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FROST, III, Milton Arthur, 1940-
SPECTROSCOPIC STUDY OF ELECTRON-ION RECOMBINATION IN AN ARGON PLASMA.

Rice University, Ph.D., 1967
Engineering, mechanical

University Microfilms, Inc., Ann Arbor, Michigan
RICE UNIVERSITY

SPECTROSCOPIC STUDY OF ELECTRON-ION RECOMBINATION IN AN ARGON PLASMA

by

Milton A. Frost, III

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY IN MECHANICAL AND AEROSPACE ENGINEERING AND MATERIALS SCIENCE

Thesis Director's signature:

Houston, Texas
August, 1966
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Introduction

Recent studies of electron-ion recombination have shown that a model including recombination into highly excited bound levels through electron-electron-ion collisions and transitions between the bound levels through electron-atom collisions, as well as the usual radiative transitions, is in good agreement with experiments\textsuperscript{1-4}.\footnote{Numbers refer to References listed on page 38.}

Experiments on the recombination of a decaying helium plasma of an arc-jet using detailed spectroscopic measurements have been performed\textsuperscript{4} which led to a determination of average excitation cross sections of several levels of helium which were in approximate agreement with values calculated by the classical expression of Gryzinski\textsuperscript{5}. The recombination coefficient calculated by Bates, Kingston, and McWhirter\textsuperscript{3} were in fair agreement with the values inferred from these experiments.

It is the purpose of this thesis to report experimental investigations of the recombination of a decaying argon plasma jet in a low-density wind tunnel. Detailed spectroscopic studies were made. It was not possible to measure the decay rate of the plasma directly, and all results were based on measurements of the absolute intensities of the various radiative transitions of the argon atom. Excitation cross sections were calculated using the approach of Gryzinski with an approximation suggested by Byron, Bortz, and Russell\textsuperscript{6}. With these cross sections together with radiative transition probabilities which have been compiled by Adcock and Plumtree\textsuperscript{7} for argon lines, the rate of electron-ion recombination can be calculated using the approach developed by Bates, Kingston, and McWhirter\textsuperscript{3}. 
It has been pointed out\textsuperscript{8} that the presence of metastable atoms in inert gas plasmas may significantly affect experimental measurements. Their influence has been accounted for here using the approach of Brewer\textsuperscript{9}. It is interesting to note that the "continuous" afterglow of such a plasma may be accounted for by the presence of metastable atoms, whereas former approaches left this phenomenon unexplained.
I. Determination of the Recombination Coefficient from Spectroscopic Data

Before 1962, radiative and collisional mechanisms of recombination and de-excitation had been considered separately in analyses of plasmas. Such analyses actually described the recombination processes provided the plasmas considered were sufficiently tenuous (radiative) or sufficiently dense (collisional). The approach of Bates, Kingston, and McWhirter\textsuperscript{3} combined the above two classes of recombination mechanisms; the resulting method can be used to analyze most any laboratory plasma. This method reduces to the above two separate cases where the plasma is sufficiently thin or dense; the general loss mechanism in this "collisional-radiative" model is a result of interactions between the collisional and the radiative processes and is not simply a sum of the two.

The Rate Equation.

Of the types of reactions that may occur in a plasma stream, the following are expected to be the most significant:

\[
\begin{align*}
A^+ + e + e &\rightarrow A(n) + e \quad \text{Collisional (3-body) Ionization and Recombination} \\
A^+ + e &\rightarrow A(n) + hv \quad \text{Radiative Ionization (Photo-ionization) and Recombination} \\
A(m) + e &\rightarrow A(n) + e \quad \text{Collisional Excitation and De-excitation} \\
A(n) &\rightarrow A(m) + hv \quad \text{Radiative Excitation and Decay}
\end{align*}
\]

where $A^+$ refers to an ionized atom in the ground state, $A(n)$ denotes an excited atom, $m$ and $n$ refer to the principal quantum numbers of the discrete levels, and $v$ is the frequency of emitted or absorbed radiation. Because the excited levels turn out to have a Boltzmann distribution, the de-excitation rate must be considerably larger than the net rate of
recombination. Processes involving absorption of radiation were not considered, as the plasma was expected to be optically thin. Self-absorption, which was observed in the infrared spectra, will be accounted for in section V. Absorption of resonance radiation will be accounted for below.

Thus, the reactions considered were:

\[ \begin{align*}
K_{m,n} & \quad \text{Ar}(m) + e \xrightarrow{m,n} \text{Ar}(n) + e \\
K_{n,m} & \quad (n > m) \\
A_{n,m} & \quad \text{Ar}(n) \xrightarrow{n,m} \text{Ar}(m) + h\nu
\end{align*} \]

where \( \text{Ar} \) refers to an argon atom, \( K_{m,n} \) is the rate coefficient for collisional excitation, \( K_{n,m} \) is the rate coefficient for collisional de-excitation, and \( A_{n,m} \) is the radiative transition probability. Based upon these reactions, the rate equation for the number density \( N_n \) of excited atoms \( \text{Ar}(n) \) is

\[
\frac{dN_n}{dt} = \sum_{m=3}^{n-1} (N_e N_m K_{m,n} - N_e N_n K_{n,m}) + \sum_{m=n+1}^{\infty} (N_e N_m K_{m,n} - N_e N_n K_{n,m}) \\
- \sum_{m=4}^{n-1} N_n A_{n,m} + \sum_{m=n+1}^{\infty} N_m A_{m,n}
\]

(1)

Note that \( m \) in this equation refers to a level above or below \( n \). The minimum value for \( n \), corresponding to the ground level in argon, is 3. Resonance radiation trapped by the plasma has been accounted for in the rate equation (next to last term) by dropping all radiative transitions to the ground level. This is discussed further below.

The Recombination Coefficient

Following Robben, Kunkel, and Talbot\(^4\), the rate equation may be written in the form
\[
\frac{1}{N_e} \frac{dN_n}{dt} = - \sum_{m=3}^{n-1} G_{n,m} D_{n,m} + \sum_{m=n+1}^{\infty} G_{m,n} D_{m,n} - L_n
\]  

(2)

where the following definitions have been used:

\[
D_{m,n} = N_m / g_m - (N_n / g_n) \exp (U/kT_e), \quad G_{m,n} = g_m K_{m,n}.
\]

\[
D_{n,m} = - D_{m,n} \exp (-U/kT_e), \quad U = E_n - E_m,
\]

\[
L_n = \frac{1}{N_e} \left[ N_n \sum_{m=4}^{n-1} A_{n,m} - \sum_{m=n+1}^{\infty} N_m A_{m,n} \right]
\]

The principle of detailed balance which states that the forward and reverse rates are equal when all the constituents are at thermodynamic equilibrium \([N N K_{m,n}] = [N N K_{n,m}]\) has also been used.

The number density of the ground level \(N_2\) is assumed to be much larger than all the other \(N_n\), and such properties of the plasma as density and temperature change slowly with time so that all the \(dN_n/dt\) except \(dN_2/dt\) may be set equal to zero\(^{3,4}\). Therefore, \(-dN_2/dt\) is equal to the recombination rate, \(dN_e/dt\), which according to this model is

\[
\frac{dN_e}{dt} = -N_e G_{4,3} D_{4,3}
\]

(3)

since \(D_{5,3} \ll D_{4,3}\) etc. For \(n = 4\), the rate equation is

\[
L_4 + G_{4,3} D_{4,3} - \sum_{m=5}^{\infty} G_{m,4} D_{m,4} = 0
\]

(4)

and

\[
L_4 = - \frac{1}{N_e} \sum_{m=5}^{\infty} N_m A_{m,4}.
\]

Therefore, the recombination coefficient \(\alpha\) may be written

\[
\alpha = - \frac{1}{2} \frac{dN_e}{dt} = \frac{1}{N_e} \sum_{m=5}^{\infty} \left[ G_{m,4} D_{m,4} + \frac{N_m}{N_e} A_{m,4} \right]
\]

(5)
Because of the scattered spacing of the various energy levels in argon, the recombination coefficient will be much more meaningful if this equation is written summing over the various subshells rather than the shells. It can be shown that the recombination coefficient may then be written

$$\alpha = \frac{1}{N_e} \sum_{m=1}^{\infty} \sum_{l=1}^{n'} \sum_{j=1}^{l'} \left[ (K_{mn} N_{4s j^0} - K_{4s j^0} N_{mn} N_{4s j^0}) + \frac{N_{mn}}{N_e} A_{mn,4s j^0} \right]$$  \hspace{1cm} (6a)

or more simply

$$\alpha = \frac{1}{N_e} \sum_{m=1}^{\infty} \sum_{l=1}^{n'} \sum_{j=1}^{l'} \left( Q_{mn},4s j^0 D_{mn},4s j^0 + \frac{N_{mn}}{N_e} A_{mn,4s j^0} \right)$$  \hspace{1cm} (6b)

where \( m = 1,2,3, \ldots \) correspond to the \( 4p,5p,6p, \ldots \) subshells, respectively (transitions from other orbitals to the \( 4s \) orbital are not allowed). \( n' \) is the total number of levels in the \( m \) subshell, and \( n \) and \( j \) refer to various levels in the \( m \) and \( 4s \) subshells, respectively. The energy level diagram in Appendix B shows many of the observed transitions between the \( 4s \), \( 4p \), and \( 5p \) subshells. Figure 1 gives the rate coefficients \( G_{n,m} \) for transitions between various subshells.

