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Spectroscopic Study of Electron Recombination with Monatomic Ions in a Helium Plasma

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Recent work on electron-ion recombination has 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 in which processes involving molecular ions, etc., are unimportant. By making absolute intensity measurements of the lines of helium emitted from the decaying plasma of an arc jet in the visible and near ultraviolet, the number densities of the excited states of helium have been calculated. From these measurements electron temperatures between 0.15 and 0.30 eV and electron densities between 4×10^{12} and 4×10^{14} cm^{-3} are found, with a fractional ionization of the order of 1%. When interpreted by means of the collisional-radiative recombination model, these measurements also give values for $K_{n,n-1}$, the collisional de-excitation rate constant of an excited helium atom from level n to $n-1$. The constancy of $K_{n,n-1}$ over a range of electron density from 4×10^{12} to 4×10^{14} per cm^3 indicates the validity of the model, and the values of $K_{n,n-1}$ are in reasonable agreement with a recent theory of classical excitation cross sections. A square-root dependence of this cross section on the excess energy is indicated, since the values of $K_{n,n-1}$ appear to be independent of electron temperature. It is then found that $Q_{2,3}=9.3\times\sqrt{E}$, $Q_{3,4}=73\times\sqrt{E}$, and $Q_{4,5}=380\times\sqrt{E}$, where $Q_{n,n+1}$ is the averaged excitation cross section from the levels with principal quantum number n to the levels with principal quantum number $n+1$ in units 10^{-15} cm^2, and E is the excess energy in eV. These cross sections have an estimated error of about 30% in the threshold energy range to about 0.3 eV.

INTRODUCTION

THE recently developed model of electron-ion recombination, including recombination into highly excited bound levels through electron-electron-ion collisions and transitions between the bound levels through electron-atom collisions, predicts recombination rates several orders of magnitude larger than pure radiative recombination for the conditions of many laboratory experiments.[1–4] For plasmas in the rare gases where a reasonable fraction (0.1% or more) of the atoms are ionized, volume recombination of this collisional-radiative type may be expected to dominate. There is evidence that it may also be important for some of the older experiments at electron densities greater than 10^{12} cm^{-3}, where analysis based on a process involving the formation of molecular ions and subsequent dissociative recombination has not met with too much success.[3] Experiments on the recombination of helium in the magnetically confined, highly ionized plasma of a "Stellarator," where both microwave and detailed spectroscopic measurements were made, have provided good confirmation of the collisional-radiative recombination model.[2] In these experiments it was found that classical expressions for the ionization and excitation cross sections of an atom by an electron resulted in predicted values of the recombination rate, and of the number densities of the excited states of helium, in fair agreement with the experimentally measured values.

In the present experiments a steady state, recombining plasma was created by expanding a stream of arc

[1] R. G. Giovanelli, Australian J. Sci. Res. **A1**, 274, 289 (1948); N. D'Angelo, Phys. Rev. **121**, 505 (1961); L. M. Biberman, Yu. N. Toropkin, and K. N. Ul'yanov, Zh. Tekhn. Fiz. **32**, 827 (1962) [translation: Soviet Phys.—Tech. Phys. **7**, 605 (1963)].
[2] E. Hinnov and J. G. Hirschberg, Phys. Rev. **125**, 795 (1962).
[3] S. Byron, R. C. Stabler, and P. I. Bortz, Phys. Rev. Letters **8**, 376 (1962).
[4] D. R. Bates, A. E. Kingston, and R. W. P. McWhirter, Proc. Roy. Soc. (London) **A267**, 297 (1962); **A270**, 155 (1962).

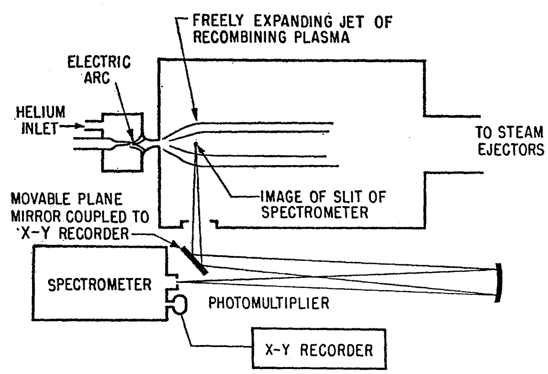

FIG. 1. A schematic outline of the plasma jet wind tunnel and the spectrometer optics.

heated helium in a large, low-density wind tunnel. Detailed spectroscopic study of the plasma was facilitated because the conditions of the source remained constant over long periods of time. It was not possible, however, to measure the decay rate of the plasma directly, and all the results are based on measurements of the absolute intensities of the various radiative transitions of the helium atom. In order to treat the rate of transitions between excited states induced by electron collisions, all of the states with the same principal quantum number are averaged together. The classical theory of excitation cross sections predicts that by far the greatest rate will occur between states with principal quantum numbers differing by unity, and using this information we are able to infer the collisionally induced transition rates from the experiment. Further, the values of the cross sections may be inferred from the temperature dependence of the transition rates. The transition rates (and cross sections) are found to be in reasonable agreement with rates calculated using recently derived classical cross sections,[5] in which the motion of the orbital electron is taken into account. Detailed calculations of recombination rates using these cross sections have been given in Ref. 4, and some general simplifications in the theory are discussed in Refs. 2 and 3.

