Recombination and Ion-Conversion Processes in Helium-Neon Mixtures*

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(Received 26 March 1970)

Detailed studies of the time dependence of the spectral emission and that of the number density of ions have been made during the decay period of plasmas produced in helium containing various concentrations of neon atoms. The occurrence of the recombination of Ne^+ via the collisional recombination process with a recombination coefficient proportional to the electron density is established for the first time. The studies confirm the occurrence of recombination processes previously reported for He_2^+ and Ne_2^+. Evidence has also been found for the population of the $4d$ levels of neon by $He(2^1S)$ metastable atoms. New values for the rate constants of the following processes were obtained: $Ne^+ + 2He \rightarrow (HeNe)^+ + He$ (2.1×10^{-32} cm^6 sec^{-1}); $Ne^+ + Ne + He \rightarrow (HeNe)^+ + Ne$ and $\rightarrow Ne_2^+ + He$ (3.0×10^{-31} cm^6 sec^{-1}); $(HeNe)^+ + Ne \rightarrow Ne_2^+ + He$ (3×10^{-11} cm^3 sec^{-1}) – all at a gas temperature of 300 °K.

I. INTRODUCTION

Several studies have been reported concerning the measurement of the electron density during the decay period of a plasma produced in neon.[1–5] The high value obtained for the electron-ion recombination coefficient was taken to be evidence for the dissociative recombination process

$$Ne_2^+ + e \rightarrow Ne^* + Ne \rightarrow 2Ne + h\nu \ . \qquad (1)$$

In 1965 Connor and Biondi[6] presented further evidence for the occurrence of process (1) during the decay period of a neon plasma. They measured the spectral line broadening caused by the kinetic energy of dissociation of the excited atoms produced. Recently, Frommhold and Biondi[7] confirmed and refined the line-profile studies.

The first simultaneous light-emission and ion-density studies of the neon afterglow were reported in 1966 by Sauter *et al.*[8] The measurements confirmed the loss of Ne_2^+ ions by the dissociative process (1). In addition, their light-intensity measurements showed that the spectral lines emitted during the decay period could be divided into two groups. The time dependence of the intensity of lines belonging to the same group was closely the same but the time dependences of the two groups were different. It was determined that the recombination of Ne_2^+ with electrons was responsible for the population of the $2p$ and $3p$ levels but not the d levels. The suggestion was made that the d levels were populated by the recombination of Ne^+ with electrons. However, no definite conclusion could be reached as to the type of recombination process involved.

The most likely processes for spectral line emission due to the recombination of Ne^+ ions are the radiative recombination process

$$Ne^+ + e \rightarrow Ne^* \rightarrow Ne + h\nu \qquad (2)$$

and the collisional recombination process

$$Ne^+ + 2e \rightarrow Ne^* + e \rightarrow Ne + e + h\nu \ . \qquad (3)$$

The latter process is the established recombination process for He^+ and Ar^+ ions.[9,10]

Studies of the time dependence of the electron density during the decay period of plasmas produced in helium-neon mixtures have been reported by Oskam.[3] He postulated the occurrence of the following sequence of processes during the decay period:

$$He_2^+ + Ne \rightarrow Ne^+ + 2He \ , \qquad (4)$$

$$Ne^+ + Ne + He \begin{cases} \rightarrow Ne_2^+ + He & (5a) \\ \rightarrow (HeNe)^+ + Ne \ , & (5b) \end{cases}$$

$$Ne^+ + 2He \rightarrow (HeNe)^+ + He \ . \qquad (6)$$

The first simultaneous spectral-emission and ion-density study of a decaying plasma in a helium-neon mixture was performed by Sauter *et al.*[8] The measurements were made for a single fixed concentration of neon in helium. The mixture was obtained by using commercially available helium which was not purified by the cataphoretic segregation process.

The present studies relate to plasmas produced in well-defined helium-neon mixtures and are concerned with the determination of the types of electron-ion recombination processes occurring in these plasmas. In addition, the rate coefficients of various ion-conversion processes will be given. Several of the rate coefficients differ from previously reported values.