**Determination of the Rate Coefficients**

The excitation rate coefficient may be written in terms of the inelastic cross section as

$$K_{n,n+1} = \frac{1}{N_e} \int_{0}^{\infty} Q_{n,n+1}(E) v(E) \frac{dN_e(E)}{dE} dE$$  \hspace{1cm} (7)

where \( U \) is the excitation energy \( E_{n+1} - E_n \), \( Q_{n,n+1}(E) \) is the excitation cross section, \( v(E) \) is the electron velocity, and \( dN_e(E)/dE \) is the electron energy distribution. In this work this distribution is assumed to be Maxwellian:
\[
\frac{dN_E(E)}{dE} = \frac{2N_e}{\pi^{3/2}(kT_e)^{3/2}} E^{1/2} \exp\left(-\frac{E}{kT_e}\right)
\]

where \(T_e\) is the (free) electron temperature. The inelastic cross sections have been calculated according to Gryzinski's theory\(^5\) for excitation of atomic levels by electron impact. These cross sections were shown to be functions of the excitation energy \(U\) and a factor \(g_j\) which depends upon the energies of the bound \((E_1)\) and the free \((E_2)\) electrons and the excitation energy. Gryzinski plotted curves for the variation of \(g_j\) with \(E_2/U\) for four values of \(E_1/U\). Byron, Bortz, and Russell\(^6\) have plotted additional curves and have suggested an approximation to the variation of \(g_j\) to simplify computations. They note that if the "gap" energy \(U\) is of the order of or less than the electron thermal energy, and regarding the energy levels above and below the gap as continuous, the simpler expression for the cross section (Ref. 5, Eqn. 26) is a good approximation. They use the linear approximation \(g_j = B(E_2/U)\), where \(B\) is a constant for a particular value of energy \(E_1\). This linear approximation is shown to be valid when \(kT_e\) lies in the range \(0.01E_1 \leq kT_e \leq U + 0.1E_1\). As an example, if \(E_1\) (the same as the binding energy of the level being excited) is 1 ev. and the excitation energy is 0.2 ev., the linear approximation is valid for electron temperatures between 1160 K and 3480 K. The variation of \(B\) with \(E_1/U\) plotted by Byron, Bortz, and Russell can be analytically represented by

\[
B = 0.2 \left(\frac{E_1}{U}\right)^{-0.323}
\]

to better than 5%.

The excitation rate using this approximation is then found to be
\[ K_{n,n+1} = \sum_{\text{number of outer shell electrons}} \left[ \frac{8kT_e}{\mu m_e} \right]^{\frac{1}{2}} \frac{2\sigma_0 B}{2U^2} \exp \left( -\frac{U}{kT_e} \right) \left( 1 + \frac{kT_e}{U} \right) \] (10)

where \( \sigma_0 = 6.56 \times 10^{-14} \text{ cm}^2 \text{ ev}^{-2} \). Using the principle of detailed balance and the Boltzmann relation for \( N_n \) and \( N_m \), the de-excitation rate coefficient becomes

\[ K_{n,n-1} = \sum_{\text{number of outer shell electrons}} \left[ \frac{8kT_e}{\mu m_e} \right]^{\frac{1}{2}} \frac{2\sigma_0 B}{2U^2} g_n \frac{g_{n-1}}{U} \left( 1 + \frac{kT_e}{U} \right) \] (11)

Values for the de-excitation rate constant as given by this approximate relation for various transitions in argon for an electron temperature of 0.17 ev. are plotted against the binding energy \( E_1 \) in Figure 1.

Two curves calculated for helium are shown for comparison: the upper curve is for transitions between levels whose principal quantum numbers differ by unity; the lower curve is for transitions between levels having principal quantum numbers differing by two. The upper curve agrees very well with a similar curve given by Robben, Kunkel, and Talbot\(^4\). This agreement justifies the use of the linear approximation for \( g_j \). Values of the energies and degeneracies for the calculations were taken from C.E. Moore\(^{10}\).

**Resonance Radiation Trapping**

The absorption coefficient \( \mu \) of an atomic line is determined by the oscillator strength \( f \) of the line and the line shape. Only the strongest transitions (\( f \approx 1 \)) will be considered since these dominate the effect of radiation on excited state populations. Considering Doppler broadening only (other types of broadening are negligible\(^{11,12}\)), the line width \( \Delta \nu \) and the absorption coefficient at the line center...
are given by\textsuperscript{13-15}:

\[
\mu = \frac{\pi e^2 N_f^2 f_{l,u}}{m_e c \Delta \nu}
\]

\[
\Delta \nu = 2 \left[ \frac{2kT_a}{m_a} \ln 2 \right]^{1/2} \frac{\nu}{c} = 7.12 \times 10^{-7} \left[ \frac{T_a}{M_a} \right]^{1/2} \nu
\] (12)

where \( \nu \) is the line frequency, \( m_a \) is the atomic mass, \( M_a \) is the molecular weight of the atom, \( T_a \) is the atom temperature, \( N_f \) is the number density of the lower state, \( f_{l,u} \) is the oscillator strength from the lower level \( l \) to the upper level \( u \), and \( m_e \) is the electron mass. Oscillator strengths may be computed from known transition probabilities using the relation\textsuperscript{16}

\[
f_{l,u} = \frac{m_e c^3}{8(\pi e)^2} \frac{g_u}{g_l} A_{l,u}
\] (13)

Assuming that an optical depth of 1 cm. is the division between optically thin and optically thick (\( \mu = 1 \) cm\(^{-1} \)) and using \( \nu = 3 \times 10^{15} \) sec\(^{-1} \) (corresponding to a wavelength of 1060 \AA; approximately the wavelength of argon resonance radiation), it can be shown that for \( 1000^\circ K \leq T_a \leq 20,000^\circ K \), the ground state number density (\( l = 3 \)) would lie between \( 1.43 \times 10^{11} \) and \( 6.52 \times 10^{11} \) cm\(^{-3} \). This means that for a number density in the neighborhood of \( 10^{17} \) cm\(^{-3} \), the intensity of this radiation will have fallen to 1/e of its initial value in \( 10^{-5} \) cm. Such a plasma would therefore be optically opaque for all transitions from upper states to the ground state.

In the rate equation, absorption of resonance radiation was taken into account by neglecting all transitions to the ground level. This means that the energy that such transitions would have released, only to be completely absorbed by the plasma, is considered never to have been released at all. Thus, only collisional de-excitation to the ground level is included in the rate equation.
II. Effect of the Metastable States on Experimental Measurements

An atom is in a metastable state when its excited electron is in an energy level from which it may not radiatively decay. The lowest excited level of the inert gas atoms have metastable states. The lifetime of a metastable atom depends upon a mode of de-excitation other than radiative decay. The lowest excited configuration of an argon atom is the \( 3p^5 4s \). As seen from the energy level diagram (Figure 2), the 4s subshell is split into four levels. The \( 3P_0 \) and the \( 3P_2 \) levels are designated metastable because a transition to the ground level \( ^1S_0 \) would violate the inner quantum number selection rule. Transition from the other two levels, \( ^1P_1 \) and \( ^3P_1 \), are allowed; wavelengths of the emitted radiation are 1049 Å and 1067 Å, respectively, which lie in the ultraviolet region of the spectrum.

Explanation of the Afterglow.

The influence of metastable atoms upon measurement evaluation in an argon plasma was brought out by Brewer and McGregor in their attempt to explain some of the spectral characteristics and the luminosity of the expanding plasma stream produced in an arc-jet. The intense radiation from the core was attributable to excitation of the argon atoms by electrons in the DC arc and by spontaneous decay of excited atoms; since the lifetime of an excited state is of the order of \( 10^{-8} \) seconds, an atom would be expected to radiate within a distance of 0.01 mm from its point of excitation in a stream having a velocity of 1 km/sec.

