BASIC PROCESSES OCCURRING IN GASEOUS PLASMAS
IN VARIOUS CHARGE DENSITY AND ENERGY STATES

by

The Gaseous Electronics Laboratory
Department of Electrical Engineering
University of Illinois

Final Report

NASA Grant NGR 14-005-037
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This laboratory has undertaken studies of "Basic Processes Occurring in Gaseous Plasmas in Various Charge Density and Energy States" in agreement with NASA Lewis Research Center under NASA Grant NGR 14-005-037. This report reviews the work performed under this grant, suggests how the results of these studies can be applied to future projects and lists the technical reports, theses and scientific articles in which the results of these investigations have been published.
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I. INTRODUCTION

This grant has been used to investigate a wide range of topics in Gaseous Plasmas. The initial work performed was concerned with the development of sensitive laser diagnostic procedures for the investigation of plasma properties. This project has met with considerable success, although the grant was terminated before the laser diagnostic techniques could be applied to the determination of significant plasma properties. The remaining projects were concerned with investigations of basic processes occurring in gaseous plasmas using the currently available diagnostic procedures. These projects are all thoroughly covered in the next section.

All of the projects initiated under this grant have shown an indication of future promise. There is every indication that the results of these investigations will play an important part in future scientific studies.
II. THE LASER HETERODYNE SYSTEM FOR PLASMA DIAGNOSTICS

The Gaseous Electronics Laboratory of the University of Illinois has been a pioneer in the development and use of laser diagnostic techniques for the probing of gaseous discharges. This background experience provides us with a unique capability to conduct experimental studies of the charge distribution inside plasma devices. Indeed, we have developed one particular aspect of laser diagnostics, the "Laser Heterodyne System" to the point where it is now possible to measure electron densities as low as $5 \times 10^{10}/cm^3$ over path lengths of 20 cm with a cross sectional resolution of $< 1 \text{ mm}^2$. We can also make simultaneous measurements of neutral density variations and the variations of one other plasma component, possibly excited states.

To put the laser heterodyne system in perspective, it is useful to first examine its forerunner, the laser interferometer. In the laser interferometer, the plasma being studied is placed within the laser cavity. Changes in the densities of the plasma constituents cause corresponding changes in the optical path length of the laser cavity which in turn alter the laser frequency. The change in laser frequency is determined by an external optical cavity which operates essentially as a fixed frequency meter. As the laser frequency scans through the various resonances (and anti-resonances) of the external cavity, the laser output will exhibit intensity peaks and nulls (fringes) corresponding to the presence of these resonances. Obviously, the sensitivity of this system is limited by how closely the resonances of the external cavity are placed as a function of frequency. A full description of the laser interferometer is given in many papers by the staff of the Gaseous Electronics Laboratory. The various strategems used to increase the sensitivity of this system have
given a lower limit, for single shot experiments, of one fringe per 7.6 \times 10^{13} \text{ electrons/cm}^3 over a path length of 40 cm. Even measurements of fractional fringe shifts do not improve this figure by more than an order of magnitude. In order to measure lower densities, we have therefore devised a system with essentially "infinite resolution," the laser heterodyne system.

In the laser heterodyne system, the plasma to be observed is also placed in the cavity of a probe laser. As in the case of the interferometer, changes in the plasma refractive index cause a shift in the laser frequency. In this case, however, the frequency shift is detected by heterodyning the probe laser's output with the signal from a reference laser of essentially constant frequency, the local oscillator. The changes in the frequency of the resulting beat note are detected with conventional video-frequency equipment.

The device containing the plasma whose spatial and temporal behavior is to be measured is placed within the cavity of a He-Ne laser. The laser beam is parallel to the axis of the device so that the laser beam interacts with the maximum volume of plasma. The output of this laser is mixed with the output of the reference laser and the frequency difference is detected by a square law photodetector. Only the low frequency components are retained and these are displayed using conventional video frequency electronics. As the plasma refractive index changes, the frequency of the probe laser will change and this will be displayed on an oscilloscope. Since the lasers we use operate at three wavelengths, 0.6328 \mu, 1.15 \mu and 3.39 \mu, the data obtained at these three
wavelengths can be used to determine electron density variations, neutral density variations, and possibly the contribution due to certain excited atoms or molecules.