EXPERIMENTAL EQUIPMENT

A schematic outline of the plasma jet wind tunnel[6] and the spectrograph is shown in Fig. 1. A $\frac{1}{4}$-in. tungsten cathode was placed about two inches axially from a copper nozzle which served as the anode. The nozzle had a throat diameter of 0.44 in., and all parts were water cooled. With a helium flow rate of 0.05 g moles/sec and an arc current of 500 A, the pressure in the arc chamber was about 90 mm Hg and the arc voltage was about 33 V. In most of the experiments a free jet expansion of the helium plasma was used, and the nozzle ended at the sonic throat. The main body of the wind tunnel, which contained a traverse mechanism for positioning various probes in the flow and a quartz window for the spectrograph, was maintained at about 0.25 mm Hg by a steam ejector pumping system.

The problem of a free jet expansion in an inviscid fluid has been analyzed,[7] and the solution indicates a rapid acceleration of the flow to a high Mach number, followed by a normal shock wave. The lower the pressure in the chamber in which the expansion occurs, the higher the Mach number reached before the shock wave. Unfortunately, under the conditions for stable operation of the arc, the Reynolds number of the free jet was quite low, so that the viscous effects in the jet expansion were large. Isentropic flow theory could not then be applied to determine the Mach number and other flow characteristics, a fact that was borne out by a comparison of the axial variation of the spectroscopically measured electron densities with the gas densities deduced from measured impact pressures and isentropic flow theory. The spectroscopic measurements indicated that the free jet Mach number never reached a value higher than 3, although the impact pressure measurements when interpreted by isentropic flow theory yielded Mach numbers in excess of 10.

It was not possible to run at higher flow rates to reduce these viscosity effects, as the arc voltage would break into oscillations of as much as 20-V amplitude at a frequency of a few kilocycles, accompanied by similar oscillations in the luminosity of the plasma. At low flow rates all oscillations (up to about 100 Mc/sec) were either absent or small.

The temperature of the helium at the throat of the orifice can be estimated from the ratio of arc chamber pressure with and without arc operation. It can be shown that the mass flow through a sonic throat is proportional to $p_0/T_0^{\frac{1}{2}}$, where p_0 and T_0 are the stagnation pressure and temperature, and a comparison of the measured stagnation pressures with and without arc operation gave a mean stagnation temperature of about 7000°K. The temperature, density, and pressure in the free jet expansion drop rapidly, although not as rapidly as predicted by isentropic flow theory, until the shock wave is reached, about 4 in. downstream. After the shock wave, which is quite diffuse, the plasma had a diameter of about 6 in. and appeared quite uniform to the end of the tunnel, four feet farther downstream. When the arc was operating "well," this plasma was remarkably stationary and symmetric, giving the appearance of a large glow discharge tube. At other times the jet was less symmetric, as the arc showed a tendency to attach itself to one side of the anode, and considerable flickering could be noticed in the boundaries of the jet.

[5] M. Gryzinski, Phys. Rev. **115**, 374 (1959).
[6] L. Talbot, J. E. Katz, and C. L. Brundin, Phys. Fluids **6**, 559 (1963); University of California Eng. Proj. Rept. HE-150-181, 1960 (unpublished).
[7] K. Bier and B. Schmidt, Z. Angew. Phys. **13**, 493 (1961); P. L. Owen and C. K. Thornhill, Aeronautical Research Council Reports and Memo 2616, Great Britain, 1952 (unpublished).

The spectrograph used was a Jarrell-Ash 3.4-m Ebert-type, which can take photographic spectra, or as was the case in these experiments, can be fitted with an exit slit and a photomultiplier and be operated as a direct recording instrument. Wavelength coverage was obtained by rotation of the grating, with readout on a counter directly in angstroms. An image of the slit of the spectrograph was focused on the plasma column with a spherical mirror and a plane mirror as shown in Fig. 1, resulting in the detection of the radiation emitted by a cross section of the plasma column about 3.5 mm high and 0.5 mm wide (in the direction of flow). The plane mirror swiveled on ball bearings about two axes with calibrated micrometer drives, so that any region of the plasma within about eight inches of the orifice could be observed. This plane mirror also rotated sufficiently so that various discharge tubes and radiation standards could be focused on the spectrograph.

SPECTRUM

A survey of the radiation from the helium plasma on photographic plates revealed only neutral helium atoms, with the various series such as $2\,^3P-n\,^3D$ merging into a continuum at about $n=12$ for the region of the jet close to the orifice. Farther downstream, these series disappeared at about $n=18$ and there was no observable continuum. With long exposure (10^4 times that necessary to observe the stronger lines of helium), N_2 and N_2^+ molecular bands were observed, principally in the boundary of the jet, and hydrogen and argon lines were observed more uniformly across the plasma. The nitrogen bands were probably due to air leaks in the tunnel, while the hydrogen and argon lines indicated that the gas purity was better than 100 ppm. In particular, no tungsten or copper from the arc was found.