Downstream, the radiation could not be explained by collisional excitation from the ground level followed by collisional or radiative de-excitation because of the low temperature there. Radiative re-
combination proceeds at such a slow rate that most of the radiation would be expected to occur downstream of the test section.

The process of dissociative recombination has also been suggested to help explain this problem. However, little evidence has been given of the presence of molecular ions in an arc-jet. Their lifetime is of the order of $10^{-6}$ seconds or less, so their decay in the stream would not be observed.

It therefore appears that the plasma afterglow may not be attributed to any of the above excitation processes in the arc. What, then, could be causing the excitation downstream? The answer can be found by examining the peculiarities of metastable atoms. A metastable atom is in a state of high potential energy, and only a small amount of energy need be supplied to further excite it to a higher level. Radiative transition from the metastable level is impossible, and because of the large amount of energy that must be released in a collisional de-excitation process, it is quite likely that a metastable atom will be further excited. Radiative transition can then occur from this higher level. A considerable number of metastable atoms are produced in the arc, and because of the rather long lifetime of these atoms (of the order of $10^{-3}$ seconds), such a process of collisional excitation followed by radiative decay could well account for the continuous afterglow of the plasma stream.

Influence of Metastable Atoms on Measured Excited State Populations

Brewer and McGregor have accounted for the influence of metastable atoms on spectral line emission by considering two different types of atoms present in the plasma: (1) the usual argon atom having its ground state energy equal to zero and (2) the metastable argon atom having a
ground state energy equal to the first excitation potential of the argon atom. A result of this modification is that populations of excited levels are considerably increased by the presence of only a small concentration of metastable atoms. Therefore, temperature determination schemes using absolute line intensity methods must be corrected. Other temperature determination procedures, such as the relative line intensity, line slope, and peaking function methods, are not affected.

The intensity of radiation emitted from the plasma is given by

\[
I_{n,m} = \frac{\nu_{n,m} A_n A_m^m n}{Z_g} \frac{N_g \exp \left( -\frac{E_n}{kT_e} \right)}{Z_g}
\]  

(14)

where \(I_{n,m}\) is the intensity of the emitted radiation whose frequency is \(\nu_{n,m}\) from the level \(n\) having energy \(E_n\) and degeneracy \(g_n\) to the level \(m\). \(N_g\) is the ground state number density of the emitting specie, and \(Z_g\) is the specie partition function. Contributions to the intensity due to absorption and induced emission have been neglected, as they are usually small.

An approximate distribution function which takes into account the metastables is formed considering a gas composed of argon atoms having a zero energy ground state (type 1) and atoms having a ground state energy \(E_M\) (type 2) from which level further decay is assumed to be impossible. Assuming that the excitation process is governed by collisions as usual, the Boltzmann relation may be used to obtain the number of each type of atom in a state \(n\). For the first type,

\[
N_{n1} = N_1 \frac{g_n}{Z_3} \exp \left( -\frac{E_n}{kT_e} \right)
\]

and for the second type,
\[ N_{n2} = N_2 \frac{g_n}{Z_M} \exp \left[ -(E_n - E_M)/kT_e \right] \]

where \( Z_M \) is the partition function for the metastables. The above distributions are correct since only very weak interactions are expected between the two distributions. Recognizing that \( N_{n1} + N_{n2} = N_n \) and that \( N_g = N_1 + N_2 \), and denoting the number of metastables \( N_2 \) as \( N_M \),

\[ N_n = \frac{g_n}{Z_3} (N_3 - N_M) \exp \left[ -(E_n/kT_e) \right] + \frac{g_n}{Z_M} N_M \exp \left[ -(E_n - E_M)/kT_e \right] \]

Therefore, the intensity of a spectral line is now written

\[ I_{n,m} = h \nu_{n,m} A_{n,m} g_n \left( N_3 - N_M \right) \exp \left( \frac{-(E_n/kT_e)}{Z_3} \right) \sum_{p=0}^{\infty} g_p \exp \left( -(E_p/kT_e) \right) \]

\[ + N_M \frac{\exp \left[ -(E_n - E_M)/kT_e \right]}{Z_M} \sum_{p=1}^{\infty} g_p \exp \left[ -(E_p - E_M)/kT_e \right] \]

(15)

For an estimate of the proportion of metastable atoms present in the plasma, assuming a one-specie gas in the stagnation chamber of the arc-jet at a temperature \( T_0 \), the density of metastables is given by

\[ \frac{N_M}{N_3} = \frac{g_M}{Z_3} \exp \left( -E_M/kT_0 \right) \]

(16)

Comparing the old and the new expressions for \( N_n/N_3 \):

\[ \frac{N_n}{N_3} \text{old} = \frac{g_n}{Z_3} \exp \left( -E_n/kT_e \right) \]

(17a)

\[ \frac{N_n}{N_3} \text{new} = \frac{g_n}{Z_3} \exp \left( -E_n/kT_e \right) \left[ 1 - \frac{N_n}{N_3} + \frac{N_M Z_2}{N_3 Z_M} \exp \left( E_M/kT_e \right) \right] \]

(17b)

Using a \( T_0 = 8000^\circ K \), and since \( Z_g = Z_3 = 1 \) (accurate to \( 20,000^\circ K \)), it is found that \( N_M/N_3 \approx 10^{-7} \) (using an average of 11.637 ev. for the
energy of the metastable level and taking $g_M = 6$). Considering $T_e = 5000^\circ K$ at a station of observation in the plasma stream, $N_n$ is found to be a factor of about $10^5$ larger than would have been computed neglecting the influence of the metastable atoms. Since the plasma in the experiments corresponds closely to the conditions specified for this example, clearly the first two terms in the brackets in Equation (17b) may be neglected, and the expression for the number density may be written

$$N_n = N_M g_n e^{E_n - E_M/kT_e}$$

which is just a statement of a Maxwellian distribution of excited levels in a metastable atom gas.

The value used in the example for the partition function for the metastable atoms, $Z_M$, was 7.32 for $T_e = 5000^\circ K$. The partition function was determined using data from Reference 10. The difference in the value of $Z_M$ calculated terminating the series after 50 terms was less than 1% smaller than the value calculated using 74 terms. The variation of $Z_M$ with temperature was found to be

$$Z_M = 6 + 36 \exp(-18,200/T_e) + 60 \exp(-28,200/T_e) + 12 \exp(-29,300/T_e) + 36 \exp(-34,200/T_e) + 60 \exp(-37,200/T_e) + 12 \exp(-38,200/T_e) + 82 \exp(-38,600/T_e) + 36 \exp(-40,200/T_e) + \ldots$$

where $T_e$ is in $^\circ K$. In the determination of $Z_M$, the energy levels within a particular configuration of the atom have been averaged, and the degeneracy of the set is the sum of the degeneracies of the individual levels. It is apparent that each additional term used in the series approaches zero very slowly for temperatures in the vicinity of 5000$^\circ K$. In fact, for temperatures nearing 10,000$^\circ K$, calculations of meaningful
values for $Z_M$ will not be possible. For $T_e = 10,000^\circ$K, the difference in the value of $Z_M$ computed terminating the series after 50 terms is 12% smaller than the value calculated using 74 terms.

It should be mentioned how these results affect the theory of section I regarding the recombination coefficient. It may be observed that the number density enters only in ratios like $N_n/N_m$. Using the new expression for $N_n$, it is found that

$$\frac{N_n}{N_m} = \frac{g_n}{g_m} \exp \left[ -\frac{E_n - E_m}{kT_e} \right]$$

which is the same as before. Therefore, there are no changes necessary.
III. Interpretation of Spectroscopic Data

Each line in the observed spectra emitted by the plasma jet corresponds to a transverse slice of the plasma approximately 0.3 mm wide. Because of the stigmatic nature of the optics in the spectrograph, there is a one to one correspondence between points along this slice of the plasma and points on each spectral line. As a result, the distribution of intensity along a spectral line corresponds to the distribution of intensity across the plasma.

However, before the intensity distribution may be ascertained for a given line, the response of the photographic film must be determined (since response is a function of wavelength). Further, since the observed intensities represent the intensity at the center of the plasma plus contributions from the plasma surrounding it, a correction must be employed which effectively subtracts out these additional contributions. Finally, the resulting intensities must be placed on a common scale.

Once true intensity distributions are obtained, the electron temperature may be determined; when the intensities are placed on an absolute scale, determination of the populations of the excited levels is possible.