The relation between densities and frequency shifts can be determined in the following manner. The frequency of the laser (within the Doppler broadened line) is determined by the requirement that the round trip phase shift of the optical signal within the cavity must be a multiple of \(2\pi\) radians. Therefore, with no plasma the frequency of the probe laser, \(\omega_0\), is determined by the equation:

\[
\frac{\omega_0}{c} d_k + \Delta \phi (\omega_0) = q\pi
\]

where \(q\) is the longitudinal mode number of the cavity (\(\sim 10^6\)), \(d_k\) is the length of the cavity and \(\Delta \phi (\omega)\) is the extra phase shift introduced by the dispersive nature of the laser transition. When a plasma is formed, the frequency of the probe laser changes to \(\omega_1\), where

\[
\frac{\omega_1}{c} d_k + \frac{\omega_1}{c} \Delta n_p (t)d_p + \Delta \phi (\omega_0) + \frac{3\Delta \phi}{3\omega} \bigg|_{\omega_0} (\omega_1 - \omega_0) = q\pi
\]

\(\Delta n_p (t)\) is the refractive index change produced by the plasma and \(d_p\) is the length of the plasma being probed. As can be seen from this equation, \(d_p\) should be a maximum for greatest sensitivity. For the case when \(\omega_p^2/\omega^2 \ll 1\), which is always true for our region of interest, the refractive index of the plasma is given by

\[
n_p = 1 - \frac{1}{2} \frac{\omega_p^2}{\omega^2} + 2\pi N_m a_m + 2\pi \sum_{i} a_i (\omega) N_i
\]
where

\[ \omega_p^2 = \frac{N_e e^2}{m\varepsilon_0} \]

is the plasma frequency

\[ \alpha_m = \text{polarizability of the ground state atoms or molecules per unit volume} \]

\[ \alpha_i = \text{polarizability of ions or excited atoms per unit volume} \]

\[ N_m, N_i = \text{number density of ground state atoms/molecules, excited atoms/ions.} \]

There are three variables in \( n_p; \Delta N_e, \Delta N_m \) and \( \Delta N_i \); so, in general, three independent equations of the form of Equations (1) and (2) are required.

Our three wavelength operation may provide this. Normally, however, excited species play an insignificant role and so only two wavelength measurements are necessary. We will consider only this case for simplicity. Therefore,

\[ \Delta n_p = \frac{1}{2} \frac{e^2}{m\varepsilon_0} \frac{\Delta N_e}{\omega^2} + 2\pi \alpha_m \frac{\Delta N_m}{m} \tag{4} \]

Since the contribution due to \( \Delta N_m \) is frequency independent and the contribution due to \( \Delta N_e \) varies inversely with frequency, two wavelength measurements will provide solutions for \( \Delta N_e \) and \( \Delta N_m \). Let us assume laser operation at nominally two wavelengths \( \lambda_1 \) (say 6328 Å) and \( \lambda_2 \) (say 1.15 μ).

From Equations (1), (2) and (4)

\[ \Delta f_{1,2} \left[ 1 + \frac{c}{d_\lambda} \frac{\partial \Delta \phi_{1,2}}{\partial \omega} \right] = -2\pi \alpha_m \lambda_2 \frac{\Delta N_m}{m} \tag{5} \]

and therefore,

\[ \Delta N_m = \frac{1}{2\pi \alpha_m C} \left[ \frac{\lambda_1^2}{\lambda_2^2 - \lambda_1^2} \Delta f_2 (1 + \frac{c}{d_\lambda} \frac{\partial \Delta \phi_1}{\partial \omega}) \right] \tag{6} \]
and \[ \Delta N_e = \left(\frac{8\pi^2 \varepsilon_0 c}{e^2} \frac{d \Delta f_2}{d \omega} \right) \frac{\lambda_2}{\lambda_2 - \lambda_1} \left[ \Delta f_2 \left(1 + \frac{c}{d \omega} \frac{\partial \Delta f_2}{\partial \omega}\right) \right. \\
\left. - \frac{\lambda_1}{\lambda_2} \Delta f_1 \left(1 + \frac{c}{d \omega} \frac{\partial \Delta f_1}{\partial \omega}\right) \right] \] (7)