The argon impurity lines in helium show a peculiarity. When the arc is run on pure argon, only the lines belonging to argon I, the neutral atom (resulting from recombination of electrons and argon ions into excited states of neutral argon) are found. This is presumably due, as in helium, to the fact that the doubly ionized species recombine very rapidly, before the plasma reaches the point of observation. However, the argon impurity lines in helium are all of argon II (singly ionized), with no argon I observable. It seems as if some process must very rapidly excite the ground-state argon ions before they have a chance to recombine. One possibility is that collisions of the second kind with helium atoms in the $n=2$ levels may excite the ground-state argon ions. Because of resonance radiation trapping, all of the $n=2$ levels of helium will have fairly high densities, although considerably less than the electron density (because of collisional de-excitation). Since none of the observed argon ion lines had an excitation energy greater than 21.2 V, the highest energy of the $n=2$ helium levels, this process would seem to be indicated. In order to suppress the argon I radiation, it is necessary that the rate of excitation of the argon ions be considerably greater than their rate of recombination. Upon estimating the recombination rate (from the theory presented in this paper) and assuming the resonance radiation trapping to be complete so that the density of helium atoms in the $n=2$ levels can be calculated assuming collisional de-excitation, it is found that the cross section for excitation of an argon ion in a collision with an excited helium atom must be somewhat greater than 10^{-14} cm^2. In view of such a large cross section, this explanation must be regarded as only tentative.

MEASUREMENTS

The measurement of the radiation intensity from a region of plasma in the jet cannot be made directly, since observations on the center line of the plasma column, for instance, include contributions from all regions out to the boundary. If the jet is assumed to be cylindrically symmetric, the radiation reaching the spectrograph is given by the Abel integral equation, whose inversion is well known. This inversion requires the complete profile of the intensity of each line across the jet. To obtain this information the intensity of each helium line was recorded as the movable plane mirror, driven by a motor, performed a transverse scan of the jet. A Moseley x–y recorder, with x axis coupled to the mirror, was used to record this scan. The exit slit of the spectrometer was set at 0.71 mm and the entrance slit at 0.13 mm, so that quite uniform response was achieved over a wavelength range of 3.4 Å in order to measure all of the radiation belonging to a spectral line. The spectral response was determined by scanning a singlet helium line from a discharge tube. (It might be noted that adjustment of the RCA–1P28 photomultiplier tube behind the exit slit was required to find a region of the photocathode with uniform response.)

The intensity calibration of the spectrograph-photomultiplier system was made with a tungsten ribbon filament lamp, viewed with the same optical arrangement as the plasma except for the quartz window in the tunnel. The primary standard, made by General Electric (#30 A/T24-3), and equipped with a quartz window, was supplied by the Bureau of Standards and calibrated for spectral intensity from 2500 to 7500 Å, when run at 35.0 A ac. This lamp was used only occasionally, and another identical lamp calibrated against the primary standard was used for normal operation. The measurement of current had to be quite precise, as a change of only 0.3% in current gave a noticeable change in intensity. The ammeter was periodically checked against a laboratory standard accurate to 0.1%. Scattered light from the internal parts of the spectrograph necessitated the use of a uv transmitting, visible absorbing filter for calibration below 3400 Å. With this filter, Corning 7–54, the calibration was extended to 2500 Å, with about 65% scattered light at 2500 Å. A red transmitting filter, Corning 2–73, was used to

TABLE I. Helium oscillator strengths. (All values of f_{lu} in units of 10^{-2}.)

Principal quantum number	$2\,^3P-n\,^3D$	$2\,^1P-n\,^1D$	$2\,^3P-n\,^3S$	$2\,^1P-n\,^1S$	$2\,^3S-n\,^3P$	$2\,^1S-n\,^1P$
3	62.3	72.5	5.71	16.5	7.64	4.64
4	12.3	12.1	2.08	5.08	1.16	0.824
5	4.67	4.31	1.19	2.29	0.385	0.300
6	2.33	2.05	0.709	1.26	0.190	0.151
7	1.32	1.15	0.414	0.720	0.114	0.087
8	0.849	0.740	0.268	0.465	0.071	0.055
9	0.580	0.490	0.186	0.311	0.048	0.037
10	0.408	0.351	0.131	0.223	0.034	0.026
11	0.300	0.280	0.099	0.168	0.025	
12	0.225	0.195	0.077	0.129	0.018	
13	0.175	0.152	0.060	0.103	0.014	
14	0.140	0.120	0.049	0.083		
15	0.112	0.098	0.039	0.069		

eliminate the second-order radiation for wavelengths larger than 6000 Å. The whole system had less than 2% variation in sensitivity over periods of several days. The photomultiplier was powered by batteries and left on continuously, and only currents less than 10^{-6} A were drawn from it to minimize fatigue effects.

A complete set of measurements of all observable lines of helium required about $1\frac{1}{2}$ h, and during this time the plasma jet was generally quite steady. The intensity of any given line of helium varied by less than 5% if the arc current was maintained constant, while the arc voltage would typically drop about 10%. The largest errors in the final results probably arise from the fact that small errors in measurement, or in lack of symmetry of the jet, have a large effect on the Abel integral inversion.

