation does.

4.5 Pressure and Power Variation of the Microwave Pulses

Figures 36 and 37 show the incident, reflected, and transmitted pulses for both pulse durations and at pressures ranging from 220 to 19mmHg. The oscilloscope sweep speeds are 0.5 and 0.02 microseconds per division for A and D and 0.5 and 0.05 microseconds per division for B, C, E, and F. The faster sweeps apply to the second exposures which were taken to reveal the leading edge characteristics. Most of the pulses represent high average powers of 37 watts for the 2.1 microsecond pulse and 42 watts for the 0.6 microsecond pulse. At the lower pressures of 40.5 and 19mmHg a range of powers is shown. For the longer pulse these average power levels are 37, 20, and 10 watts while for the shorter pulse
Figure 36. Incident (I), reflected (R), and transmitted (T) pulses for the 2.1 microsecond pulse in A, B, and C. Similarly for the 0.6 microsecond pulse in D, E, and F.
Figure 37. Incident (I), reflected (R), and transmitted (T) pulses for the 2.1 microsecond pulse in A, B, and C. Similarly for the 0.6 microsecond pulse in D, E, and F.
they are 42, 20, and 10 watts.

The most significant result of these traces is the comparison of the incident, reflected, and transmitted pulses very near the beginning of the pulse. This was mentioned before in discussing part of Figure 32 where close observation shows the incident and reflected pulses to be very similar except for the initial edge of the reflected pulse. The traces show that during the initial short time that high power levels are transmitted through the gas there is little or no reflection. This can be seen best in A, B, and C at 220mmHg in Figure 36 where the leading edge of the reflected pulse is missing the part which trace C shows is transmitted. Thus one sees that the reflected pulse shape is significantly different from the incident pulse only before the plasma formation while the gas is transmitting a short pulse of microwaves and that the subsequent adjustment of the secondary light intensity peak during the discharge period from a maximum to the slow decay and shoulder formation is not evident at all in the character of the reflected pulse. What is happening to the plasma does so in such a way so as to not exhibit any significant external change in time to the incident microwave pulse.

Another aspect which was discussed in connection with Figures 33, 34, and 35 is the earlier occurrence
of the main light intensity peak occurring during the discharge period as the pressure is decreased. A corresponding behavior of the transmitted pulse can be observed in Figure 36 in the fast second exposures of either C or F. These second exposures were taken at 0.05 microseconds per division and show the detail of the leading and trailing edges of the transmitted pulse. It can be seen that the width of the pulse increases with the pressure and that most of this change occurs in the trailing edge. At the low pressures the pulse is almost symmetric while at higher pressures the slope of the trailing edge is much less than that of the leading edge.

In Figure 37 at 40.5 and 19mmHg one can examine the effect on the reflected and transmitted pulses of reducing the power level of the incident pulse. Traces B and E at these pressures show the slight irregularities of the reflected pulses when compared with the corresponding incident pulses in A and D. While no variations appear which are comparable to the effects observed in the light intensity variation during the same time period, still some aspects of the discharge prevent an exact duplication of the wave form of the incident pulse. This is especially true in the case of the medium power 0.6 microsecond pulse in E.
Traces C and F of Figure 37 at 40.5 and 19mmHg show the effect of a change in microwave power on the shapes of the transmitted spike. This effect is very similar to the pressure effect just described. At low power the waveform lacks symmetry, while as the power is increased the width of the pulse decreases and it becomes more symmetrical. At the same time, the microwaves are also able to penetrate the discharge tube section of the waveguide much later in the pulse for high power. This penetration corresponds to the shoulder formation of the light emission. It thus becomes very probable that the high light output early in the power pulse arises from high absorption of microwave power which soon is transmitted as the steady state is obtained.
V. THE INTENSITY VARIATION FOR OTHER GASES

5.1 Apparatus Modification

At the conclusion of the helium scans and time resolved studies, the microwave excitation system was modified to allow a survey of the integrated light intensity (vs) time response with different gases in the discharge. The modification involved only the section beyond the ferrite isolator in Figure 6. One end of a 90° waveguide twist was attached to the ferrite isolator and the other end was attached to a 90° H-plane waveguide bend. The original x-band waveguide discharge section as used by Tynes (103, p. 11) was then mounted vertically to the bend and a high power terminator which was also mounted vertically, completed the pulse power arrangement.