If \( \Delta f_2 \) and \( \Delta f_1 \) are experimentally measured, \( \Delta N_e \) and \( \Delta N_m \) can be determined since the other quantities involved are known or are measurable. The dispersion terms \( \frac{\partial \Delta f}{\partial \omega} \) are the only troublesome terms in this respect. For the He-Ne laser, \( \frac{\partial \Delta f}{\partial \omega} \) is negligible for operation at .6328 \( \mu \) or 1.15 \( \mu \) since these are low gain transitions. Neglecting this term would contribute an error of no more than a few percent. For He-Ne lasers operation at 3.39 \( \mu \), \( \frac{\partial \Delta f}{\partial \omega} \) can contribute an error of 20 to 30% and so must be evaluated. Normally, operation at this wavelength would be avoided, however, the 3.39 \( \mu \) operation should give the most sensitivity to electron density changes. In order to take advantage of the fact, we have developed an experimental technique for the determination of \( \frac{\partial \Delta f}{\partial \omega} \) under actual measurement conditions. This has provided us with a three wavelength laser heterodyne system of high resolution and sensitivity.

The laser heterodyne technique has been developed to the point that it can be used to make reliable and routine plasma measurements. The latest breakthrough in the use of the 3.39 \( \mu \) transition should allow us to get appreciably more sensitivity than our previous value (using the 1.15 \( \mu \) and .6328 \( \mu \) transition) of 5 x 10\(^{10}\) cm\(^{-3}\) over a 20 cm path length. We are in an excellent position therefore, to investigate the temporal and spatial variations of low density plasmas.

Future investigations could very profitably use the laser heterodyne techniques that we have partially developed under NASA sponsorship. For
instance, the behavior of plasmas near surfaces is of direct relevance and has been the subject of an enormous amount of conjecture and theory, but a direct experimental confirmation of existing theories has not been obtained. It is easy to see why. Before the advent of the lasers, the two primary methods of plasma density determination were microwaves and probes. Probe characteristics depend upon the plasma-surface interface characteristic and hence cannot be used to give an independent measurement of plasma-surface behavior. Microwave techniques are independent in this respect but suffer from the lack of spatial resolution obtainable with a probe. Laser diagnostic techniques, however, should provide us with measurements which are independent of plasma-surface properties, coupled with fine spatial resolution.

The laser techniques are closely related to the microwave methods with the obvious modification of increased frequency. The increased frequency capability afforded by lasers allows much greater spatial resolution. Figure 1 illustrates the spatial resolution obtainable in the heterodyning system with the gas laser. Let us suppose that the plasma-surface interface to be studied is located within the laser cavity. Then the electric field of the normal modes within this cavity are described by the Hermite-Gaussian functions. For the lowest order TEM mode, the field varies in the transverse plane according to the following relation:

$$E(r) = \exp \left( -r^2/\omega^2 \right)$$

where $\omega$ = spot-size of the laser beam (see fig. 1 for definition).

For a typical set of parameters, let us pick $d \sim 98$ cm, $b \sim 100$ cm,
Radius of Curvature = b

Figure 1 Geometry of Laser-Plasma Interaction

\[ W_0^4 = \left( \frac{\lambda}{\pi} \right)^2 d_e b \left( 1 - \frac{d_e}{b} \right) \quad \lambda = \text{ laser wavelength} \]

\[ W_1^4 = \left( \frac{\lambda}{\pi} \right)^2 \frac{d_e b}{1 - \frac{d_e}{b}} \]

\[ W^2(z) = W_0^2 \left[ 1 + \left( \frac{\lambda}{\pi} \frac{z}{W_0^2} \right)^2 \right] \]
The minimum spot-size, $w_0$, equals 0.168 mm, and the beam size is doubled at a distance of 42 cm from the flat mirror. Thus one has at least a 0.6 mm spatial resolution over a plasma length of 40 cm. Obviously, one can trade plasma length, laser cavity length, and mirror separation for increased spatial resolution.