DATA REDUCTION

Numerical techniques for solving the inverted Abel equation giving the intensity of radiation $I(r)$ (in, for example, watts/cm^3) at a point in the cylindrically symmetric jet from the experimentally determined values of $J(x)$ (in watts/cm^2) have been described in the literature.[8–10] The accuracy of these various techniques was tested, and the method using a cubic interpolating polynomial passed through the experimental points[9] was found to be the best for our data. (A more complete discussion of several experimental details is given in Ref. 11.) It was found that a relative error of 1% in the first two or three values of $J(x)$ near the center line would result in about 5% error in $I(r)$ on the center line. Since the experimental curves were at best of this order of symmetry, a 5% error is about the best obtainable. Only values on the centerline were used in the results to be presented, as the errors were even higher off the center line.

Because the sensitivity of the spectrometer was made uniform over a range of 3.4 Å, the photomultiplier current was proportional to the total intensity of the helium line selected if the width of the line was much less than 3.4 Å. This was satisfied for all but those lines originating from very highly excited states, where the Stark broadening is appreciable. In this latter case integration over a spectral scan of the line was sometimes used to obtain the intensity; however, the over-all accuracy for these lines was considerably lower than for the rest of the measurements.

The light intensity indicated by the photomultiplier current was put on an absolute scale by calibration with the standard lamp. The number density of atoms can then be found if the oscillator strength for the line is known. The theoretical values of the oscillator strengths found by the Coulomb expansion method of Bates and Damgaard[12] and the variational technique of Trefftz *et al.*[13] agreed quite closely, and were used as the basis of our values. The tables of Bates and Damgaard give values for all six series of lines in the visible up to $n=5$ or 6, and Trefftz *et al.* calculated a number of the astrophysically important oscillator strengths, with some values at $n=10$. These values were extrapolated graphically to give all oscillator strengths up to $n=15$ by using the hydrogen oscillator strengths for the same series as a guide, and the resulting values used in this work are given in Table I.

RESULTS AND DISCUSSION

Measurements were made on all the lines of helium that were intense enough to be observed. The center line values of N_u/g_u were plotted on a logarithmic scale versus the energy level, and a typical result is shown in Fig. 2. These graphs are similar to those obtained by Hinnov and Hirschberg,[2] and, as was the case with their results, all the states of higher excitation seem to fall on

[8] O. H. Nestor and H. N. Olsen, Soc. Industr. Appl. Math. Rev. **2**, 200 (1960).
[9] K. Bockasten, J. Opt. Soc. Am. **51**, 943 (1961).
[10] M. P. Freeman and S. Katz, J. Opt. Soc. Am. **50**, 826 (1960).
[11] F. Robben, University of California Eng. Proj. Report HE-150-211, 1963 (unpublished).
[12] D. R. Bates and A. Damgaard, Phil. Trans. Roy. Soc. (London) **A242**, 101 (1949).
[13] E. Trefftz, A. Schlüter, K. Dettmar, and K. Jorgens, Z. Astrophys. **44**, 1 (1957).

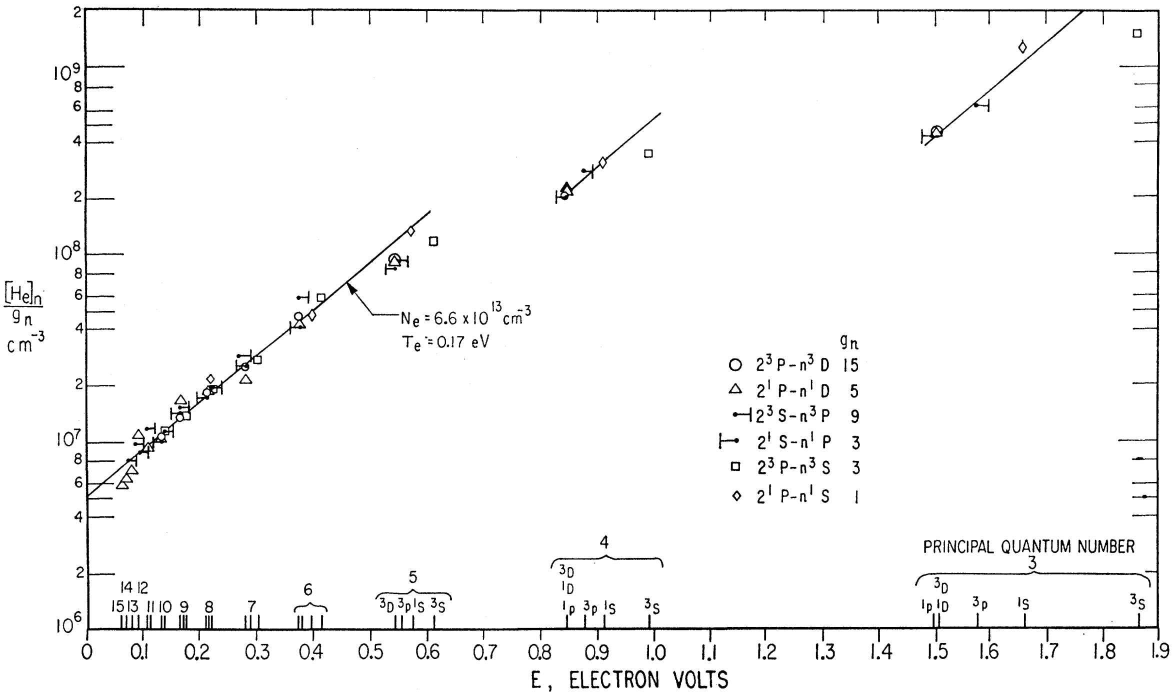

FIG. 2. The density of the excited states of helium, in cm^{-3} divided by the multiplicity of the state, versus the binding energy of the state in eV. The data were taken 2.8 cm downstream of the sonic orifice.

a straight line, indicating a Boltzmann distribution whose temperature is given by the slope. The Boltzmann distribution arises from the fact that transitions between the levels induced by electron collisions are much more rapid than the net rate of recombination, a result leading to an estimate of the cross section in agreement with the results to be presented later.