The high vacuum pumps were by-passed and a simple system was devised to allow filling, flushing, and evacuating the plasma tube.

5.2 Comparison of the Intensity Traces

Figures 38, 39, and 40 show the time resolved integrated intensities for tank oxygen, air, tank helium, tank argon, tank hydrogen, and natural gas at the limiting high, intermediate, and limiting low pressures for the formation of the plasma. The plasma was produced
Figure 38. Integrated light intensity as a function of time with the 0.6 microsecond pulse for tank oxygen (LEFT) at pressures of 70, 20, 10, 2, and 1 mmHg for A through E. Similarly for air (RIGHT) at pressures of 75, 55, 10, 2, and 1 mmHg for A through E. The sweep speed is one microsecond per division while the average power is 24 watts.
Figure 39. Integrated light intensity as a function of time with the 0.6 microsecond pulse for tank helium (LEFT) at pressures of 955, 750, 335, 54, and 2 mmHg for A through E. Similarly for tank argon (RIGHT) at pressures of 84, 80, 17, 2, and 1 mmHg for A through E. The sweep speed is one microsecond per division while the average power is 24 watts.
Figure 40. Integrated light intensity as a function of time with the 0.6 microsecond pulse for tank hydrogen (LEFT) at pressures of 55, 30, 5, 2, and 1 mmHg for A through E. Similarly for natural gas (RIGHT) at pressures of 140, 14, 3, 2, and 1 mmHg for A through E. The sweep speed is one microsecond per division while the average power is 24 watts.
with the 0.6 microsecond incident power pulse at 1000 pulses per second. Observations were made with the oscilloscope at a vertical sensitivity of 50 millivolts per division, a sweep speed of 1 microsecond per division and an external attenuation of 32db. All of the tank gases were of commercial grade while the natural gas was obtained from the building outlets and when analysed with a mass spectrometer was found to be composed of methane (93.6%), ethane (4.3%), propane (1.3%) and air (0.8%).

The limiting high pressure intensities shown in trace A are similar for all of the gases. This pressure, while different for the various gases represents the highest pressure at which it was just possible to sustain a stable plasma at full incident microwave power. The intensity is low but there is still very little delay in the production of the plasma except for helium. For oxygen and air the high pressure light intensity follows an exponential type charge and decay shape without the customary light spiking at the beginning of the power pulse. No shoulder effect is evident at the high pressures for any of the gases with perhaps the exception of the above mentioned hydrogen. In this case even though the intensity is low, the light rises very sharply and roughly holds the same level until the power
pulse declines. These higher limiting pressures were found to be 70mmHg for oxygen, 75mmHg for air, 955mmHg for helium, 84mmHg for argon, 55mmHg for hydrogen, and 140mmHg for the natural gas. A 5mmHg pressure increase above these high pressure limits was sufficient to prevent the attainment of a stable plasma.

The delay in the high pressure helium light intensity production shown in trace A of Figure 39 is not unique with this gas but was characteristic of the high pressure limit for all gases. Because of the instability of the plasma formation for the other gases, the delay is shown only for helium at 955mmHg. The very high pressure at which one is able to produce the helium plasma again indicates that collisions play an important part in the energy transfer for this gas. In the case of the other gases such frequent collisions evidently dissipate the energy before excitation and ionization become possible.

As the pressure is lowered in trace B of Figure 38 for oxygen and air, the light intensity increases and the sharp early rise in light intensity begins to characterize the initial optical emission. For oxygen in trace B of Figure 38 the light spike is very sharp and rises almost to the third division on the illuminated vertical scale. It is regrettable that the exposure was
not sufficient to record this very sharp light intensity spike. Again in trace C for oxygen the exposure does not show the initial sharp rise. It is shown in D and E but here it is very much reduced in height. There seems to be very little delay in the production of the plasma for oxygen and air, while a significant fraction of the pulse period is required in traces A and B for helium. For argon, hydrogen, and natural gas the sharp early rise in the light intensity is not well developed in trace B but at least it becomes more evident than at the higher pressures in trace A.

