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Recombination Spectrum and Electron Density Measurements in Neon Afterglows*

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The spectrum emitted by the recombination of positive ions and electrons in the afterglow of a pulsed microwave discharge through neon has been studied as a function of time. Simultaneous electron density measurements on the plasma were made by the cavity resonant frequency shift method, and the recombination coefficient was calculated from these data. The total radiated visible light was found to be proportional to the square of the observed electron density. The observation of light intensities allowed recombination measurements to be carried out at electron densities much too high to be observed directly by the cavity resonant frequency shift method. The *relative* intensities of several strong lines in the spectrum remained essentially constant during the afterglow, while the *relative* intensities of several others showed consistent changes.

INTRODUCTION

The general problem of the mechanism of the disappearance of electrons from a gas discharge plasma by various processes has been the subject of an enormous amount of theoretical and experimental work.[1] In many cases, it is possible to choose experimental conditions such that virtually all of the electrons are lost by one process (*e.g.*, recombination with positive ions). In the present investigation simultaneous spectroscopic and electron density measurements were made on neon discharges under conditions favoring electron-positive ion recombination.

Recombination spectra have been studied in argon[2] and metal vapors.[3] The results of these studies have been used in conjunction with probe measurements to determine the recombination coefficient,[4] α, for electron-positive ion recombination. The values of α obtained were of the order of 10^{-10} cm^3/ion-sec., compared to a theoretical value (based on the use of hydrogen-like wave functions)[5] of about 10^{-12} cm^3/ion-sec. These values are in poor agreement with the values of α of the order of 10^{-6}–10^{-8} cm^3/ion-sec. which have recently been obtained[6] for several gases by means of microwave electron density measurements.

METHOD AND APPARATUS

A pulsed microwave (3000 mc/sec.) electrodeless discharge through spectroscopically pure neon at pressures between 10 and 30 mm Hg was used in this work. Any other method of exciting the original discharge would probably have been equally satisfactory; but, since it was desired to make microwave electron density measurements on the plasma, the microwave breakdown was selected for convenience. The measurements consisted of observing the spectrum as a function of time after cutting off the discharge, and measuring simultaneously the electron density by determining the shift of the resonant frequency of the cavity in which the discharge occurred. As can easily be seen from the definition of α, a plot of $1/n$ *vs.* time should result in a straight line provided virtually all of the electrons originally present in the plasma disappear due to recombination. If the electrons disappear due to ambipolar diffusion, it can easily be shown that the plot of log n *vs.* time should be a straight line—this gives a simple method of distinguishing experimentally between the two processes. If the total intensity of light emitted is simply proportional to the rate of disappearance of electrons, then a linear relationship between the square root of the intensity and time would be expected, for the range of conditions under which recombination is the principal process of electron removal.

Figure 1 is a block diagram of the apparatus used. The operating sequence, which repeated 200 times per second, follows. The timing of the sequence was controlled by a rotating wheel. Light from an incandescent bulb was sent through the slit in the wheel and reflected by a mirror onto a photo-multiplier, which generated an electrical trigger. The trigger was passed through a

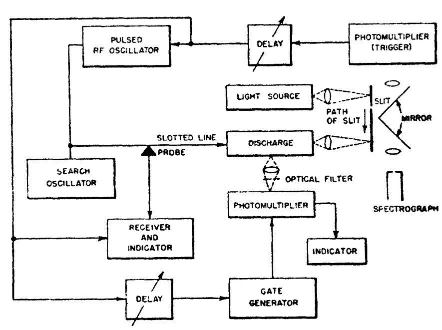

Fig. 1. Block diagram of apparatus used for studying recombination spectrum and variation of electron density in neon afterglow.

* The work described in this paper was assisted by the ONR.

[1] For a good summary of work before 1939, see L. B. Loeb, *Fundamental Processes of Electrical Discharge in Gases* (John Wiley and Sons, Inc., New York, 1939) Chapters II and III.

[2] C. Kenty, Phys. Rev. **32**, 624 (1928).

[3] F. L. Mohler and C. Boeckner, Bur. Stand. J. Research **2**, 489 (1929); C. Broeckner, Bur. Stand. J. Research **6**, 277 (1931); F. L. Mohler, Bur. Stand. J. Research **10**, 771 (1933); J. Research Nat. Bur. Stand. **19**, 447 (1937); and **19**, 559 (1937).