For the lower levels, even though they deviate from a Boltzmann distribution, it appears that all levels with the same principal quantum number are approximately maintained with relative populations given by a temperature equal to that found from the higher levels. The $3\,{}^3S$ and $4\,{}^3S$ levels seem to deviate the most from this relationship, but an error in the oscillator strengths for these levels could change the results. It seems, then, that collisionally induced transitions between the triplet and singlet states belonging to the same quantum numbers must be fairly frequent, compared to the rate at which transitions occur from other levels. The general over-all agreement of results from the different series for the higher levels indicates that the oscillator strengths used in the data reduction are accurate to at least about 10%, and that the measurements themselves are accurate to about 10%.

The high quantum number levels may be assumed to have rapid transitions induced by electron collisions with levels lying above the ionization limit, that is, with the free electrons. Thus, the whole system of upper levels is in equilibrium with the free electrons, and the Saha equation,

$$\frac{N_e N_i}{N_u}=\frac{g_i g_e}{g_u}\left(\frac{2\pi m_e kT}{h^3}\right)^{3/2} e^{-E_u/kT}, \qquad (1)$$

can be used to give the density of the free electrons. Upon setting $N_e=N_i$ and inserting numbers, Eq. (1) becomes

$$N_e=(1.2\times10^{22}T^{3/2}N_\infty/g_\infty)^{1/2},$$

where N_e is in electrons/cm^3, T is in eV, and

$$N_\infty/g_\infty=N_u/g_u e^{-E_u/kT}.$$

The quantity N_∞/g_∞ is found graphically by the extrapolation of the upper levels in Fig. 2 with a straight line to the $E=0$ axis. The electron density and temperature obtained are given on the figure.

Verification of the above assumptions can be taken from the work of Hinnov and Hirschberg[2] and from the internal consistency of the results to be presented here, but two independent determinations of the electron density were also attempted. First, about 1% of hydrogen was added to the helium and the Stark broadening of the H_0 line was used. The spectral half-width of this line was measured, and after corrections for the instrumental and Doppler broadening the electron density was inferred using published tables of the Stark broadening of the hydrogen lines. Because of the low electron densities and resultant small Stark broadening, the agreement with the values of electron density from the Saha equation was only within an order of magnitude. Secondly, the Inglis-Teller relation[14]

$$N_e = 0.014 a_0^{-3} n^{-7.5},$$

where a_0 is the Bohr radius, 0.53×10^{-8} cm, and n is the quantum number of the highest observable discrete level before the lines merge into a continuum, was used. This formula was derived for the Stark broadening of the highly excited states of hydrogen, but can also be expected to apply to helium. Quite good agreement was obtained with the values of electron density from the Saha equation and from the Inglis-Teller relation.

The analysis of the recombination process follows the same model as employed in Refs. 2, 3, and 4. A rate equation is written down for the number density N_n of atoms with principal quantum number n, taking into account both radiative processes and collisional processes involving electrons.

These equations are given by

$$\frac{dN_n}{dt} = \sum_{m=n+1}^{\infty} N_m A_{m,n} - N_n \sum_{m=1}^{n-1} A_{n,m} + N_e \left\{ \sum_{m=n+1}^{\infty} (N_m K_{m,n} - N_n K_{n,m}) - \sum_{m=1}^{n-1} (N_n K_{n,m} - N_m K_{m,n}) \right\}, \quad (2)$$

where $K_{m,n}$ is the collisional rate constant for the transition from level m to n. The absorption of radiation is not explicitly included, but will later be taken into account by dropping all radiative transitions to the ground level, and the sums over the discrete levels can be generalized to include the continuum. For the plasmas considered here, N_e is much larger than any N_n except N_1, the ground level, and the parameters of the plasma such as temperature and density change slowly enough in time so that all the dN_n/dt except dN_1/dt in the complete set of Eqs. (2) can be set equal to zero. This approximation is discussed in greater detail in Ref. 4. By then making some reasonable approximations regarding the nature of $K_{m,n}$ and using the experimental measurements as summarized in Fig. 2, values of $K_{m,m-1}$ can be obtained for those levels which are important in determining the recombination rate. This method depends on knowledge of the Einstein coefficients, and knowledge of the approximate behavior of $K_{m,n}$.