At still lower pressures in C the intensity continues to rise for all of the gases except natural gas. These pressures were adjusted for each gas to illustrate a significant change in the intensity trace. Oxygen yields in trace C at 10mmHg its maximum sharp spike at the beginning of the trace. As mentioned before, this sharp light intensity spike is not sufficiently exposed in Figure 38 but does reach a full four division vertical deflection at this pressure. It is very probable that this effect is a result of the previous pulse and occurs as a particular electron energy is attained just prior to atomic excitation and ionization.

For air the sharp initial spiking effect is also most noticeable in trace C at 10mmHg. The helium
discharge exhibits a high initial spike at the relatively high pressure of 335mmHg in trace C while at the same time showing a slight afterglow. The third trace at 17mmHg for argon fails to show dominant initial spiking but the sharp light intensity rise does bring the intensity very quickly to about two thirds of the maximum value attained later in the pulse period. A very weak afterglow begins to appear in Figure 39 for argon but does not assume any real importance.

The third trace for hydrogen at 5mmHg is almost identical in form to that of argon while trace C for natural gas at 3mmHg is again very similar but with the important difference that the light spike is definitely evident above the main pulse level. Notice in trace C for argon, hydrogen and natural gas that the sharp early rise in light intensity is recognizable as it rises out of the more pronounced emission which generally peaks during the later part of the power pulse for these gases.

Near the lower pressure limit in trace D the light intensity maximizes at 2mmHg in the case of oxygen while a slight delay between the arrival of the power pulse and the formation of the plasma becomes evident. Some afterglow is not evident and the very sharp light spike is still present as the plasma is formed but is now much reduced in height. Trace D for air at 2mmHg shows
that the maximum intensity occurs at a slightly higher pressure since the early light spike is now decreasing. Notice that the shoulder is still almost exactly the same as it is in trace C at 10mmHg, thus distinguishing between the early plasma formation and these processes which correspond to the equilibrium condition later in the pulse period. A very slight afterglow is observed and the slight delay in the formation of the plasma begins to appear.

Trace D for helium at 54mmHg shows the most extensive afterglow of all of the gases. It has a duration of several microseconds and the initial afterglow intensity is higher than the shoulder which occurs during the power pulse. In the case of argon at 2mmHg in trace D the results are very similar to those already described in trace C. The afterglow is very insignificant and the power pulse region is still rounded. However, the sharp early spike even though present previously, now becomes most evident for this gas. Much the same can be said of trace D for hydrogen and natural gas both at 2mmHg.

The E traces are representative of the low pressure limit at which it is just possible to sustain the plasma. In every gas it was possible to make slight adjustments of the pressure to illustrate the delay in formation of the plasma for a time of roughly one half of the pulse
period. For oxygen the reduced early sharp spike is still present and the intensity is still high in comparison to the other gases. In addition there is still a measurable afterglow. For air the sharp spike is also still present but the shoulder effect now begins to disappear. A very similar appearance is observed for helium. The last three gases, argon, hydrogen, and natural gas are very similar to one another in that the intensity is very much reduced, the early sharp spike is still visible, and the shoulder effect is also still visible.

The effect of pressure on the light intensity behavior, especially for oxygen, makes it possible to advance several conclusions. In the first place at the high pressure of 70mmHg in Figure 38, the problem of sustaining a plasma involves the loss of energy by the frequent collisions. The light spikes at the beginning of the light intensity traces and which are presumed to be due to particle species still present from the previous pulse are not in evidence at these high limiting pressures. This is very probably due again to depopulation of these particles by the frequent collisions. At the low pressure limit it is easier to adjust the pressure to the point where a delay of about one half of the pulse period occurs. Several things can be said
concerning this delay. First it is probably due to the problem of electron heating from the lack of collisions rather than the energy dissipation of the excited atoms and molecules resulting from "hot" electrons as at the higher pressures. The existence of the light spike at the beginning of the light intensity response in trace E supports this explanation. If in fact the sharp light intensity spike is due both to the existence of certain particle species from one pulse to another and also to the slight electron heating due to the arrival of the microwave power pulse then the appearance at the low pressure is expected since the reduced number of collisions will allow the existence of these particle species at the higher pressures and thus no initial light intensity spike at the beginning of the trace. At the lower pressures the particle species still exist from one pulse to another and after the delay as shown in trace E of Figure 38 for oxygen the electron energy is high enough to release the energy.
VI. RESULTS AND CONCLUSIONS

6.1 Introduction

It is not unusual to read that the understanding of the helium afterglows is not yet complete. As an example, one may examine a recent paper by Ferguson et al. (37, p. A381) where dissociative recombination of He$^+_2$, which has been used in the past to explain helium afterglow studies, is seriously questioned.