[4] Defined by the equation $dn/dt = -\alpha n^2$, where n is the electron concentration.

[5] Stueckelberg and Morse, Phys. Rev. **36**, 16 (1930).

[6] Quarterly Progress Report, Research Laboratory of Electronics, M.I.T. (January 15, 1949), p. 6 ff.

variable delay (0 to 3000 microseconds) and used to excite the pulsed r-f oscillator for 10 microseconds. The power thus generated (variable to approximately 1-kw peak) was passed through a slotted coaxial line and used to break down the vycor discharge tube, which was contained in a cylindrical cavity tuned near the pulsed oscillator frequency. The spectrum of the discharge was examined by allowing the light from the discharge to fall on the wheel a little distance from the triggering light and to pass through the same slit which was responsible for the generation of the trigger. After passing through this slit, the light from the discharge was reflected by a mirror onto the slit of a Kipp high intensity liquid prism spectrograph. The spectrographic plates obtained were run through a microphotometer to determine wave-lengths and relative intensities of spectral lines. Type 103a Eastman plates were used in some of the runs involving widely different exposure times to minimize reciprocity law failure. By varying the electrical delay, the light from the discharge could be examined during the discharge or at any time after the discharge up to a maximum of 3000 microseconds. The size of the slit and angular speed of the wheel were such that a time resolution of 2 microseconds could be achieved.

The apparatus was arranged so that the light from the discharge could also be viewed by a photo-multiplier (or, for the infra-red, by a Type 1P25 infra-red image tube). Because of the tremendous differences in light intensity encountered at various times after the discharge, it was necessary to "gate" the photo-multiplier or image tube so that it was sensitive only at the time after the discharge at which an observation was being

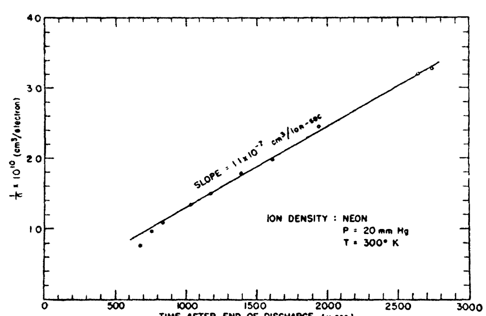

FIG. 2. Variation of electron density as a function of time.

made. This was accomplished by the use of a second variable delay circuit which operated from the original electrical trigger and furnished a delayed trigger to activate the photo-multiplier pulsing circuit. A synchroscope plus a calibrated video attenuator constituted the indicating equipment for the photo-multiplier.

Finally, the electron density in the plasma was measured in the following manner. A small signal from the search oscillator,[7] which was tuned to an accurately known frequency differing from the natural resonant frequency of the cavity by a few megacycles, was injected into the slotted line. The presence of electrons in the gas discharge plasma causes the resonant frequency of the cavity to be shifted from its normal value; and, in fact, it can be shown that under suitable experimental conditions, which were maintained here, the frequency shift is directly proportional to the concentration of electrons present, the exact factor of proportionality depending somewhat on the spatial distribution. By placing the probe of the slotted line at a suitable point in the standing wave pattern, it is possible to cause a minimum amount of power to flow into the receiver and indicator if the injected frequency and the cavity frequency coincide. Since the concentration of electrons in the plasma, and therefore the cavity frequency, was changing due to recombination, such a minimum did occur, and its development was fast enough to produce a pulse when receiver output was displayed on the indicator time base. It thus sufficed to observe the search frequency and the time after the discharge at which the pulse occurred in order to obtain one point on the electron density *vs.* time curve. To obtain other points, the search frequency was varied.

RESULTS

The variation of electron density as a function of time is shown in Fig. 2 for a typical run at a pressure of 20 mm Hg. The $1/n$ *vs.* time plot gives a good straight line, indicating that, in the range of experimental conditions used, recombination far exceeded diffusion as a mechanism of electron loss. The slope of the line gives $\alpha = 1.1\times 10^{-7}$ cm³/ion-sec., and is considered in satisfactory agreement with the value 2.1×10^{-7} cm³/ion-sec. obtained previously for neon with this same method at a lower peak input power.[6] We observed that the measured value of α increases with decreasing peak input power, varying over a 2:1 range which contains the previously reported figure. These observations are satisfactorily explained by noting that unless a large density of electrons are present initially in the plasma, a uniform spatial distribution (which is assumed in order to calculate the density) is not fully achieved from the non-uniform one existing at the end of the discharge by the time measurements begin. If such experimental conditions exist, some other distribution function may be reasonably assumed in order to preserve the value of α.

