ELECTRON-ION RECOMBINATION COEFFICIENTS IN NITROGEN AND IN OXYGEN*

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Currently, there is considerable interest in predicting free-electron lifetimes in various regions of the ionosphere following ionizing events, such as solar flares. An important process in determining the electron loss is recombination between electrons and the positive ions present in the upper atmosphere. It has been reasonably well established from analysis of ionospheric observations[1] and from laboratory experiments[2,3] that the recombination coefficient α (defined by $dn_e/dt = -\alpha n_e n_+$, where n_e and n_+ are the electron and positive ion densities, respectively) may be as large as 10^{-8} to 10^{-6} cm^3/sec and that the capture process is dissociative recombination.[4]

Since no adequate theoretical calculations of the rate of the dissociative recombination reaction,[5] e.g.,

$$(XY)^+ + e \longrightarrow (XY)^* \xrightarrow[\text{unstable}]{} X^* + Y, \qquad (1)$$

are available, appeal is generally made to laboratory measurements of the recombination coefficients in atmospheric gases.[2,6] Unfortunately, previous measurements were made without identification of the ions responsible for the recombination and under conditions where more complex ion types, e.g., O_3^+ in oxygen and N_3^+ and N_4^+ in nitrogen, are observed. This Letter presents the preliminary results of an experiment involving microwave determinations of the decay of electron density during the afterglow following ionization of gas in a cavity,[7] coupled with the simultaneous identification, by an rf-type mass spectrometer, of the afterglow positive ion currents to a wall of the cavity. From these measurements, recombination coefficients appropriate to particular positive ion species, e.g., N_2^+, can be assigned.

The apparatus consists of a rectangular, stainless steel, microwave cavity containing in one wall a quartz iris for coupling microwave energy, in the opposite wall a 0.008-inch diameter effusion orifice leading to a Boyd-type[8] rf mass spectrometer and its differential pumping system, in a third wall an optical viewing port, and in a fourth wall a lead to an ultra-high vacuum, gas-handling system. The gas-handling system, the cavity, and the mass spectrometer differential pumping system are baked at 350°C for 14 hours to minimize impurity effects. The high-purity gas samples,

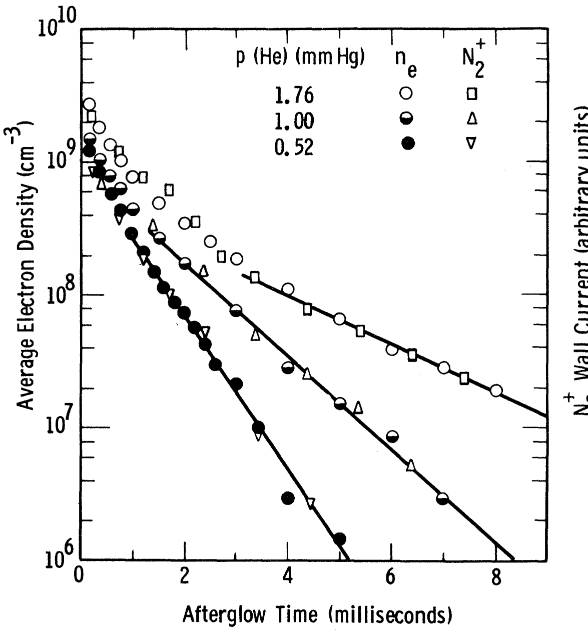

FIG. 1. Electron and N_2^+ ion decay in the afterglow of N_2-He microwave discharges having constant nitrogen pressure, 0.006 mm Hg, and variable helium pressure. The N_2^+ wall currents are normalized to the respective electron density decay curves at an afterglow time of approximately 2 milliseconds.

contained in steel pressure tanks,[9] are fed into the system through controlled leaks.

In order to carry out measurements at low concentrations of nitrogen or oxygen, an inert buffer gas is used to inhibit particle loss by diffusion to the walls. It was observed that in either pure nitrogen or nitrogen-helium mixtures, the predominant afterglow ions for nitrogen pressures in the range 0.1 to 7.0 mm Hg were N_3^+ and N_4^+, while at nitrogen pressures less than 0.01 mm Hg, N_2^+ was the only significant ion in the afterglow.[10] Similarly, in oxygen, at partial pressures of less than 0.005 mm Hg, O_2^+ predominated.