II. EXPERIMENTAL METHOD

The experimental tube used to study the time dependence of the ions during the afterglow period consists of a differentially pumped mass spectrometer which samples ions diffusing to the walls of

a discharge tube. The mass spectrometer used is of the electric-quadrupole type and has been described in detail elsewhere.[11]

The discharge region is a glass cylinder with metal endplates. One endplate is a molybdenum electrode, while the other is made of Kovar metal and contains a small hole (60-μ diam and 40-μ length) through which the ions effuse into the mass spectrometer region.

The gas-handling system is analogous to that developed by Alpert.[12] The ultimate pressure was about 10^{-9} Torr following a system bakeout at 350 °C for a period of 24 to 36 h. All research-grade pure gases admitted to the discharge region were purified by means of the cataphoretic segregation method.[13] For the studies of plasmas produced in gas mixtures each gas was separately cataphoretically purified before mixing. The final cleaning of the discharge region was achieved by covering the discharge-tube wall with a molybdenum layer obtained from sputtering the discharge electrode. This cleaning process was continued until the impurity-ion signal was less than 0.5% of that of the dominant ion throughout the afterglow. This condition was necessary to achieve reproducibility of the data. The gas pressure was measured by a capacitance manometer which controlled a servo-operated valve to maintain a constant preset pressure in the discharge tube.

A block diagram of the measuring system is shown in Fig. 1. The discharge was produced by a high-voltage dc pulse applied between the discharge-tube electrodes. The ions passing through the quadrupole mass spectrometer are detected by a 14-stage ion multiplier. The resulting anode pulses, each due to a single ion, are amplified by a wide-band amplifier and those above a minimum pulse height are selected by a discriminator in order to reduce the background count rate. The pulses from the discriminator are then fed into a multichannel scaler. The afterglow is divided into 100 to 400 equal time intervals which have a minimum duration of 25 μsec. As the multichannel scaler advances from channel to channel, the number of pulses in the corresponding time intervals in the afterglow are recorded in the memory section. By accumulating the afterglow counts for a sufficient number of afterglow repetitions, a statistically significant number of counts can be recorded in each channel of the memory.

The time dependence of the intensity of the spectral lines studied was obtained using a 0.5-m Ebert light spectrometer. The photomultiplier had a S-20 photocathode and was cooled to reduce the dark count rate. The signal pulses resulting from single photons were detected and counted by the same technique as that used for ions.

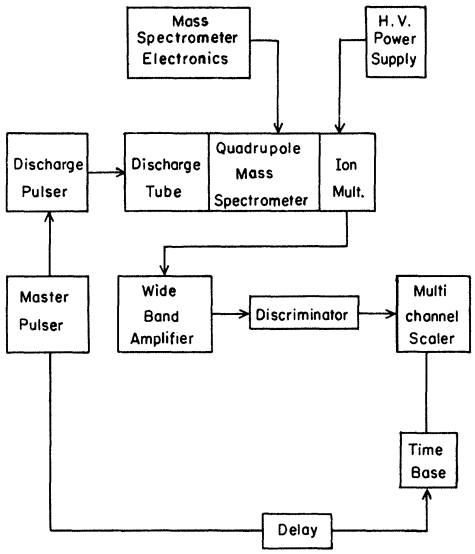

FIG. 1. Block diagram of measuring system.

III. RELEVANT AFTERGLOW PROCESSES

A. Ion-Density Time Dependence

The production of He^+ by mutual collisions between helium triplet metastable atoms

$$He^m(2^3S) + He^m(2^3S) \rightarrow He^+ + He + e \qquad (7)$$

has been shown to have a strong influence on the loss rates of charged particles in a mixture of 5×10^{-3}% neon in helium.[14] The reaction scheme (4)–(6) proposed for a decaying plasma in a helium-neon mixture by Oskam did not include this source of He^+. The present studies show that reaction (7) must be included. The production of He^+ influences the production of He_2^+ via the conversion reaction[15]

$$He^+ + 2He \rightarrow He_2^+ + He\ . \qquad (8)$$

Moreover, recent studies[16] as well as the present results show that an additional ion-conversion process

$$(HeNe)^+ + Ne \rightarrow Ne_2^+ + He \qquad (9)$$

needs to be added to the reaction scheme proposed by Oskam[3] to account for the measured decay rate of the $(HeNe)^+$ ion.[17]