It should be emphasized that one can still infer finer spatial resolution by utilizing the perturbation theory developed for microwave cavities.\textsuperscript{7} One would assume a power series expansion for cross-sectional variation of the electron density as a function of the distance away from the surface; i.e.,

$$\text{Ne}(x) = a_1 \left( \frac{x_0 - x}{L} \right) + a_2 \left( \frac{x_0 - x}{L} \right)^2 + a_3 \left( \frac{x_0 - x}{L} \right)^2 + \ldots$$

(9)

$L = \text{arbitrary scale length, (x}_0 \text{ and x are defined in figure 1)}$

The resonant frequency of the optical cavity (and thus the laser frequency) is given by:

$$\frac{\omega - \omega_i}{\omega_i} = \frac{1}{2} \frac{e^2}{\kappa c \omega_i^2} \int_V \text{Ne}(x) E^2 dV$$

(10)

where

- $E = \text{Optical field intensity}$
- $\omega_i = \text{Initial resonant (laser) frequency}$
- $\omega = \text{New laser frequency}$
- $V = \text{Volume of the optical cavity}$

In order to evaluate the coefficients, one repeats the experiment by changing parameters of the measurement process. For instance, one could adjust the plasma-surface interface with respect to the center of the laser beam, thereby changing the parameter $x_0$ in Equation (9). Alternatively, one could change
modes in the laser (say to the TEM$_{1,0}$ mode) or change $d$ which changes the spot size. The coefficients $a_1, a_2, a_3$ are adjusted to fit the data. If the scale length over which the plasma changes rapidly is much larger than the spot-size $w(z)$, then Equation (8) can be approximated by $\delta(x)$, and Equation (10) yields the familiar equation:\textsuperscript{2,8,9}

$$\omega - \omega_0 = \frac{1}{2} \frac{\omega_p^2}{\omega^2} \cdot \frac{2\pi c}{\lambda} \cdot \frac{dp}{de} = \Delta n \frac{2\pi c}{\lambda} \cdot \frac{dp}{de}$$

or

$$\Delta f = \Delta n \frac{dp}{de} \cdot \frac{c}{\lambda}$$

where

$\Delta n = \text{refractivity of the plasma}.$

Equation (10) has been in successful use for many years in the microwave cavity technique, even in some cases where the validity of perturbation theory can be questioned. However, at optical frequencies, the perturbation approach is surely valid inasmuch as $\omega_p^2/\omega^2$ is typically $10^{-8}$ or less.

In principle then, measurements of plasma density variations over scale lengths smaller than a spot-size are possible. The question mark at this time is whether the frequency stability of the lasers and the repeatability of the plasma production system is good enough to make meaningful measurements. If these experiments are successful, then variations on the above experiments can be performed involving changing the type of surface (i.e., conducting as opposed to insulating) and biasing the surface with respect to the plasma potential.

Other projects could be pursued which are closely related to the previous topic. For instance, an independent check of probe measurements of plasma densities could be made by comparing these measurements to simultaneous microwave and laser measurements.
III. EFFECTS OF LASERING UPON THE ELECTRON GAS AND EXCITED-STATE POPULATIONS IN XENON DISCHARGES

The work of R. Freiberg\textsuperscript{10,11} consisted largely of demonstrating the usefulness of a gaseous laser for the investigation of the relative importance of various electron-atom collisions in an active gaseous discharge. The laser radiation used was the $3.51\mu (5d_{33} \rightarrow 6p_{22})$ transition of xenon. The gas investigated was the same xenon gas used for lasering. The discharge tube was placed in an optical cavity and the discharge current, electron density, electron temperature and spectrally resolved discharge radiations were compared in the presence and absence of lasering. Lasering was interrupted by means of a mechanical chopping wheel positioned within the optical cavity. Studies were made in a pressure range of 12.5 to 25.5 m Torr and with discharge currents up to about 150 m amps. The discharge was through a very small bore capillary (2 mm I.D.) and this resulted in large anode directed pressure gradients and associated electron density gradients, both originating from electrophoretic effects known to be important in a closed capillary discharge tube. Because of these gradients it was necessary to make the observations of electron density and temperature and discharge light at various axial positions along the tube. Mechanically chopping the laser cavity made it possible to utilize phase sensitive detection and so it was possible to record the electron temperature variations which were \( < 0.4 \) per cent of an electron temperature of 8,000 to 10,000 \( ^{\circ}\)K, electron density increases and decreases of \( \sim 10^{9} \) electrons/cc of electron densities between \( 8 \times 10^{10} \) to \( 2 \times 10^{12} \) electrons/cc and resulting changes of discharge current during lasering up to a maximum of 0.02 \%. Changes in electron density were made by observing shifts in the resonant frequency of a microwave cavity.
Electron temperature was sometimes determined by observing changes in the Q of the cavity but principally by measurements of the microwave noise emitted by the plasma.