Transformation of Film Transmission Distributions to Intensity Distributions via the Characteristic Curve

When the exposed region of a film is analyzed using a densitometer, light of constant intensity is passed through that region of the film. The transmitted light activates a photomultiplier tube which drives a recorder having a scale of 0 - 100% transmission. Transmission is defined as the ratio of the transmitted light to the incident light.
If different portions of the film are exposed to different known relative intensities, or to a constant intensity source at various exposure times, it is then possible to determine a relationship between the photographic response and the relative exposure. Exposure is defined as the product of the intensity of the source and the time of exposure. The characteristic curves used in this thesis were curves of log (percent transmission) versus log (relative intensity). Since photographic response is a function of the wavelength of the radiation to which the film is exposed, there is a different characteristic curve for each wavelength line studied. Exposure of the film to an argon bulb for several different lengths of time was used to obtain the information needed to construct this calibration curve. This curve was then used to convert the percent transmission distributions of the spectral lines taken of the plasma jet to relative intensity distributions.

Transformation from Observed Intensity Distributions to True Intensity Distributions via the Abel Inversion Technique

When an axially symmetric plasma is viewed from the side, the spectrograph sees an intensity which is different from the actual radial intensity. This is due to contributions from the radiating plasma distributed symmetrically about the axis. The true intensity distribution may be generated from the observed distribution using an Abel inversion scheme. Following Pearce \(^{19}\), the plasma is imagined to be divided into concentric rings as shown in Figure 3a. The intensity is assumed to be constant in each ring. The observed distribution is made up of contributions from one or more of these rings. The radial distribution is essentially equal to the observed intensity along the x-axis in Figure 3a minus the radial intensities of the off-axis segments.
contributing in the same slice of width $\Delta x$ viewed by the spectrograph. The sizes of the various segments are tabulated as area coefficients. 

If the plasma is assumed to be divided into $k$ rings, the true intensity $I(r)_k$ of the segment having an area coefficient $a_{k,k}$ will be

$$I(r)_k = C \frac{I(x)}{a_{k,k}}$$  \hspace{1cm} (20)

The true intensity $I(r)_n$ of some segment having an area coefficient $a_{n,n}$ is found to be

$$I(r)_n = \frac{C}{a_{n,n}} \left[ I(x)_n - \sum_{p=n+1}^{k} a_{p,n} I(r)_p \right]$$  \hspace{1cm} (21)

which indicates a stepwise solution for the radial distribution beginning at the edge of the plasma and working inward toward the axis of symmetry.

$C$ is a constant that is adjusted to place the true and the observed distributions on the same relative scale which is done by making the tails of the $I(x)$ and $I(r)$ curves coincide.

If the plasma is found to exhibit some degree of self-absorption, then this must be taken into account when calculating the radial intensity distribution. The determination of various absorption coefficients and the resulting inversion are based on a similar approach by Pearce. Referring to Figure 3b, the radiation from the plasma observed at one point, $I(x)$, is compared to the radiation observed at the same point, $I'(x)$, with a plane mirror behind the plasma having a reflectance $R$. Defining the absorption coefficient as $\Delta I \propto \mu I a_{p,n}$, it is easily shown that the absorption coefficients may be found for each of the radial rings from the following equations:

$$\mu_k = \frac{1}{a_{k,k}} \left[ 1 - \left( \frac{I'(x)_k - I(x)_k}{R I(x)_k} \right)^2 \right] \hspace{1cm} (n = k)$$  \hspace{1cm} (22)
\[ \mu_n = \frac{1}{a_{n,n}} \left[ 1 - \frac{\left( I'(x)_n - I(x)_n \right)}{R \ I(x)_n} \right] \frac{1}{k} \prod_{r=n+1}^{k} \left( 1 - \mu_r a_{r,n} \right) \quad (n < k) \quad (23) \]

Thus, taking two spectra, one without a mirror and one with a mirror, is sufficient to determine all the \( \mu_n \)'s. The inversion is performed in the same manner as before; the resulting equations are

\[ I(r)_k = \frac{I(x)_k}{(2 - \mu_r a_{k,k}) a_{k,k}} \quad (n = k) \quad (24) \]

\[ I(r)_n = I(x)_n - \sum_{r=1}^{n-1} I(r)_r a_{r,r,n} \left[ \prod_{i=1}^{r-1} \left( 1 - \mu_i a_{i,i,n} \right) \right] \times \]

\[ \times \left[ 1 + (1 - \mu_r a_{r,n}) \prod_{i=r+1}^{n} (1 - \mu_i a_{i,i,n})^2 \right] \frac{a_{n,n}}{\prod_{i=1}^{n-1} \left( 1 - \mu_i a_{i,i,n} \right)(2 - \mu_n a_{n,n})} \quad (n < k) \quad (25) \]

A Fortran I program is given in Appendix D for the Abel inversion that can be used for either of the above two cases. In the experiments, a concave spherical mirror was used rather than a plane mirror; it is expected that because its radius of curvature is quite a bit larger than the plasma diameter, the assumption of a plane mirror is satisfactory.

It should be emphasized that when several distributions are being compared, the number of rings used in the inversion processes must be the same. Also, a spectral line should be photographed such that each end drops off to the background level, if possible, where the plasma meets the unexcited gas surrounding it in the test section.

Another approach for deducing the radial distribution was suggested by Pearce. This makes use of a chart of \( I(x) \) curves which are matched with the observed intensity curves. Solutions for the radial distri-
bution are presented in terms of a table of shape factors. Use of these shape factors as presented become quite tedious when inverting entire distributions since each entry $A_{k,k}$ must be divided by an area-coefficient $a_{k,k}$; therefore, in Appendix D, a table is presented which gives values of $I(r)$ directly.

**Determination of Electron Temperature**

There are a number of methods for determining the electron temperature. One method, selected because of its superior accuracy over the other methods, was used in the experiments and is the only one described here.

The distribution of the population of the different energy levels was found in the previous section to be given by

$$\frac{N_n}{g_n} = \frac{N_M}{Z_M} \exp \left[ - \frac{E_n - E_M}{kT_e} \right]$$

to a good approximation. Taking the logarithm

$$\ln \left[ \frac{N_n}{g_n} \right] = \ln K - \frac{E_n}{kT_e}$$

(26)

where $K$ is a constant for a fixed $T_e$. Therefore, a plot of $\ln \left( \frac{N_n}{g_n} \right)$ versus $E_n$ is a straight line whose slope yields $T_e$ directly. Notice that only the constant $K$ would have been altered had the Maxwellian distribution been used for an ordinary argon gas.

**Determination of Excited State Number Densities**

Inspection of the equation for the intensity of a spectral line

$$I_{n,m} = h \nu_{n,m} A_{n,m} N_n$$

shows that the number density of an excited state $n$ can be found provided the absolute intensity and the spontaneous transition probability are known. Values of transition probabilities
for argon have been gathered by Adcock and Plumtree\textsuperscript{7}; a portion of their tabulation has been reproduced in Appendix C. Absolute intensities were determined using a tungsten ribbon filament lamp calibrated for the wavelength range of 2500\textmu m to 26,000\textmu m. The procedure for calculating absolute intensities using a standard of spectral radiance is described in Appendix F.

Determination of Free Electron Density

The higher quantum levels are assumed to undergo rapid transitions caused by atomic collisions with free electrons. Therefore, the upper levels are assumed to be in equilibrium with the free electrons, and the Saha equation may be used to compute their density:

$$\frac{N_e N_{Ar^+}}{N_{Ar(n)}} = \frac{g_e g_{Ar^+}}{g_{Ar(n)}} \left(\frac{2\pi m_e kT_e}{h^3}\right)^{3/2} \exp \left[-\frac{E_I - E_n}{kT_e}\right]$$  \hspace{1cm} (27)

where $E_I$ is the ionization energy. Assuming the plasma to be electrically neutral ($N_e = N_{Ar^+}$) as usual, the electron density ($\text{cm}^{-3}$) may be determined from the relation

$$N_e^2 = 3.63 \times 10^{22} T_e^{3/2} \frac{n_{\infty}}{g_\infty}$$  \hspace{1cm} (28)

where $T_e$ is in ev. and $n_{\infty}/g_\infty = n_n/g_n \exp \left[-(E_I - E_n)/kT_e\right]$. $n_{\infty}/g_\infty$ may be found from extrapolating the curve of $\ln (n_n/g_n)$ versus $E_n$ to the ordinate where $E_n = E_I$. 
IV. Experimental Equipment

The experimental investigations reported in this thesis were carried out in a low-density wind tunnel using argon as the working fluid. A plasma was produced by passing argon through a DC arc. The arc was initiated by a high frequency starter; power to support the arc was supplied by a 24 kw. welding generator. For the present experiments, the plasma generator was run at about 5 kw. Arc chamber pressure was typically 4.75 psia; arc chamber temperature was estimated to be 8200°K.