Recently Gryzinski[5] has calculated cross sections for excitation of atomic levels by electron impact from classical considerations that appear to be in fair accord with experiment. The excitation rate can be written in terms of the cross section as

$$N_e K_{n,m} = \int_{E_{\mathrm{ex}}}^{\infty} \frac{dN_e(E)}{dE} v(E) Q_{n,m}(E) dE,$$

where E_{ex} is the excitation energy of the level, $dN_e(E)/dE$ is the electron energy distribution, $v(E)$ is the electron velocity, and $Q_{n,m}(E)$ is the excitation cross section. The principle of detailed balance, which states that at equilibrium the forward and reverse rates of all possible reactions are equal, implies that

$$N_e N_n K_{n,m} = N_e N_m K_{mn},$$

when all the constituents of the reaction are at thermodynamic equilibrium. Using the equilibrium values of N_n and N_m at temperature T and the Boltzmann distribution for $dN_e(E)/dE$ and $v(E)$, the de-excitation rate coefficient, in terms of the excitation cross section, can be shown to be

$$G_{m,n} \equiv g_m K_{m,n} = \frac{g_n}{\sqrt{\pi m_e}} \left(\frac{2}{kT}\right)^{3/2} \times \int_0^{\infty} (E+E_{\mathrm{ex}}) Q_{n,m}(E+E_{\mathrm{ex}}) e^{-E/kT} dE, \quad (3)$$

for a Boltzmann distribution of electron velocities at a temperature T.

In terms of this expression Eq. (2), with $dN_n/dt=0$, can be rewritten as

$$\sum_{m=n+1}^{\infty} G_{m,n} D_{m,n} = L_n + \sum_{m=1}^{n-1} G_{n,m} D_{n,m}, \quad (4)$$

where

$$D_{m,n} = N_m/g_m - N_n/g_n \exp[(E_n - E_m)/kT]$$

and

$$L_n = \frac{1}{N_e} \{ N_n \sum_{m=1}^{n-1} A_{nm} - \sum_{m=n+1}^{\infty} N_m A_{mn} \}.$$

Calculations of $G_{m,n}$ using Gryzinski's cross sections show that the following approximations:

$$G_{m+2,m} \approx 0.24 \quad G_{m+1,m}$$
$$G_{m+3,m} \approx 0.10 \quad G_{m+1,m}$$
$$G_{m+1,m-1} \approx 0.03 \quad G_{m+1,m}$$

are obeyed for all n of interest. Using these approximations and neglecting $G_{m+1,m-1}$ and all other collisional transitions, Eq. (4) can be written as

$$G_{n+1,n}(D_{n+1,n} + \Delta_{n+1,n}) = L_n + G_{n,n-1} D_{n,n-1}, \quad (5)$$

where

$$\Delta_{n+1,n} = 0.24 D_{n+2,n} + 0.10 D_{n+3,n}.$$

The factors $D_{n+1,n}$, $\Delta_{n+1,n}$, and L_n can be determined from the experimentally measured densities of the various levels for $n \geq 3$. The method of obtaining $G_{n+1,n}$ then consists of estimating $G_{3,2}$ (in some cases this term is negligible), and solving Eq. (5) for $G_{4,3}$. This value of $G_{4,3}$ is then used to solve the equation for $n=4$, etc., until the terms $D_{n+1,n}$ become zero as the density of level n approaches equilibrium with the free electrons.

[14] D. R. Inglis and E. Teller, Astrophys. J. 90, 439 (1939).