Another point that should be made is that the naming of certain processes is not at present clear in two of the more recent papers dealing with the helium problem. Ferguson et al. (37, p. A382) discusses "collisional-radiative recombination" in terms of two "coupled" processes $X^+ + e \rightarrow X^* + h\nu$ and $X^+ + 2e \rightarrow X^* + e$. The first is simple two-body radiative recombination while the latter is referred to as collisional recombination and involves three bodies. $X^*$ represents an excited atomic or molecular state which may then be ionized, transferred to higher or lower states, or radiate to a lower state. Biondi (10, p. A791), in referring to the paper by Ferguson et al., mentions the predominance of three-body "collisional-radiative recombination" as if only the latter process listed above is acting. He then concludes that the precise nature of
the helium recombination process is not at all clear at the present time.

The extensive effort to achieve an understanding of the important processes occurring in helium has been directed not to the early light emission during the plasma formation and immediately afterwards, but to the late afterglow. The work of Tynes (103, p. 1) however, is concerned with the early light emission from the helium plasma and the details of the atomic emission were stressed in the thesis. Brief mention is made by Janin and Eyraud (53, p. 1073) of the double peaked atomic emission during and after the pulse and also the weak molecular emission.

This study presents for the first time important details of the molecular emission, as well as the atomic line emission, for this early period of the pulsed microwave produced plasmas. In so doing, several temporal variations of the atomic line emission seem more clear than in Tynes' work. It is this overlapping of the problem, that makes it important to know, if only qualitatively, what is occurring early in the plasma production because there are still difficulties in explaining the late afterglow.

In the following sections the most significant results will be summarized and then related to the
individual variations of the intensity (vs) time behavior for both the atomic line as well as the band emission.

6.2 The Results of the Pressure and Power Variation

Figures 15 through 17 show that the band intensity is directly proportional to the power at about 200mmHg for most bands. This is not however, the pressure for a maximum band intensity but at this particular pressure the intensity is directly proportional to the power and the slope of the intensity (vs) power curves is greatest for the $3d^{3}A_{u}^3 - 2p^{3}F_{g}$, 5733A molecular transition. This indicates a higher net probability for population of this high angular momentum triplet state which is generally true at all of the pressures. Two important questions suggested by this result are as yet not answered. In the first place, is this preferential population the same both during and after the pulse? Secondly, is this a primary population of the $3d^{3}A_{u}$ state or is it just a step resulting from some other primary process?

These same Figures also show that at a pressure of 1mmHg the band intensity is independent of the excitation microwave power over the range from 25 to 40 watts. Two possibilities, both dependent upon collisions, can account for this. First, the build-up of electron
energy from the microwave field may not increase with power because of the fewer collisions at this lower pressure. This has also been mentioned in connection with the light intensity traces for oxygen in Chapter V. Secondly, the lifetime of excited atomic states is short compared to the reciprocal collision frequency thus preventing an increase in ion formation by process 1 of Table I.

Figures 18 through 20 show that the pressure, at which the maximum band intensity is reached, shifts to higher values at higher powers. The \(3d\sigma^3\Sigma^+ \rightarrow 2p\nu^3\Pi\) g 5958A band and especially the \(3p\nu^1\Pi\ g \rightarrow 2s\sigma^1\Sigma^+\ u\), 5133A band have very broad light intensity maxima as the pressure is increased from 100 to 200mmHg. This shift to higher pressures at higher microwave power for maximum band intensity is an indication of the importance of collisional processes in the energy transfer from atomic excitation and ionization to molecular ion population and consequent molecular radiation.

In contrast to the band emission, Figures 22 and 23 show that the pressure, at which the maximum intensity for the atomic lines is reached, decreases as the power is increased. This is again what one would expect if collisional processes were depleting the excited atomic states. In general, for this type of microwave plasma
production in the waveguide, the bands are prominent at pressures greater than 10mmHg while the atomic line emission is dominant at pressures less than 10mmHg.