The observation that discharge current was altered by lasering was found consistent with measured changes in electron density. Electron density at a given axial position along the tube was found to decrease due to lasering at low discharge currents but above a given current (a function of pressure) it was found to increase with discharge current. The behavior at low current was interpreted as due to the laser induced depopulation of the upper laser level (5p_{33}). Normally there is a contribution to free electron density from electron collisional ionization of 5p_{33} states. The partial depopulation during lasering decreases this contribution and thereby decreases the discharge current. However, after the normal discharge current is increased until the electron density becomes greater than about $10^{12}$ electrons per cm$^3$ the electron density increases due to lasering. It was concluded that increased ionization from the enhanced 6p_{22} and 6s metastables (below 6p_{22}) overcomes the reduced 5d_{33} ionization under these conditions.

Thus small laser induced changes in excited state populations can permit assessment of the relative importance of contributions of various excited state populations to ionization in an active discharge.
IV. METASTABLE MEASUREMENTS IN FLOWING HELIUM AFTERGLOW

Knowledge of the populations of neutral metastables, both atomic and molecular, is important in understanding the decay of a helium plasma following the cessation of active discharge (i.e., the afterglow). It is possible to produce a steady state afterglow by producing a breakdown at some point in a flowing gas. Distance downstream from the breakdown point corresponds to time in the afterglow. The feasibility of metastable population studies in a flowing helium gas was demonstrated by observations over a range of mean helium pressures from 5 to 35 Torr with the gas nominally at room temperature. Gas purity was maintained by taking the helium from below the liquid level of a liquid helium storage dewar.

The gas flowed in a 1 cm diameter quartz tube making it necessary to assume a parabolic distribution in velocity across the tube. The mean (across the tube) population of the triplet atomic metastable ($2^3S$) was determined as a function of distance from the breakdown point by solving the differential equation describing rate of change of $2^3S$ density due to loss by diffusion and by three-body conversion into the molecular metastable, $He_2(2^3\Sigma)$, by the reaction

$$He(2^3S) + 2He \rightarrow He_2(2^3\Sigma) + He.$$  

By experiment the diffusion coefficient was found to be 490 cm$^2$ sec$^{-1}$ Torr and the three-body combination coefficient 0.25 sec$^{-1}$ Torr$^{-2}$. The three-body coefficient is in good agreement with that reported elsewhere. In order to determine these numbers it was necessary to obtain relative mean $2^3S$ density as a function of distance from the breakdown point. This was done by injecting molecular oxygen at various distances from the breakdown point and interpreting the light observed from the $O_2^+$ second negative
system as resulting from Penning ionization of O₂ by He(2³S).

A second detection method was used in order to obtain absolute measurements of the mean He(2³S) densities. In this method argon was injected instead of oxygen. Again Penning ionization occurred by He(2³S) collisions with the injected gas. However, this does not lead to visible radiation as in the oxygen case so the effect of adding argon was monitored by measuring the increase in mean electron density with a microwave cavity. By attributing the increase in electron density solely to the efficient reaction

\[ \text{Ar} + \text{He}(2³S) \rightarrow \text{Ar}^+ + \text{He} + \text{e} \]