The plasma left the arc chamber through an axially symmetric converging nozzle, emerging as a free jet in the test chamber where the pressure was maintained between 1 and 2 mm Hg. In the test section is a traverse mechanism for positioning various probes in the flow. Quartz windows for spectrographic observations are mounted in the walls of the test section. A schematic diagram of the plasma tunnel is shown in Figure 4.

Argon was supplied to the arc chamber from a reservoir at 30 psig, through a metering orifice. A 0.043 inch diameter orifice was used which produced a flow rate of 0.575 grams per second at 74°F. The flow rate was measured with a Fischer and Porter Tri-Flat variable area flowmeter type FP-1/4-25-G-5, and its calibration was accomplished using data given in the handbook for such flowmeters.

In the arc chamber, a thoriated tungsten anode 1 inch in diameter was located about 0.2 in. below the copper nozzle which served as the cathode. The nozzle had a 0.25 in. diameter throat. All parts were water-cooled. Power to the arc was supplied through water-cooled copper coils built into the electrodes which had the desirable effect of magnetically inducing a force on the arc causing it to spin. The
argon entered the arc chamber tangentially and traveled in the same sense as the rotating arc. No arc attachment and negligible electrode erosion has been observed. The free jet was generally quite steady and symmetrical in appearance at the operating conditions described above.

Most of the test data was recorded on a Honeywell model 1012 Visi- corder which provided continuous graphical data for the duration of the tests. Tests generally lasted from 1 to 10 minutes. Occasional data were taken with a Tektronix type 545A or 561A oscilloscope.

Pressure in the arc chamber was measured with a Statham type PA208TC strain gauge pressure transducer. Supply pressure was monitored with a Bourns model 304 pressure potentiometer. Test section and probe pressures were measured using Statham type PM5TC differential pressure transducers.

A set of three consoles designed to automatically run the experiments and record the test data was built by the author and W.L. Boddie, a graduate student also doing research in the Plasma Dynamics Laboratory at Rice University. Tests were automatically run by a Test Control Console which essentially is a programmable sequential timer containing a plugboard that is programmed by each user for his particular test sequence. Besides the required test functions, certain "checks" are also programmed into the test sequence to ensure that all equipment is functioning properly before the test is initiated as well as during the test; several failsafe checks have been built into particular subsystems where such a system must operate only under certain conditions. Tests may be operated from this console, but generally are operated by remote control.

An Instrumentation Console handles all the excitation circuits for the various transducers and channels the incoming data through compensating,
filtering, or other networks to the Visicorder. This console is also programmed by each user. Additional circuitry needed to modify the incoming data may easily be wired in on any one of twenty plug-in cards. Each card handles up to four circuits. Capacity of the Visicorder is thirty-six channels; design capacity of the Instrumentation Console is twenty channels, although up to thirty-six can be handled if necessary.

A photograph of this equipment is shown in Figure 5. The Test Control Console is at the left; the Instrumentation Console is on the right. The Visicorder rests on a small console between the two; this contains DC power supplies for the excitation circuits plus storage room for Visicorder accessories.

Figure 6 shows a general view of the laboratory; the group of consoles appear at the left, the test chamber is in the center, and the spectrograph is on the right. All power, water, instrumentation, and gas lines are brought in from overhead. Figure 7 is a photograph of the argon free jet having the operating conditions described above (a scale is superimposed to indicate its size). Figure 8 shows the free jet with a flat-ended cylindrical probe in the flow.

Spectroscopic observations were made with a Bausch and Lomb model 11, 1.5 meter stigmatic spectrograph. A schematic diagram of the optical train is shown in Figure 4. A lens system was set up to provide a demagnification of 5, so that an entire transverse slice of the plasma could be photographed. In order to obtain greater wavelength coverage, the spectrograph was modified to contain two gratings which can easily be interchanged during a single test. One grating gives coverage in the 3700Å to 7400Å range; the other gives coverage
in the 4900Å to 9800Å range. Spectra were recorded on Kodak type 1-N spectroscopic film which is sensitive from below 3700Å to about 8800Å. The films were analyzed on a Jarrell-Ash model 23-050 micro-densitometer.

Located on the spectrograph table is a radiation standard used for absolute intensity calibration of plasma jet spectra. The primary standard is a tungsten ribbon filament lamp which is a General Electric type 30A/T24/17 bulb calibrated by Eppley Laboratories to National Bureau of Standards specifications for spectral radiance from 2500Å to 26,000Å when run at 35.0 amperes AC. Current flowing through the standard is monitored within 0.1 percent using an Electro-Instruments precision AC-DC differential voltmeter. A schematic of the standard lamp circuit is shown in Figure 9. The standard is viewed by the spectrograph using an alternate, but identical, optical path to that used in observing the plasma. Also located on the spectrograph table is a modified Cenco 5000 volt power supply and an argon Pluecker tube used for emulsion calibration. A photograph of this equipment is shown in Figure 10.

Numerical calculations were carried out on an IBM 1620 Data Processing System.
V. Experimental Results

Spectroscopic measurements were taken at a point one inch downstream of the exit of the converging nozzle; at this location the Mach number was estimated to be 2.2. The line profiles were symmetrical and were found to fit curve 19 used in the shape factor method of radial inversion discussed in section III (see Figure 11). Because most of the line profiles were similar in shape, the shape factor technique was by far the fastest way of inversion. Infrared lines, however, showed some self-absorption, and the numerical inversion procedure had to be used for these lines.

The spectra revealed, besides argon I, only N₂ bands and the Hα and Hβ lines of the Balmer series. No argon II lines were observed. The argon used contained approximately 6 ppm of impurities, half of which was N₂; other impurities were: O₂ - 1 ppm, H₂ - 1 ppm, others - 1 ppm. The absence of other lines indicated the cleanliness of operation of the plasma jet. No continuum radiation was observed.

Results of the experimental measurements are shown in Figure 12, a plot of centerline values of \( \ln \left( \frac{N_n}{g_n} \right) \) versus \( E_n \). From the slope of this curve, the electron temperature has been determined to be 4830°K with an estimated error of less than 20 percent. (The size of the error is largely due to operations involving interpretation of the photographic films.) Assuming the plasma to be electrically neutral, \( N_e \) was determined to be \( 9.3 \times 10^{12} \text{ cm}^{-3} \). The number density of argon atoms was estimated to be \( 3 \times 10^{16} \text{ cm}^{-3} \).

Equations (12) for the absorption of radiation may be combined to give

\[
N_t = \frac{2 \mu m c}{\pi e^2 f_{t,u}} \left[ \frac{2k}{ma} \ln 2 \right]^{1/2} \frac{T_e^{3/2}}{\lambda}
\]  

(29)
Using \( f_{1, \mu} = 1 \) and \( \mu = 1 \text{ cm}^{-1} \), the equation can be written

\[
N_L = 1.288 \times 10^{13} \frac{T_a^{3/4}}{\lambda}
\]

where \( \lambda \) is in angstroms, \( T_a \) is in \(^0\text{K}\), and \( N_L \) is in cm\(^{-3}\). Using \( T_a = 450^0\text{K} \): at 4200\(\text{Å} \), \( N_L = 6.50 \times 10^{10} \text{ cm}^{-3} \) and at 7724\(\text{Å} \), \( N_L = 3.53 \times 10^{10} \text{ cm}^{-3} \). Since \( N_L \) is the approximate number density required for absorption within 1 cm at 450\(^0\text{K} \), and since the radiation at the longer wavelengths is partially absorbed whereas no absorption occurs at the shorter wavelengths, it is estimated that \( N_M \approx 4 \times 10^{10} \text{ cm}^{-3} \). (Because of the assumptions made here, \( N_M \) could be off an order of magnitude.)

Extrapolation of the curve in Figure 12 to the ordinate where \( E_n = E_M \) gives \( N_M = 1.3 \times 10^9 \text{ cm}^{-3} \) (using \( Z_M = 7.2 \)). Based on arc chamber measurements, the density of metastable atoms there was found to be \( 1.2 \times 10^{11} \text{ cm}^{-3} \); if none of these had decayed to the ground level by the time they reached the point of observation, the density of metastables would be approximately \( 1.2 \times 10^{10} \text{ cm}^{-3} \).