6.3 Contributions from the Time Resolved Effects

Figure 28 is very probably the single most important result of this study. It shows, first of all, that while not intense, the band emission does begin as fast as the atomic line radiation on a microsecond time scale. These pictures are much more distinct than those of Tynes (103, p. 92) and show the time resolved band emission occurring both during and immediately following the microwave excitation pulse. The band emission during the excitation pulse attains an equilibrium level in a few tenths of a microsecond at about the same time as the atomic line radiation. This equilibrium level is maintained over a time interval equal to the sum of the interval for no-transmission plus the interval for reduced transmission. It is lacking, however, the sharp light intensity spike both at the very arrival of the microwave power pulse and also just prior to the attainment of the light intensity equilibrium condition during the major part of the pulse period. Since both of these intensity spikes are so typical of the atomic emission it is thus evident that the light emission processes are
different for the molecules.

Another point to emphasize is that not only does the band emission appear early, but it is again the 0,0 vibrational levels that are prominent. In fact, in this study, these are the only transitions observed. This very early appearance of the 0,0 band emission is not discussed in the current helium afterglow studies.

Ferguson et al. (37, p. A382) quotes an estimate of $10^{-5}/v$ seconds for the mean vibrational lifetime of the vth vibrational level of $\text{He}_2^+$ (v) at 1mmHg. This is later used to de-emphasize the process of dissociative recombination in the late afterglow or about $10^{-3}$ seconds after the plasma production. But Ferguson makes no comment about early vibrational pulse period de-excitation as is evident from the transitions shown in Figures 28, 33 and 34.

A result as important as the early appearance of the band emission and the attainment of the equilibrium band intensity condition is the fact that the band light intensity does not in general decay as the microwave power pulse ends. This again is in marked contrast to the atomic line emission and is also shown in Figures 28, 33 and 34. This independence of the molecular radiation from the decrease of the microwave power at the end of the pulse shows that the energy for molecular
emission is not immediately dependent upon inelastic electron excitation and ionization of the helium atoms by the microwave field.

An abrupt rise in the band light intensity occurs at the end of the power pulse and is followed by a monotonic decay which is almost complete in about 10 microseconds. The fact that the band emission does not drop at the end of the power pulse but immediately increases, suggests that the radiation both during and after the pulse is essentially due to the same process, probably recombination of the helium molecule ion, together with quenching of the molecular radiation by the microwave field during the pulse period.

Figure 29 shows prominent atomic light spiking as the plasma is produced and as yet has not been satisfactorily explained. It is very probable that this really represents an energy resonance of the electrons, now being "heated" by the incident microwave power, with an energized particle still in the plasma tube from the previous pulse. It is probable that these particles are metastable atoms with the sharpness of the spike representing both the speed at which they are depleted and perhaps that the average electron energy is not favorable for longer times for transitions to higher radiating states.
Figure 31 indicates that the atomic D to P afterglow transitions are more intense than the P to S transitions. In contrast to this, during the microwave excitation pulse period all of these traces are very similar. This is another evidence of the need to emphasize different processes for the atomic light emission during the pulse period as opposed to the afterglow. In this particular case for the atomic emission during the pulse, the major process is assumed to be electronic excitation, as in 17 of Table I, of the singlet states with collisional transfer at higher excited states to populate the triplet series as discussed by St. John and Fowler (101, p. 1813).

6.4 Results from the Time Resolved Microwave Reflection and Transmission Data

Figure 32 shows the time response of the reflected microwave pulse and one concludes that in all major respects it is the same in form as the incident pulse. Careful study of traces B and C on the left of Figure 32, expecially the fast 0.05 microsecond per division second exposures show that the beginning of the reflected pulse in trace B is delayed for a time of about 10 nanoseconds. Thus as the incident microwave pulse first contacts the plasma tube there is no significant reflection for this very short time. However, during this time in trace C
transmitted power penetrates the gas.

Tynes suggested (103, p. 166) that reduced reflection early in the pulse period is responsible for the main atomic light intensity spiking early in the pulse. This seems difficult to defend with the time behavior of the reflected pulse as shown in Figure 32.

In trace C of this Figure, however, there is a suggestion of something unusual. It is not that it is strange to see a transmitted spike at the arrival of the incident pulse. One would naturally expect microwave penetration of the helium gas prior to the build-up of ionization and a subsequent increase in the electron density. The unusual observation is that after the short transmitted pulse is observed, the plasma becomes essentially opaque to the microwaves only for a very short time on a microsecond time scale and then begins to transmit again but at a reduced level. This is clearly shown in trace C of Figure 32 on the left.