In order to test the assumption that the light intensity should be proportional to the square of the electron density, the data for total light intensity for two regions of the spectrum and the corresponding electron density data are plotted in Fig. 3. It should be remembered that the most that can be said of the light intensity is that the total energy radiated (regardless of the spectral distribution) should, in the absence of com-

[7] The method used was developed by S. C. Brown, M. A. Biondi, M. A. Herlin, E. Everhart, and D. E. Kerr, and is described in detail in Technical Report No. 66, Research Laboratory of Electronics, M.I.T. (1948).

plicating factors such as long-lived metastable levels, be proportional to the square of the electron density. The fact that the data show that this proportionality holds at least fairly well for two broad regions of the spectrum agrees with the assumption, and furthermore indicates that the mechanism of the recombination process probably does not change radically during the period of the afterglow studied, since a change in the mechanism would probably affect the spectral distribution. This is not intended to be a conclusive argument.

It should be noted that the light intensity measurements have allowed us to take data to shorter times after the discharge than would be possible with the electron density measurements. This is so because of the limitations imposed on the electron density method at relatively high electron densities (greater than about 10^{10} electrons/cm^3) because of the breakdown of the assumption (which is made in deriving the equations) that the presence of the electrons perturbs the cavity fields only slightly.

It was noted that the intensity of the afterglow at a particular time was the same whether the search signal was on or off for sufficiently low signal levels. This verifies the fact that it is possible to inject a signal large enough to make electron density measurements without affecting the plasma appreciably.

The microphotometer traces of the plates taken with the spectrograph showed several interesting features. Only lines identifiable as atomic neon were obtained. Secondly, the spectra were virtually pure line spectra. It would be expected that the initial process of an electron possessing thermal kinetic energy falling into a quantized level would give rise to a continuum within the resolution of our instrument beyond the series limit associated with the particular quantized level involved. Line spectra would then be expected as the secondary consequence of the atom (which would be left in an excited state unless the level into which the electron was captured was the ground state) returning to the ground state by the emission of one or more quanta. The absence of strong continua in the visible and near ultraviolet shows qualitatively that the upper quantum levels (which would give rise to continua in the far infra-red) are probably more important in the recombination process than has previously been supposed.

The *relative* intensities of various spectral lines were found to change very rapidly in the first few microseconds after the discharge, and then much more slowly during the remainder of the afterglow. The rapid change in the early part of the afterglow is to be expected, since a little time is required for the establishment of a stationary state of electron temperature in the plasma, etc. As an example of the type of change found in the spectrum of the later afterglow, the line 6678A ($^2P_{\frac{1}{2}}3p8-3p4$ transition) gave a photographic density roughly twice that produced by the line 5852A ($^2P_{\frac{1}{2}}3p10-3s4$ transition) at a time of 100 microseconds after the discharge, while the two lines produced the same photographic density 700 microseconds after the discharge. Similar somewhat smaller changes could be observed with many other lines, but the majority of the lines observed did not change appreciably in relative intensity, thus agreeing in detail with the photo-multiplier data already cited for various fairly wide regions of the spectrum. At any rate, a careful examination of the data did not reveal any striking *systematic* shift in the relative intensities of the lines which might have been correlated with a change in the relative importance of various quantum levels in the initial capture of electrons.

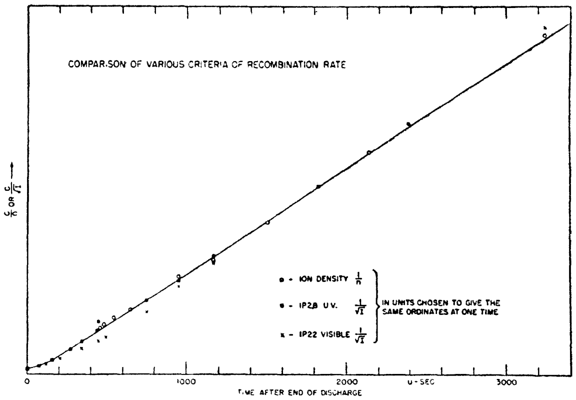

FIG. 3. Electron density; visible and ultraviolet light intensities, as a function of time.

ACKNOWLEDGMENT

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