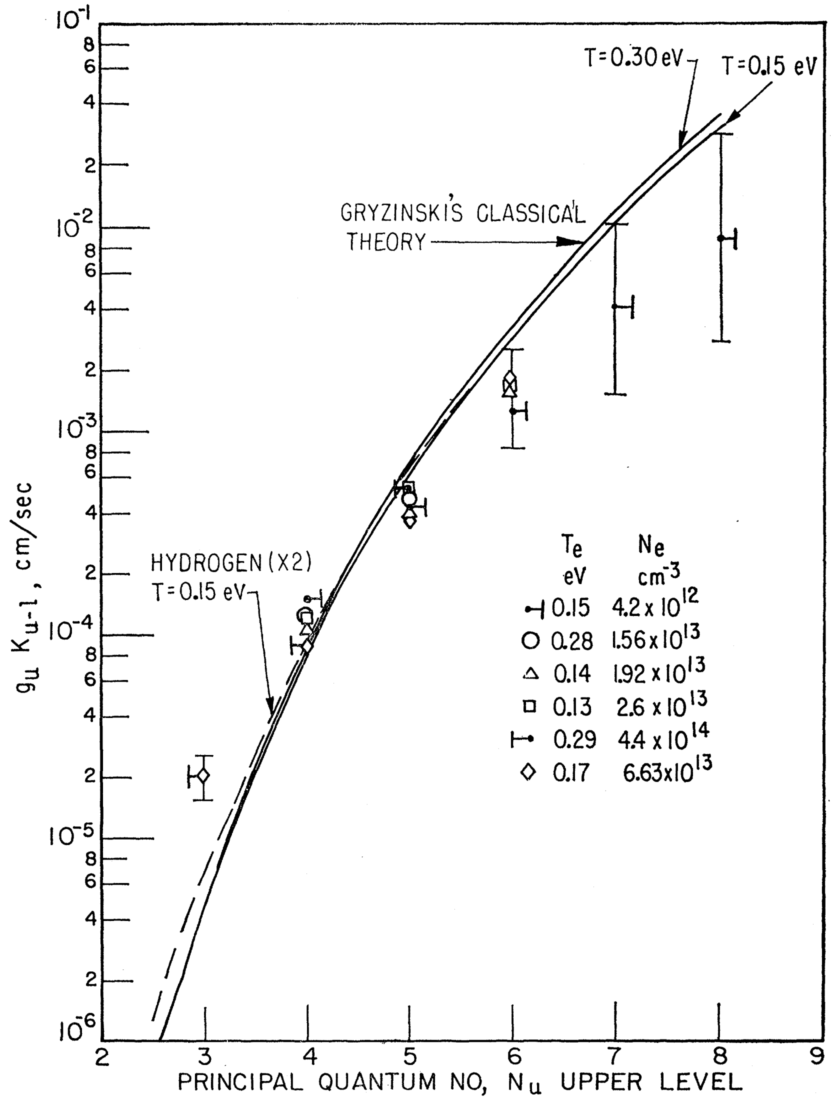

FIG. 3. The collisional de-excitation rate constant multiplied by the multiplicity, versus the principal quantum number. This is the rate constant from the averaged level with principal quantum number n to the averaged level with principal quantum number $n-1$.

In the calculation of the radiative factor L_n in Eq. (5) the terms involving radiation to the ground level are dropped, as the radiation is trapped. Assuming Doppler broadened lines (Stark and collision broadening are negligible), the spectral absorption coefficient at the line center in cm^{-1} atm^{-1} is given by[15]

$$P(0)=S_{lu}(mc^2/2\pi kT)^{1/2}\lambda, \qquad (6)$$

where S_{lu} is the integrated absorption coefficient in cm^{-2} atm^{-1} and related to the absorption oscillator strength by[15]

$$S_{lu}=2.38\times10^7 f_{lu}.$$

In Eq. (6) m is the atomic mass and λ is the wavelength of the transition in cm. For the helium resonance transition $1\,^1S-2\,^1P$ at 537 Å, $f_{lu}\approx0.072$. Assuming typical conditions in the plasma jet, $T=1000$°K and 1.7×10^{17} atoms in the ground state, the absorption at line center given by Eq. (6) is found to be 50 cm^{-1}. Thus, the mean free path of resonance radiation at the line peak is about 0.2 mm, so that to a good approximation in Eq. (5) the resonance radiation can be neglected compared with the other terms.

The results of this calculation of $G_{n,n-1}$ are shown in Fig. 3, as well as the values calculated using Gryzinski's cross sections with averaged energies of the helium levels. The value of $G_{3,2}$, important at the higher electron densities, was adjusted so as to bring all the points into reasonable agreement. Agreement with the classical theory is quite good; however, it appears to give values somewhat too large for the levels with $n>5$, and somewhat too small for levels with $n<4$. The relative constancy of $G_{n,n-1}$ over a factor of 100 in electron density

[15] S. S. Penner, *Quantitative Molecular Spectroscopy and Gas Emissivities* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1959), p. 31.

TABLE II. Comparison of recombination rates.

N_e (cm^{-3})	T_e (eV)	α in 10^{-11} cm^3 sec^{-1} From present experiment	From Bates *et al.*	From Hinnov and Hirschberg
4.2×10^{12}	0.15	5.8	14	13
1.6×10^{13}	0.28	1.9	3.3	1.5
1.9×10^{13}	0.15	30	68	58
2.6×10^{13}	0.13	89	150	130
6.6×10^{13}	0.17	34	65	111
4.4×10^{14}	0.29	25	33	13

can be regarded as a verification of the model of recombination.

The rate of recombination, according to the model, is given by

$$\frac{dN_e}{dt}=-\sum_{m=3}^{\infty} N_m A_{m,2}+N_e G_{3,2}(D_{3,2}+\Delta_{3,2}), \qquad (7)$$

with the radiative term being dominant in all cases except those with the highest electron density. As stated previously, resonance radiation to the ground level is neglected as it is trapped. In Table II the recombination coefficient α as defined by

$$dN_e/dt=-\alpha N_e^2$$

is given for most of the experiments, using the results of Eq. (7). Values of α interpolated from the table of Bates, Kingston, and McWhirter[4] for trapped resonance radiation, and from the graph given by Hinnov and Hirschberg,[2] are also shown in Table II for comparison. These values depend on the experimentally determined T_e and N_e. The values of Bates *et al.* are generally about a factor 2 greater than the experimental values from Eq. (7). This is a somewhat surprising result, since their calculations were based on the same model of recombination used in the present data reduction and relied on Gryzinski's expression for the collisions cross sections, implying that the two results should agree, within uncertainties of 50% or so.

[*Note added in proof.* A check on our procedure (and their results) was performed by application of our method of analysis to their calculated number densities of excited atoms.[16] The collisional de-excitation rates calculated were found to be in agreement with Gryzinski's theory as shown in Fig. 4, and the recombination rates were in agreement with their tabulated values. It appears that the discrepancy is due to a combination of minor differences in the electronic rates, and to inaccuracies in the long interpolations necessary in the tables of recombination coefficients given by Bates *et al.*]

The results of Hinnov and Hirschberg are a more approximate calculation, still based on the same model of recombination, using collisional cross sections estimated from the classical ionization cross section and the oscillator strengths of the allowed optical transitions. They were made for helium, and agree with their experimental measurements on the recombining helium plasma in a stellarator with an error of less than 50%. Their values are also generally somewhat greater than those of the present experiment, but probably within the maximum experimental uncertainties.