The results of comparing this transmission with the behavior of the atomic light intensity is shown on the right of Figure 32. In the first place trace A at high power shows that at the beginning of the plasma formation when the light intensity is high, the transmission is low. Later, however, as the shoulder formation of the light intensity develops, the transmission assumes the reduced-level value. It thus appears that at least part of the
variation of the light intensity with time during the pulse period is related to a corresponding behavior of the transmitted microwave power pulse. It is surprising that there is not a change in the reflected power at the same time that increased transmission occurs. It is possible that this is a manifestation of strong wall effects which tend to isolate the inner plasma tube volume.

Another result previously not available is the coincidence of the transmitted spike at the beginning of the pulse with the initial sharp light intensity spike as shown in trace C of Figure 32 on the right. It is regrettable that such a comparison is not available for Figure 29 taken with the plasma tube. However, from the results now available for the larger 18mm plasma tube in Figure 32, it is likely that the very sharp light intensity spike shown at the beginning of the pulse in Figure 29 coincides with an equally sharp transmitted pulse just prior to the plasma formation.

It should be emphasized that if a sharp light intensity pulse appears at the same time as an equally sharp transmitted pulse there is good evidence that the plasma has not yet formed and consequently the light originates from properly excited particles still present in the plasma tube from the previous pulse. As stated before, these particles are believed to be metastable atoms.

Two other types of behavior indicate that this
interpretation is qualitatively correct. In the first place it has been shown in Figure 32 on the right in traces A, B, and C that as the incident power is reduced the initial sharp light intensity spike remains about the same while the main light intensity pulse which accompanies the plasma production is delayed more and more as the incident microwave power is decreased. This simply reflects the longer time to build-up the electron energy, ionize the gas, and thus produce the plasma at the lower incident microwave power for the main light intensity pulse. The apparent lack of sensitivity of the sharp initial light intensity spike to power changes is expected since the electron energy required to change a metastable level to a radiating one is not large.

Secondly, Figures 33 and 34 have shown that at fixed incident microwave power, the delay in the appearance of the main light intensity pulse increases as the pressure increases. This is typical for the main pulse atomic emission which is most easily excited at the lower pressures of this study. At higher pressures, where the main light intensity is delayed, the initial sharp light intensity spike appears at the same time as the incident microwave pulse but at a reduced intensity.

The initial sharp light intensity spike decreases with a pressure rise, indicating a collisional depletion of the metastable atoms between microwave power pulses.
Figure 32 on the right shows the initial sharp light intensity spike increasing with an increase of incident microwave power. It is assumed that this indicates a higher metastable density at higher incident microwave powers.

6.5 The Dual Nature of the Main Atomic Light Intensity During the Incident Power Pulse Period

From the behavior of the light intensity variation with time it has been concluded that the major molecular radiation results from recombination both during and after the incident microwave pulse. In the case of the atomic emission however, the major source of radiation during the pulse is undoubtedly electronic collisional excitation. It is possible to deduce that this is not the complete source, however. This information comes from Figures 33 and 34 where one observes for example, from the 5876A atomic line emission, the decrease in light intensity at the end of the microwave excitation pulse. At low pressures this decrease reduces the intensity almost to zero while at the high pressure limit the minimum light intensity at the end of the excitation pulse is not close to zero at all. If this residual light emission actually occurs during the entire pulse period, it contributes significantly to the total emission at the high pressures but is negligible at low pressures. Notice
that this pressure behavior is in contrast to that of the initial sharp light intensity spike discussed previously and which tends to disappear at high pressure.

6.6 Summary

In order to see the results of this study, Figure 41 shows a representation of the atomic line light intensity and the molecular light intensity as functions of time with different sections numbered for identification. The information acquired regarding the interpretation of the time behavior of the light intensity will then be summarized using the identifying number from Figure 41.