At low total pressures, see Fig. 1, the predominant loss of electrons and N_2^+ ions in the late afterglow is by ambipolar diffusion to the walls, as indicated by the fact that the rate of particle loss varies inversely with gas pressure and exponentially with time. The curvature in the top curve shows significant electron-ion recombination in the early afterglow. From the known fundamental diffusion length of the cavity, $\Lambda = 1.126$ cm, a $D_a p$ value of approximately 900 (cm²/sec) × (mm Hg) is obtained for N_2^+ ions and electrons moving in helium. At this low partial pressure of nitrogen, 0.006 mm Hg, N_3^+ and N_4^+ ions are essentially absent. The decay of the normalized wall current of N_2^+ ions (which, for fundamental mode diffusion, is proportional to the average density of N_2^+ inside the cavity) follows the average electron density, within the accuracy of the present measurements.[11]

In order to study recombination under conditions where diffusion loss is unimportant, it is necessary to increase the pressure of the buffer gas to reduce the diffusion rate. Neon is more suitable than helium, the gas conventionally used,[6,12] since, at a given pressure, neon inhibits diffusion more strongly[13] and effuses into the mass spectrometer more slowly than helium, thus reducing choking of the mass spectrometer pumping system. The ionization potential of neon is sufficiently high that one observes only ions of the minority nitrogen or oxygen constituent.

Measurements of electron-ion recombination have been carried out under conditions where one particular ion species (except for the N_3^+-N_4^+ combination) predominates during the afterglow. Under these conditions, and when diffusion loss can be neglected, the solution of the electron density equation becomes $(1/n_e) = (1/n_{e0}) + \alpha t$. Examples of data obtained for recombination loss of N_2^+ ions and electrons and for O_2^+ ions and electrons are shown in Fig. 2. The neon gas pressure is sufficiently high so that the ratio of recombination loss to fundamental mode diffusion loss is large over the entire range, varying from ~200 to ~10 in the data shown. From the initial straight portions of the curves, covering a factor of ~10 in electron density, taken at several neon partial pressures, we obtain the values $\alpha(N_2^+) = (5.9 \pm 1) \times 10^{-7}$ cm³/sec and $\alpha(O_2^+) = (3.8 \pm 1) \times 10^{-7}$ cm³/sec. The uncertainty in these values arises largely from imperfect knowledge of (a) the correction of the recombination curve for diffusion effects in this geometry,[14] (b) the spatial density distribution of the electrons, which affects the determination of absolute electron density from the measured cavity frequency shifts,[7] and (c) the time-resolved afterglow ion currents, which indicate the importance of any other positive ions in the afterglow. Electron attachment in oxygen should not be a significant factor in the recombination analysis, since the oxygen partial pressure was so low,

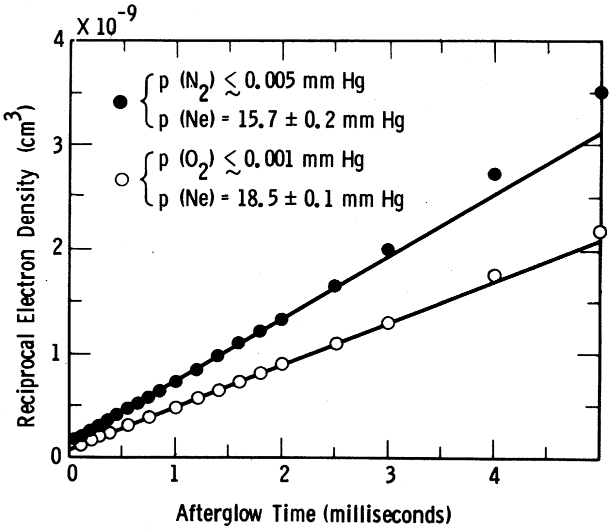

FIG. 2. Time variation of the reciprocal electron density in the afterglow of high-pressure N_2-Ne and O_2-Ne microwave discharges.

i.e., less than 10^{-3} mm Hg.