The temperature dependence of the de-excitation rate constant, Eq. (3), is related to the form of the excitation cross section, $Q_{n,m}(E+E_{ex})$. If we assume that

$$(E+E_{ex})Q_{n,m}(E+E_{ex})\propto E^r$$

then it is easy to show that

$$G_{m,n}\propto T^{r-\frac{1}{2}}.$$

An examination of the experimental data plotted in Fig. 3 reveals no systematic variation of $G_{m,n}$ with electron temperature, indicating that $r=\frac{1}{2}$ for the range of electron energy around kT. With the approximation that $kT\ll E_{ex}$, the expressions given in Table III can be calculated for the average excitation cross sections. This excitation cross section must be considered to be a sum over all the levels belonging to the upper principal quantum number, and an average, according to the distribution, over all the levels belonging to the lower quantum number. Theoretical considerations[17] show that these cross sections should have a square root dependence on E near $E=0$, and the results given here are consistent with this behavior up to an energy of about 0.3 eV. Assuming, then, this dependence on E from threshold up to an energy of about 0.3 eV, the values of the proportionality constants in Table III have an estimated error of about 30%.

The classical cross section derived by Gryzinski[5] applies to the energy transfer between two free electrons. The expression given by his Eq. (23), when written in the form $\sigma(E_1,\Delta E,E_2)d(\Delta E)$, is the cross section for excitation of an electron with energy E_1 to an energy between $E_1+\Delta E$ and $E_1+\Delta E+d(\Delta E)$, in a collision with another electron of energy E_2. The direction of the velocity of electron 1 has been assumed to be isotropic in space, and collisions with long interaction times have been neglected. In applying the above results to excitation of an atom, the bound electron is assumed to be in a circular Bohr orbit, from which the

TABLE III. Excitation cross section for helium atoms. (Averaged over all levels with principal quantum number n and summed over all levels with principal quantum number $n+1$.) In units of 10^{-15} cm^2 with E in eV.

$Q_{2,3}(E+E_{ex})=9.3\times\sqrt{E}$
$Q_{3,4}(E+E_{ex})=73\times\sqrt{E}$
$Q_{4,5}(E+E_{ex})=380\times\sqrt{E}$

[16] D. R. Bates and A. E. Kingston, Planetary Space Sci. **11**, 1 (1963).

[17] M. J. Seaton, in *Atomic and Molecular Processes*, edited by D. R. Bates (Academic Press Inc., New York, 1962), p. 374.

kinetic energy E_1 can be found. The cross section for excitation of a level with principal quantum number n to level m is thus given by

$$Q_{n,m}=\sigma(E_1,\Delta E,E_2)(E_{m+1}-E_m),$$

where ΔE is the excitation energy, E_m-E_n, of the level, and E_{m+1} is the energy of the level in the same series with principal quantum number $m+1$. This form gives the threshold variation of the cross section as $(E_2-\Delta E)^{\frac{1}{2}}$, in agreement with the quantum-mechanical considerations. Use of this cross section to calculate $G_{n,n-1}$ results in fair agreement with the experiment, as already shown in Fig. 3. It appears, though, that the semiclassical value of $Q_{2,3}$ is too small by a factor of 3, and for the higher quantum numbers the semiclassical values may be somewhat too large. It might be expected, from correspondence principle arguments, that the classical cross section would be asymptotic to the actual values at large quantum number. However, the approximations inherent in Gryzinski's classical theory in the limit of high quantum number do not allow such a comparison.

The cross section for excitation of hydrogen from the ground level to the $2P$ level has recently been measured by atomic beam methods[18] in the energy range near threshold, and the results fit the curve

$$Q_{1,2}(E+E_{\mathrm{ex}})=0.29E^{\frac{1}{2}} \tag{8}$$

for energies up to 3 eV. Results for excitation from $1S$ to $2S$ have also been determined,[19,20] and although they have not been shown to fit a curve of the form of Eq. (8), the threshold cross section appears to be somewhat less, but of the same order of magnitude, as that of Eq. (8). It is interesting to note that this value for hydrogen agrees quite well with Gryzinski's theory. If it is true that the excitation cross sections for hydrogen and helium are about the same for the higher quantum numbers, then it appears that Gryzinski's theory gives about the correct results for Q_{1-2}, a factor 3 too low for Q_{2-3}, and again about the correct result for Q_{3-4} and Q_{4-5}.

[18] W. L. Fite, R. F. Stebbings, and R. T. Brackman, Phys. Rev. **116**, 356 (1959).
[19] W. Lichten and S. Schultz, Phys. Rev. **116**, 1132 (1959).
[20] R. F. Stebbings, W. L. Fite, D. G. Hummer, and R. T. Brackman, Phys. Rev. **119**, 1939 (1960).

SUMMARY

Detailed measurements of the number density of the excited levels of helium in a recombining plasma have been shown to be in good agreement with the collisional-radiative model of recombination. These measurements led to a determination of the average excitation cross section of several levels of helium in the energy range from threshold to about 0.3 eV excess energy which are proportional to the square root of the excess energy, as suggested by quantum mechanical theory. These cross sections are also in approximate agreement with values calculated by the classical expression of Gryzinksi.[5] The tables of recombination coefficient calculated by Bates, Kingston, and McWhirter[4] are in approximate agreement with the recombination coefficient inferred from experiment, as expected, since Bates *et al.* used Gryzinski's cross section in their calculations.

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