it was possible to identify the increase in mean electron density in the cavity region as the mean density of He(2³S). An accurate means of measuring electron density change was developed.¹⁴ Shifts in resonant frequency were measured with an accuracy of \( \pm 10 \) kc/sec by a superheterodyning technique and this accuracy made it possible to measure metastable densities down to \( 3 \times 10^7 \) cm\(^{-3}\). In fact two cavities were used; one permitting measurements in the range \( 2 \times 10^{11} \) to \( 10^{10} \) cm\(^{-3}\) and the other in the range \( 10^{10} \) to \( 3 \times 10^7 \) cm\(^{-3}\). Measurement of the diffusion coefficient of He(2³S) (from the argon-impurity experiments) as a function of pressure at a given afterglow time indicated that Penning ionization of the argon by the molecular metastable can be important. This can be taken into account (by using the reported diffusion coefficient of the molecular metastable) and the microwave data be used to obtain the absolute sum of He(2³S) plus He(2³S) in cases where the molecular metastable is important.

In conclusion, a relatively new experimental technique, the steady-state afterglow, has been shown to be useful in the study of metastable species in helium.
V. RECOMBINATION IN LOW TEMPERATURE HELIUM PLASMAS

Previous investigations in this laboratory of an afterglow in helium gas maintained at a gas temperature of \( \sim 4.2^\circ\text{K} \) revealed an anomalous behavior of the visible afterglow light due to application of a short pulse of small microwave power.\(^{15}\) Later a similar behavior was observed with the gas maintained at \( 77^\circ\text{K} \). The work of J. Kaplafka\(^{16}\) consisted principally of studying this behavior at \( 77^\circ\text{K} \) in cataphoretically pure helium with atomic densities of about \( 10^{18} /\text{cc} \) and electron densities in the range of \( 10^8 - 10^{11} /\text{cc} \).

The discharge tube was contained within a section of x-band microwave-guide. Microwaves were used both to disturb the plasma by means of increasing the temperature of the free electron gas and as a probe to measure electron density and electron temperature. Both total and spectrally resolved visible afterglow radiations were observed. Because of the low mean energy of the electron gas the afterglow light is the result of recombination. The light was found to be overwhelmingly molecular in origin after about the first 200 \( \mu\text{secs} \) of afterglow. It originated from decay of the \( \text{He}_2 \) molecule and was found to be proportional to the square of the electron density.

The application of a square pulse of microwaves sufficient to heat the electron gas resulted initially in a partial quenching of the recombination light just as observed in room temperature afterglows. This initial quenching was used to determine the electron temperature dependence of the recombination coefficient. It was found to be approximately inversely proportional to electron temperature which supports the conclusion drawn from the electron density dependence of the light, that only one electron rather than two takes part in the recombination collision. But, unlike the room temperature afterglow, when heating is removed the light increased momentarily to a level
higher than the intensity level before heating (in some cases as much as eight times higher) and in about 70 μsecs returned to the normal level. Microwave probing revealed the surprising fact that electron density increased during heating. It was observed that increasing the electron temperature from 150°K to only 300°K increased electron density by ~50 per cent. It was concluded that there exists a high population of weakly bound electrons which are normally in a near collisional equilibrium with the electron gas. Disturbing the equilibrium by microwave heating results in a net production of free electrons. When heating is removed the electron temperature returns quickly to normal (~1 μsec), the normal weakly bound electron population is slowly re-established but the recombination is large because of the enhancement of electron density during heating.

To explain the high population of weakly bound electrons it is assumed that these states are long-lived. This then may be the first evidence of long-lived states near the ionization limit of the He₂ molecule.
REFERENCES


VI. LIST OF PERSONNEL

The following staff members were supported under this grant:

Faculty

L. Goldstein, Professor
J. T. Verdeyen, Associate Professor
J. H. Cahn, Associate Professor
B. E. Cherrington, Assistant Professor

Research Associates

J. Kaplafka
R. W. Huggins

Research Students

M. E. Fein (M.S. thesis)
R. J. Freiberg (Ph.D. thesis)
J. Kaplafka (Ph.D. thesis)
M. Brooks
VII. LIST OF TECHNICAL REPORTS, THESES, AND SCIENTIFIC ARTICLES

The following technical reports have been published under this grant:


The following articles have been published in scientific journals under this grant:


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