1. (a) It is assumed that this sharp initial light intensity spike represents the atomic light emission from metastable atoms still present from the previous pulse which are electronically excited to radiating levels. Important processes are, \( \text{He}^m + e \rightarrow \text{He}^* + e \) and \( \text{He}^*(1) \rightarrow \text{He}^*(2) + h\nu_{12} \). (b) The maximum intensity coincides timewise with the initial transmitted spike of the power pulse and is an indication that the discharge is not yet produced. (c) The maximum intensity does not noticeably shift position with either pressure or power variations. This again separates it from the behavior of the light intensity during the pulse. (d) The intensity, however, does decrease with increasing pressure indicating collisional losses. (e) The intensity increases slightly with
Figure 41. Qualitative representation of the atomic line light intensity and the molecular light intensity as functions of time.
increased power, indicating an increase of the appropriate particle density.

2. (a) The main light intensity maximum represents atomic light emission mainly from direct electronic excitation. The important processes are \( \text{He} + e \rightarrow \text{He}^* + e \) and \( \text{He} + e \rightarrow \text{He}^+ + 2e \). (b) It coincides timewise with the no-transmission part of the pulse period. This is a possible reason for the higher intensity of region 2 as compared with region 3. (c) It occurs at later times as the pressure is increased. (d) It occurs at earlier times as the power is increased.

3. (a) This region represents the equilibrium portion or shoulder of the atomic light emission which again is mainly due to direct electronic excitation. The important processes are the same as in 2 (a). (b) The intensity decreases as the pressure is increased and indicates a collisional loss of excited atomic states. (c) The intensity increases slowly as the power is increased. (d) A decrease in intensity occurs as the incident microwave power pulse ends. This is what one would expect for direct electronic impact excitation. (e) This part of the atomic emission coincides timewise with the reduced but stable transmission of microwave power which occurs after the no-transmission part of the pulse period. This loss of power probably is the cause of the intensity reduction as compared with region 2.
4. (a) This represents the residual atomic light intensity due to sources other than direct electronic excitation, and is most probably recombination by the process \[ \text{He}^+ + 2e \rightarrow \text{He}^* + e. \] (b) The intensity increases as the pressure is increased. This is an indication of the enhancement of the light intensity as the collision frequency increases. (c) The dashed line indicates that this atomic radiation may also exist during the main pulse period.

6. (a) The afterglow decays faster than the comparable molecular radiation. This is expected since the excited atoms may populate the molecular states as well as radiating. (b) The afterglow decay time is less as the pressure is increased. This indicates the importance of collisional processes as expected for recombination. (c) The intensity increases with increased power.

7. (a) This region represents the fast beginning and stable pulse period emission of the molecules probably by the recombination reaction \[ \text{He}_2^+ + 2e \rightarrow \text{He}_2^* + e \] during the power pulse. (b) The intensity increases as the pressure is increased. Notice this is the same as 4 (b). (c) The time required for reaching the equilibrium level is less at the lower pressures. This is probably due to the decreased population, at higher pressures, of excited atomic states needed to produce the molecules. (d) The low intensity level in comparison to the afterglow is
probably due to the quenching effect of the microwave field. (e) There is no drop in the intensity at the end of the microwave excitation pulse. This indicates that the net molecular source is not directly related to the power pulse.

8. (a) This region represents increased molecular light intensity by recombination as the excitation pulse ends and the electrons cool. (b) The maximum intensity occurs either at about the same time or slightly later than the corresponding maximum for the atomic emission.

9. (a) The decay in the afterglow light intensity represents the loss of the \( \text{He}_2^* \) ion and is slower than the decay of the atomic light intensity. (b) The decay time decreases as the pressure is increased. (c) It appears possible that regions 7, 8, and 9 may be qualitatively similar to regions 4, 5, and 6 of the atomic emission. This assumes the behavior of region 4 is also typical of region 2 and 3.

Finally, one can reasonably assume from the summary and Figure 41 that region 1 represents light emitted from particles left from the previous pulse which are raised to radiating levels as a result of the increased electron energy as the next pulse arrives. Region 2 is principally light emitted by radiating atoms produced by direct electron excitation. The same may be said of region 3 with the reduced intensity corresponding to increased
transmission of the microwaves. The increased transmission is probably due to a change in spatial distribution of the particle species in the discharge.