The plasma was checked for optical thinness by comparing the lines from the plasma with those obtained with a concave spherical mirror behind the plasma, the centerline of the plasma being observed at the center of curvature of the mirror. If the plasma is optically thin, then the intensity of the radiation with the mirror in place should be greater than the intensity observed without the mirror by a factor of \( (1 + R) \), \( R \) being the reflectance of the mirror. It was found that all the radiation emitted from 5p-4s transitions was optically thin, but that the infrared radiation from 4p-4s transitions was optically thick to varying degrees. For an idea as to how much self-absorption can affect the radial inversion, a curve is plotted in Figure 13 showing the variation in centerline values of radial intensity versus \( I'(x)/I(x) \),
all being based on curve 19 as the I(x) curve (see Figure 1l). Such a correction has been applied to the infrared data, and it can be seen in Figure 12 that the corrected points are grouped together and fall generally along the line through the other group of data points.

Notice that the number densities of the excited states that have been determined correspond only to transitions from the 4p and 5p levels to the 4s level. The question naturally arises: what about transitions between other levels? Referring to Figure 14, it is observed that the transitions 4d-4p, 5d-4p, 6s-4p, and 7s-4p are within the range of the equipment, but none of these lines were observed. The rest of the transitions in the figure involve radiation outside the range of the equipment. Transition probability information for the 4p-4s and 5p-4s transitions is given in Appendix C; transition probabilities for the 4d-4p and 5d-4p transitions are also available but are not included in this appendix.

Before the rate coefficients plotted in Figure 1 are used in any calculations, the validity of the linear approximation to the function $g_j$ must be shown. Since for the 4p-4s transition, $U = 1.57$ ev., for the 5p-4s transition, $U = 2.95$ ev., and $E_\perp = 4.12$ ev. for both, the linear approximation is valid if

$$0.0412 \leq kT_e \leq \begin{cases} 1.98 & (4p-4s) \\ 2.37 & (5p-4s) \end{cases}$$

which clearly is the case here, since $kT_e = 0.416$ ev.

Regarding the effect of the self-absorption of the infrared radiation on the rate equation: Just as the resonance radiation terms were dropped because the radiation was trapped, the radiative term in the rate equation for the self-absorbed lines were multiplied by a factor $f$ which is essentially the percentage of radiation that
is released from the plasma. This factor is equal to the observed radiation from the plasma divided by the maximum possible radiation, both measurements being made with the spherical mirror in place.

The recombination rate is found from Equation (6a); for purposes of estimating the relative sizes of the terms in the equation, this equation is re-written

$$\alpha = \frac{10}{N_e} \sum_{m=1}^{3} \left( \overline{K_{m,4s}N_m} - \overline{K_{4s,4s}N_{4s}} \right) + \frac{1}{N_e^2} \sum_{m=1}^{3} f(\Sigma_{m,4s}N_m)N_m$$

where only the first three terms of the series are included, $m = 1,2,3...$ correspond to $m = 4p,5p,6p,...$, and the bars indicate averages. From the experimental data and the values computed for the rate coefficients plotted in Figure 1, it can be shown that

$$\overline{K_{6p,4s}N_{6p}} : \overline{K_{5p,4s}N_{5p}} : \overline{K_{4p,4s}N_{4p}} \equiv 0.002 : 0.01 : 1.00$$

and

$$f(\Sigma_{5p,4s}N_{5p}) : f(\Sigma_{4p,4s}N_{4p}) \equiv 0.003 : 1.00$$

The process going on in the gas was imagined to be collisional excitation from the metastable level followed by radiative or two-body de-excitation; this is the process which has been used to explain the afterglow phenomenon in the plasma. The collisional excitation rate was found to be $3.5 \times 10^{-10} \text{ cm}^3/\text{sec}$; the radiative de-excitation rate was $2.70 \times 10^{-10} \text{ cm}^3/\text{sec}$; and the collisional de-excitation rate was $0.60 \times 10^{-10} \text{ cm}^3/\text{sec}$. The recombination coefficient was found to be $0.8 \times 10^{-12} \text{ cm}^3/\text{sec}$, based on the de-excitation rate of the first excited level.

Early investigations of electron-ion recombination carried out by Kentry, Mohler, Boeckner, and Sayers for electron densities of about
$10^{12}$ cm$^{-3}$ and electron temperatures in the range 1000°K - 4000°K gave recombination coefficients for argon of about $2 - 3 \times 10^{-10}$ cm$^3$/sec. More recent studies by Griem for electron densities of about $10^{17}$ cm$^{-3}$ and an electron temperature of about 10,000°K gave apparent recombination coefficients in the range $10^{-11} - 10^{-12}$ cm$^3$/sec. Similar investigations by Olsen and Huxford for $N_e \approx 10^{17}$ cm$^{-3}$ and $T_e \approx 10,000$°K gave recombination coefficients near $10^{-13}$ cm$^3$/sec. A review of these investigations in argon plasmas may be found in Reference 20.
VI. Conclusions

The recombination rate derived in section I was expressed in terms of collisional and radiative transitions between excited levels. It was calculated that the plasma employed in the experiments had collisional and radiative contributions of the same order of magnitude, so that the collisional-radiative approach was truly necessary. The excitation and de-excitation rates were on the order of $10^{-10}$ cm$^3$/sec; the overall recombination rate was about two orders of magnitude faster. De-excitation rates were computed from the approximate equation developed in section I; excellent agreement of this equation with the more exact equation for the case of helium at 0.17 ev. was used as justification of the use of this relation within the range of the linear approximation.

It was shown that the gas in the free jet could be well represented analytically if it was assumed that it consisted of atoms whose ground state was inaccessible and that the temperature associated with the gas was the electron temperature. Thus, the electron density was found from the Saha equation based on the equilibrium reaction: $Ar^M + e \rightleftharpoons Ar^+ + e + e$ at temperature $T_e$; the superscript $M$ refers to the atoms having their "ground" level equal to the metastable level.

The determination of excited state populations calculated from absolute intensity measurements was influenced greatest by errors introduced in the film measurements. These errors resulted from difficulties in keeping the line images focused on the densitometer slit and film calibration procedures. Had photomultiplier tubes together with a strip chart recorder been used for direct readout of the line intensities, much of this error could have been eliminated. Another source of error was the values of transition probabilities
which were generally known within 15 percent.

One further difficulty which may arise in computing excited level populations depends on the value of the electron temperature. The expression for the partition function in section II indicates that for temperatures greater than about 5000°K, the series will diverge so rapidly that this quantity \( Z_M \) cannot be analytically determined.

The presence of the metastable level in argon has been found to account for the afterglow characteristic and the large value of the population of the excited levels of an argon plasma. Reactions in the plasma stream were presumed to be collisional excitation followed by either collisional or radiative de-excitation. The very long lifetime of the metastable level and the closeness in energy of this level to the higher excited levels, together with the fact that the electrons had a sufficiently large amount of energy (nearly 0.5 ev.), is enough evidence for the plausibility of such a process.

In deriving the rate equation, it was noted that the net rate of recombination of the ions was significantly smaller than the rate of excitation and de-excitation. In future work, it is suggested that additional processes be considered. One of these involves the reaction: \( \text{Ar}^+ + \text{Ar}^M \rightarrow \text{Ar}^+ + \text{Ar} + e \), which may occur to an appreciable extent. Such a reaction constitutes a source of electrons in the free jet and could influence the overall recombination rate. By measuring the electron density at two or more points, downstream of each other, one could perhaps discover if the electrons were indeed being produced between the locations. This reaction was disregarded here on the same grounds as the other recombination reactions. Another process that is well worth investigating is that of diffusion. Although the walls of
the test chamber are about three jet diameters away, diffusion to the surrounding gas may be significant.

In the preceding section, the recombination coefficient was determined from the collisional decay of the 4s level. The de-excitation rate coefficient calculated at $T_e = 4380^\circ K$ was used. Although this temperature is probably not very good for this calculation, the result is not expected to be off by more than half an order of magnitude. The excitation rate of the 4s level was assumed to be known more accurately than the collisional de-excitation rate of the 4p level, so the collisional de-excitation rate was calculated from the former value.