At higher partial pressures of nitrogen, N_3^+ and N_4^+ ions appear in nearly equal abundance and seem to vary together with both gas pressure and time during the afterglow. The curves of $1/n_e$ vs time are linear in this case also, indicating an effective recombination coefficient of the order of 2×10^{-6} cm³/sec.

The present results indicate that at the pressures used in the earlier work,[2,6] the ions under study were not N_2^+ and O_2^+, as had been assumed. Our preliminary value for $\alpha(N_3^+$-$N_4^+)$ agrees with that reported in the earlier nitrogen work[2] and with some recent results of Van Lint et al.,[15] while our value for $\alpha(N_2^+)$ is substantially smaller, thus indicating the need for ion identification in atmospheric gas studies of this type. Studies of the recombination coefficients of other ions, such as $(NO)^+$, of importance in upper atmosphere problems, are being considered.

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[1]See, for example, D. R. Bates and H. S. W. Massey, Proc. Roy. Soc. (London) A187, 261 (1946); H. S. W. Massey, Advances in Physics, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1952), Vol. 1,

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[8]R. L. F. Boyd and D. Morris, Proc. Phys. Soc. (London) 68, 1 (1955). The instruments used in the present study were manufactured by Mullard Ltd., Century House, Shaftesbury Avenue, London, England.

[9]The gases were obtained from the Matheson Company, Inc., East Rutherford, New Jersey. Undesirable impurities are less than 1 part in 10^5.

[10]According to the observations of Faire and Champion, the recombination coefficient for electrons and nitrogen ions is constant for nitrogen pressures from 0.011 mm Hg to 4 mm Hg. It is difficult to understand this result since the present measurements yield more than an order-of-magnitude change in the relative concentrations of the various nitrogen ions over the same pressure range.

[11]The time-resolved afterglow ion currents reaching the collector of the spectrometer are small, $\sim 10^{-15}$ amp, leading to small signal/noise ratios. A modified instrument using an ion multiplier at the collector is undergoing tests.

[12]M. A. Biondi, Phys. Rev. 93, 1136 (1954).

[13]Studies of ambipolar diffusion in neon at low total pressures gave values of $D_a p \sim 450$ (cm^2/sec)(mm Hg) for both N_2^+ and O_2^+ ions; however, diffusion cooling effects in neon (see reference 12) caused some difficulty in these studies.

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PHOTOPRODUCTION OF SINGLE NEUTRAL PIONS FROM HYDROGEN AT 60° IN THE 600-1100 Mev REGION

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In a recent experiment Cortellessa and Reale[1] measured the differential cross section for the process $\gamma + p \to \pi^0 + p$ in the neighborhood of the second resonance at a center-of-mass angle for the neutral pion of 57°. Their results, obtained with a proton telescope in coincidence with a lead glass Čerenkov counter used to detect one of the π^0 decay photons, are considerably different from those which had been reported previously. Earlier data were obtained by Vette[2] using a proton magnetic spectrometer, by Stein and Rogers[3] and Worlock[4] using proton telescopes, and by DeWire, Jackson, and Littauer[5] using a proton telescope plus Čerenkov gamma-ray counter. Some of the difference can be attributed to the better resolution of Cortellessa and Reale which was ±30 Mev in incident photon energy, about half that of the previous experiments. However, even taking into account this difference in resolution, a serious disagreement exists between the experiment of Cortellessa and Reale and the earlier ones in that Cortellessa and Reale obtain values for the cross section which are, in general, considerably lower than those previously obtained. Furthermore, Cortellessa and Reale find a sharp peak in the cross section at a photon energy 700 Mev, which is about 50 Mev lower than the energy corresponding to the "second resonance" peak observed in π-nucleon scattering.[6] Although the excitation curve at any given angle need not have a maximum at the same energy as the total cross section, the angular distributions for π^0 photoproduction do not seem to be changing rapidly in this region, so that one might expect the peak at 57° to be characteristic of the total cross section and to be centered closer to 750 Mev than 700 Mev as measured by Cortellessa and Reale. (The fact that π^+ photoproduction shows a peak at 700 Mev is probably the result of an interference effect with the meson current term which does not appear in π^0 photoproduction.[7])

Intrigued by this apparent discrepancy and by the interesting shape of the curve reported by Cortellessa and Reale, we have measured the π^0 cross section in this region and extended the measurements through the third resonance using the