One may deduce that direct electron excitation is not the only process occurring during the microwave pulse by observing that the intensity of region 4 does not drop to zero as the microwave pulse ends. It is possible that this non-zero component is also present in region 2 and 3 as shown by the dashed line in Figure 41. Since the intensity of region 4 increases with increasing pressure, one concludes that probably collisional-radiative recombination (37, p. A381) is dominant. This process is also used to explain the light emission of region 5 and 6. The summary shows that the pressure behavior of region 7 is the same as that of region 4, showing that part of the atomic emission during the pulse is essentially the same as the molecular emission for the same time period. The similarity of the atomic and molecular afterglows has already been mentioned in 9 (c).
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APPENDIX A

THE GAS HANDLING SYSTEM

1. NRC 4D Rotary Gas Ballast Mechanical Pump—Type 4512-3.3 cubic feet per minute. (NRC Equipment Corporation. Newton Highlands 61, Massachusetts)

2. 3 Stage Mercury Diffusion Pump — Model M40110 — 70 liter per second. (Research Vacuum Supply Company. 6712 North Clark Street, Chicago 26, Illinois)

3. Type C Ultra-High Vacuum Valve — Bakeable to 450°C. (Granville-Phillips Company. Box 198, Pullman, Washington)

4. Uranium Metal Disc (Fisher Scientific Company. 2850 South Jefferson Avenue, St. Louis 18, Missouri) U-238.07. Maximum impurities: 100ppm of Cr, Fe, and Ni; 50ppm of Mn; 20ppm of V.

5. Ionization Gauge — 675-P — 100 microamperes per micron at 10 µu grid current. (Vacuum-Electronics Corporation. Terminal Drive, Plainview, Long Island, New York)

6. Veeco Vacuum Gauge — Type RG-21A — (2) $10^{-10}\text{mmHg}$ (Vacuum-Electronics Corporation. Terminal Drive, Plainview, Long Island, New York)

7. Tektronix L-C Meter — Type 130 — 0.1 to 300 micromicrofarads (Tektronix Incorporated, Portland, Oregon)

8. Pressure Gauge — 1-100mmHg — Model F19160 — Serial DDO 2521 (Wallace and Tiernan Company. Bellville, New Jersey)

9. Vacuum Thermocouple Indicator Meter and Filament Supply—Model 301 (Western Electrical Equipment Corporation. Newark, New Jersey)

MICROWAVE EXCITATION SYSTEM

1. 725A Magnetron — Part of surplus radar unit RT-5A/APS-4

2. Microwave Power Meter — Model 430C (Items 2 through
APPENDIX A (continued)

7 supplied by Hewlett-Packard Company. 1501 Page Mill Road, Palo Alto, California

3. Frequency Meter - Model X532B

4. Variable Attenuator - Model X382A

5. Fixed Attenuators - Model X370C and X370D

6. Directional Couplers - Model X752D

7. High-Power Termination - Model X912A

8. Power Divider - Fabricated in the Physics Department Shop at Oregon State University

9. Plasma Tube Transition Waveguide Section - Fabricated in the Physics Department Shop at Oregon State University

10. Ferrite Isolator - Model X-126 (Cascade Research, Division of Monogram Precision Industries, Incorporated, Los Angeles, California)

11. Directional Coupler - RG52-097 (Ramage and Miller, Incorporated, 3221 Florida Avenue, Richmond, California.

OPTICAL DETECTING SYSTEM

1. JACO 2.5 Meter Adjustable Grating Spectrograph (Jarrell-Ash Company, 165 Newbury Street, Boston 16, Massachusetts)

2. Grating Monochromator - Serial Number DB548 (Bausch and Lomb Optical Company, Rochester 2, New York)

3. Cathode-Ray Oscilloscope - Type 517A (Tektronix, Incorporated, P. O. Box 831, Portland 7, Oregon)


5. Photomultiplier Tube - RCA 1P21
APPENDIX A (continued)

6. Cathode-Ray Oscilloscope - Type 551 Dual Beam
   (Tektronix, Incorporated. P. O. Box 831, Portland 7, Oregon)

7. Keithley Electrometer - Model 600-A (Keithley Instruments. 12315 Euclid Avenue, Cleveland 6, Ohio)
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# APPENDIX C

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The transitions indicated on the left apply to both the singlet and triplet series. The transition frequencies $\gamma$ and $\gamma'$ are from Gabriel (43, p. 125) and Rogers (94, p. 36) respectively. *The 1S-2P and 2S-2P $\gamma'$ transition frequencies are from Phelps (92, p. 628).