Values for the recombination coefficient for argon are scarce; some are shown in the following table:

<table>
<thead>
<tr>
<th>Observer</th>
<th>$N_e$ (cm$^{-3}$)</th>
<th>$T_e$ (°K)</th>
<th>$\alpha$ (cm$^3$/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kenty, et.al.</td>
<td>$\approx 10^{12}$</td>
<td>1000-4000</td>
<td>$2-3 \times 10^{-10}$</td>
</tr>
<tr>
<td>This Author</td>
<td>$\approx 10^{13}$</td>
<td>4380</td>
<td>$0.8 \times 10^{-12}$</td>
</tr>
<tr>
<td>Griem</td>
<td>$\approx 10^{17}$</td>
<td>$\approx 10,000$</td>
<td>$10^{-11}-10^{-12}$</td>
</tr>
<tr>
<td>Olsen and Huxford</td>
<td>$\approx 10^{17}$</td>
<td>$\approx 10,000$</td>
<td>$\approx 10^{-13}$</td>
</tr>
</tbody>
</table>

Because diffusion was neglected in the experiments by Kenty, et.al., the numerical result for the recombination coefficient may not be very meaningful. Since there are no analytical methods for predicting recombination coefficients for complicated gases like argon, it is suggested that the possibility of extending the work of Bates, Kingston, and McWhirter$^3$ be investigated.

Finally, such information as the population of excited levels, electron temperature, electron density, and reaction rates, along with
the velocity, pressure and atom temperature are essential to fluid dynamic studies in a plasma tunnel. Any model study of ablation or heat transfer phenomena, for example, requires a knowledge of the free stream conditions. The techniques employed in this work can be used to obtain such information about the plasma stream.
Acknowledgements

The author would like to express his gratitude to Dr. F.A. Wierum, Jr., of Rice University for his many helpful suggestions during the course of this work.

The author also wishes to express his appreciation to the following agencies under whose support this work has been carried out: National Aeronautics and Space Administration: NASA grant 44-006-033, NASA grant NSG (T) 9, and NASA grant NSG-3-59 and the Shell Companies Foundation, Inc. grant for fundamental research in mechanical engineering.
References


**Additional References**


Figure 1. De-excitation rate coefficients for various transitions in argon and helium. The lower curve for helium indicates the relative importance of transitions between levels whose principal quantum number differs by more than unity.
Figure 2. Energy level diagram for argon showing the lowest levels where the LS coupling designation still applies. Wavelengths are in angstroms; energy values are in cm$^{-1}$. 
Figure 3a. Division of plasma cross section for Abel inversion.

Figure 3b. The observed intensity $I'(x)$ using a plane mirror is compared with $I(x)$ to arrive at an absorption coefficient. Inversion may then be accomplished considering absorption.
Figure 4. Schematic representation of the plasma jet wind tunnel, the spectrograph, and the associated optical system.
Figure 5. Test Control, Recording, and Instrumentation Consoles.

Figure 6. View of the Plasma Dynamics Laboratory.
Figure 7. Argon free jet; scale superimposed for size comparison.

Figure 8. Argon free jet with impact probe in the flow.
Figure 10. Spectrograph table with built in radiation standard facility. Densitometer located behind spectrograph.
Figure 11. Curve 19 (Ref. 19) for the observed intensity distribution, $I(x)$, and the inverted distribution, $I(r)$. 

$k = 0.81$

$\xi = 1.00$
Figure 12. Number densities of the excited states of argon divided by the degeneracies versus the energies of the states. The data were taken one inch downstream of the sonic orifice. The plotted points correspond to values on the axis of symmetry.
Figure 13. Variation of the correction factor \( \xi \) for self-absorption with the proportion of released radiation from the plasma based on intensity curves having the shape of curve 19 (in Reference 19, Figure 3). \( I'(x) \) is the observed intensity using a mirror behind the plasma having a reflectance of 0.9.
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Figure 14. Wavelengths $\lambda_n$ of possible transitions based on the average energy $E_n$ for several subshells. The values of $\lambda_n$ are in angstroms. Transition probability data for the 4p-4s and 5p-4s transitions are given in Appendix C. Data for the 4d-4p and 5d-4p transitions may be found in Reference 7.
Figure 15. Spectrogram taken of plasma jet using Kodak type 1-N film which had been hypersensitized. This halation effect was caused by partial removal of an antihalation backing during the hypersensitizing process.
Figure 16. Simplified sketch of optical path denoting quantities necessary for making the conversion from volume radiance to spectral steradiancy.
Appendix A. Nomenclature

\( a_{n,p} \) area coefficients in Abel inversion

\( A_{k,k} \) shape factors tabulated by Pearce

\( A_{n,m} \) probability of spontaneous emission from level \( n \) to level \( m \)

\( c \) speed of light \((3.00 \times 10^{10} \text{ cm/sec})\)

\( e \) electronic charge \((4.80 \times 10^{-10} \text{ esu})\)

\( E_n \) energy of level \( n \)

\( f_{t,u} \) oscillator strength from level \( t \) to level \( u \)

\( g_n \) degeneracy of level \( n \)

\( h \) Planck's constant \((6.63 \times 10^{-27} \text{ erg-sec})\)

\( I_{n,m} \) intensity of radiation due to transition from level \( n \) to level \( m \)

\( I(r) \) radial intensity distribution

\( I(x) \) observed intensity distribution

\( k \) Boltzmann's constant \((1.381 \times 10^{-16} \text{ erg} / \text{K-molecule} = 8.617 \times 10^{-5} \text{ ev} / \text{K-molecule})\)

\( K_{n,m} \) collisional rate coefficient

\( m_a \) mass of argon atom \((6.62 \times 10^{-23} \text{ g})\)

\( m_e \) mass of electron \((9.11 \times 10^{-28} \text{ g})\)

\( N_e \) number density of electrons

\( N_n \) number density of atoms having an electron in an excited level \( n \)

\( N_3 \) number density of argon atoms in ground state

\( Q_{n,m} \) excitation cross section

\( S_{k,k} \) shape factor in Appendix D

\( T_e \) electron temperature

\( U \) excitation energy \(= E_n - E_m \) (\( n > m \))

\( v \) velocity

\( Z_3 \) argon atom partition function

\( Z_M \) metastable atom partition function
\[ \alpha \] recombination coefficient
\[ \lambda \] wavelength
\[ \mu, \mu_k \] absorption coefficient
\[ \nu \] frequency

**Subscripts**

- \[ a \] atom
- \[ e \] electron
- \[ g \] ground level
- \[ M \] metastable level
Appendix B. Argon I Energy Level Diagram

Energy values are given in cm\(^{-1}\); wavelengths are given in angstroms. The levels marked "M" are the metastable levels. The level designations are the same as used in Adcock and Plumtree.\(^7\)
Appendix C. **Argon I Transition Probability Data** for a transition from an upper level \( n \) to a lower level \( m \). Values taken from Adcock and Plumtree.\(^7\)

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<th>( E_n ) (cm(^{-1}))</th>
<th>( E_n ) (ev)</th>
<th>( g_n )</th>
<th>( g_m )</th>
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<th>Estimated Error (% of)</th>
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Appendix D. Radial Inversion Techniques

1. Fortran I Computer Program for Numerical Inversion

Symbols:

- ITOT - number of radial divisions used
- WLINE - name and wavelength of spectral line
- N1, N2, N3 - date
- N4 - film number
- A(M) - area coefficients $a_{p,n}$
- X(K) - $I(x)$
- Y(K) - $I'(x)$
- R(K) - $I(r)$
- Z(K) - $\mu_k$

The first 65 data cards contain the table of area coefficients given by Pearce, and are read in as a column with A(1) corresponding to $a_{1,1}$ and A(325) corresponding to $a_{25,25}$. The table is read in by rows from left to right beginning with the top row. As this data table is read in, the sequence of the cards is checked.

This program may be used for radial inversion for an optically thin plasma or for a plasma exhibiting some self-absorption. Sense switch 1 in the program is used to select one of these two cases.
C SOLUTION OF ABEL INTEGRAL WITH OR WITHOUT ABSORPTION
C SENSE SWITCH 1 ON WITH ABSORPTION
C MAX. VALUE OF ITOT = 25
DIMENSIONA(325),R(25),X(25),Y(25),Z(25)
K = 1
20 DO1M=1,325,5
READ 2,A(M),A(M+1),A(M+2),A(M+3),A(M+4),L
2 FORMAT(5F12.2,120)
 IF(L-K) 12,1,12
12 PRINT 14
14 FORMAT(2OHCARD OUT OF SEQUENCE)
13 PAUSE
GO TO 20
1 K = K + 1
 RR = .9
3 READ 4, WLINE,ITOT,N1,N2,N3,N4
4 FORMAT(17HELEMENT AND LEVEL,F10.2,2Ha,A,4I2,13)
 IF(ITOT - 25)11,11,3
11 DO5K=1,ITOT,5
5 READ 2,X(K),X(K+1),X(K+2),X(K+3),X(K+4)
 IF(SENSE SWITCH 1)22,23
22 DO21K=1,ITOT,5
21 READ2,Y(K),Y(K+1),Y(K+2),Y(K+3),Y(K+4)
23 PUNCH 19,N1,N2,N3
19 FORMAT(//12HMILTON FROST,4X13,2H/13,2H/13/)
 IF(SENSE SWITCH 1)24,25
24 PUNCH 16
16 FORMAT('3OHABEL INVERSION WITH ABSORPTION')
GO TO 26
25 PUNCH 9
9 FORMAT('29H SOLU TION OF ABEL INTEGRAL FOR')
26 PUNCH 4, WLINE
PUNCH 50, N4
50 FORMAT('21X4HFILM,14')
IF(SENSE SWITCH 1)47, 27
47 PUNCH 6
6 FORMAT('/3H K, 8*4H1(X), 7*5H1(T(X), 8*4H1(R), 10X2HMU/')
GO TO 28
27 PUNCH 29
29 FORMAT('/3H K, 8*4H1(X), 8*4H1(R)/')
GO TO 61
28 DO10 I = 1, ITOT
K = ITOT - 1 + 1
IF(X(K))13, 40, 41
40 Z(K) = 0.
GO TO 10
41 L = K + 1
IF(L-1) 17, 17, 7
17 M = K*(K+1)/2
Z(K) = (1.-SQRT((Y(K)-X(K))/RR/X(K))) / A(M)
GO TO 10
7 PROD = 1.
DO8 J = L, ITOT
M = K+J*(J-1)/2
\[ \text{PROD} = \text{PROD} \times (1 - Z(J) \times A(M)) \]

8 CONTINUE

\[ M = K \times (K+1)/2 \]

\[ Z(K) = (1 - \text{SQRT}((Y(K) - X(K))/RR \times X(K))/\text{PROD})/A(M) \]

10 CONTINUE

\[ \text{DO361}=1,1 \text{TOT} \]

\[ \text{SUM} = 0. \]

\[ P1 = 1. \]

\[ J = \text{TOT} - 1 + 1 \]

\[ \text{IF}(I-1)30,30,31 \]

30 \[ M = J \times (J+1)/2 \]

\[ R(\text{TOT}) = X(\text{TOT})/(2 - Z(\text{TOT}) \times A(M))/A(M) \]

\[ \text{GO TO 36} \]

31 \[ N = J + 1 \]

\[ \text{DO3211}=N,1 \text{TOT} \]

\[ KL = N - 1 \]

\[ KK = N + 1 \]

\[ \text{IF}(KK-\text{TOT})34,34,35 \]

35 \[ P1 = 1. \]

\[ \text{GO TO 37} \]

34 \[ \text{DO33111} = KK,1 \text{TOT} \]

\[ M = J + \text{III} \times (\text{III} - 1)/2 \]

33 \[ P1 = P1 \times (1 - Z(\text{III}) \times A(M)) \]

37 \[ P2 = 1. \]

\[ \text{DO38N1}=J,\text{KL} \]

\[ M = J+N1 \times (N1-1)/2 \]

38 \[ P2 = P2 \times (1 - Z(N1) \times A(M))**2. \]
M = J + II*(II-1)/2
32 SUM = SUM+R(II)*A(M)*P1*(1.+1.-Z(II)*A(M))*P2
P3 = 1.
N = J + 1
DO3911 = N,1,ITOT
M = J + II*(II-1)/2
39 P3 = P3*(1.-Z(II)*A(M))
M = J*(J+1)/2
D = A(M)*P3*(2.-Z(J)*A(M))
R(J) = (X(J)-SUM)/D
36 CONTINUE
DO18K=1,1,ITOT
18 PUNCH15,K,X(K),Y(K),R(K),Z(K)
15 FORMAT(13,3F12.2,E16.7)
GO TO 3
61 DO701=1,1,ITOT
SUM = 0.
K = ITOT - 1 + 1
77 IF(I-1)77,77,67
M = K*(K+1)/2
GO TO 69
67 L = K + 1
DO68J=L,1,ITOT
M = K+J*(J-1)/2
68 SUM = SUM+R(J)*A(M)
69 M = K*(K+1)/2
70 R(K) = (X(K)-SUM)/A(M)
DO 45 K = 1, TOT

R(K) = R(K)/2.

45 PUNCH 15,K,X(K),R(K)

GO TO 3

END
2. Radial Inversion Using Table of Shape Factors

This method simply involves fitting the experimentally determined curve for \( I(x) \) to a similarly shaped curve in Figure 3 of Reference 19. The number of the curve best fitting the data is used in Table 1 (Ref. 19) to find the shape factors \( A_{k,k} \). Since \( I(x) \) on the axis was adjusted to a value of 100, the radial intensity distribution \( I(r) = C A_{k,k}/a_{k,k} \) where \( a_{k,k} \) are the area coefficients found in Pearce's Table 14. The "C" is a constant adjusted so that the tails of the \( I(x) \) and \( I(r) \) curves coincide. Values for a new shape factor \( S_{k,k} = A_{k,k}/a_{k,k} \) are tabulated below. The quantity \( S_{k,k} \) is more useful because the values corresponding to a particular \( I(x) \) curve are now measures of relative radial intensity. A plot of the quantity \( S_{k,k} \) versus \( k \) is, therefore, a plot of \( I(r)/C \). The shape factor \( S_{k,k} \) may also be used in the same way as \( A_{k,k} \) in the determination of temperature by the two-line method discussed in Reference 19.
<table>
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Appendix E. **Processing of Kodak Type 1-N Spectroscopic Film**

Type 1-N film should be developed in Kodak Developer D-19 (for good contrast with high effective speed and low fogging tendency) for about 3 minutes at 75°F with continuous agitation. Development is quickly stopped by immersing the film in Kodak Stop Bath SB-5 for at least 30 seconds with continuous agitation. An antihalation backing on this film is removed during this process. The film is then fixed in Kodak Rapid Fixer for about 3 minutes with frequent agitation, washed for 30 minutes in running water, sponged off with a film squeegee to remove any of the remaining backing, treated with Kodak Photo-Flo Solution, and hung to dry.

**Hypersensitizing**

Hypersensitizing is a process of treating a film to increase its spectral sensitivity. This process is not recommended for this film, as experiments performed by the author showed that the antihalation backing on the film is partially removed during this process, and, as a result, there is halation in the high intensity image areas caused by internal reflections. This effect is clearly seen in the photograph in Figure 15.
Appendix F. Procedure for Establishing the Absolute Intensity Scale

The standard of spectral radiance is calibrated in terms of radiated power per steradian per millimicron per square millimeter of filament. The relation for the intensity of the plasma given by Equation (14) gives a volume radiance in terms of radiated power per unit volume. A relationship between these two quantities must be established. In order to do this, the latter quantity is converted to a spectral radiance by multiplying by an average depth $d$, dividing by the solid angle $\omega$ subtended by the limiting optic, and dividing by the wavelength interval $\delta x$. Referring to Figure 16, it is seen that the solid angle is given by $A/D^2$, where $A$ is the area of the limiting optic; the wavelength interval is given by the projection of the slit width on the film and is equal to the product of the dispersion of the grating and the slit width.

Now that the units are the same, it only remains to adjust the spectral radiance of the standard by a factor, $r$, which accounts for the difference in exposure times for the plasma ($t_p$) and the standard ($t_s$), for the difference between the observed intensity and the true intensity (a factor $\kappa$), for self-absorption (a factor $\xi$), and for the arbitrary nature of the emulsion calibration scales. When this is done, it is found that

$$h\nu_{n,m} A \frac{N}{d} \frac{D^2}{A} \delta x = W_\lambda \cdot r$$

where

$$r = \frac{t_p}{t_s} \frac{I(x)}{I_s} \kappa \xi$$

and $W_\lambda$ is the spectral radiance of the standard, $I(x)$ is the intensity of the plasma radiation read from the characteristic curve, and $I_s$ is
the intensity of the standard read from the same curve.

From the above information, the number density of an excited level \( n \) divided by the degeneracy of the level is

\[
\frac{N_n}{g_n} = \left\{ \frac{\mu^2 \delta x}{\hbar c} \frac{\tau_p A}{\tau_s D^2 I_s} \right\} I(x) \left[ \frac{\Lambda_{n,m}}{\Lambda_{n,m,g}} \right]
\]

Alternate, but identical, optical paths were used for the radiation measurements; for different optical paths, additional geometrical corrections become necessary, reflectances of the various optics may have to be considered, and even the presence of water vapor in the air may have to be accounted for.