In a helium-neon mixture, the volume destruction of the helium triplet is mainly due to three-body collisions with ground-state helium atoms and excitation transfer to neon atoms via the process

$$He^m(2^3S) + Ne \rightarrow Ne^* + He \qquad (10)$$

where Ne* is an unspecified excited state of neon. The diffusion coefficient and all the reaction rates

related to He^m (2^3S) have been measured by Phelps.[18–20]

In order to determine the time dependences of the number densities of the particles governing the decay properties of plasmas produced in helium-neon mixtures, the continuity equation of each active particle [$He(2^3S)$, He^+, He_2^+, Ne^+, ($HeNe^+$), and Ne_2^+] has to be solved. The solutions of the continuity equations are rather simple functions if the experimental conditions are chosen such that the loss processes are mainly diffusion towards the walls of the plasma container and/or linear volume-loss processes.

For particles which are not produced during the decay period the time dependence of the fundamental mode is a single exponential function. The relevant time constant is directly related to the loss processes of the particle. If the particle is also produced during the decay period, its time dependence is given by the sum of exponential functions having different time constants. One time constant is related to the loss processes of the particle, while the other(s) describe(s) the loss of the producing particles. The final decay of the particles will be determined by the largest time constant.

It can easily be shown that the time constant τ related to the loss processes of any particle is, for the assumptions made, given by

$$1/\tau = D/\Lambda^2 + \nu \quad . \tag{11}$$

Here, D is the diffusion coefficient of the particle (ambipolar diffusion coefficient for charged particles); Λ is the characteristic diffusion length of the plasma container, and ν is the volume-destruction frequency of the particle considered.

B. Time Dependence of the Light-Emission Intensity

The neon atomic-light emission can result from recombination of the atomic ion Ne^+ and/or from the dissociative recombination of the molecular neon ion Ne_2^+. Fortunately, the studies reported by Sauter *et al.*[8] in pure neon as well as the present measurements in the helium-neon mixtures show that the dissociative recombination of Ne_2^+ does not populate all energy levels of the neutral atom. This makes it possible to differentiate between the emission due to recombination of Ne^+ and that due to the recombination of Ne_2^+.

The intensity of spectral lines emitted as a consequence of a specific electron-ion recombination process will be proportional to the number of ions recombining with electrons per second by this process. This means that the time-dependent part of the light intensity $I_a(t)$ will be proportional to the product of the recombination coefficient α, the ion density $n_i(t)$, and the electron density, i.e.,

$$I_a(t) \propto \alpha n_i(t)\, n_e(t) \quad . \tag{12}$$

The radiative recombination process (2) will result in a spectral line emission intensity proportional to the product of the electron density and that of the ion involved. The same dependence will be found for spectral emission due to the dissociative recombination process (1). The collisional recombination process (3), however, will result in a different relation between the intensity of the produced spectral lines and the densities of the charged particles participating in the recombination process, since the recombination coefficient α for this process is a function of electron density. Previously, it was established that the recombination of He^+ and Ar^+ with electrons occurred via the collisional recombination process with a recombination coefficient proportional to the electron density.[9,10]

The functional relation between the spectral line emission intensity and the density of the charged particles responsible for the emission can easily be determined for experimental conditions such that the electron density is nearly equal to that of the recombining ion. Both the radiative recombination process and the dissociative recombination process will then produce emission having an intensity proportional to the square of the ion density. The collisional recombination process, however, will result in a different dependence of the emission intensity on the ion density. The intensity will be proportional to the ion density to the third power, provided the emission originates from electronic transitions with sufficiently small principal quantum numbers and provided the recombination coefficient is proportional to the electron density.

Another source of neon atomic-light emission during the decay period of plasmas produced in helium-neon mixtures is the excitation of neon by metastable helium atoms. The measurements of the decay rate of the intensity of the spectral lines involved as a function of gas pressure and gas mixture should give information about the identity of the metastable particle responsible for the emission.

IV. RESULTS AND DISCUSSION

A. Ion-Density Studies

The measured time dependence of the number density of the atomic helium ion under conditions such that the decay rate is governed by the He^+-loss processes yields an ambipolar diffusion coefficient $D_a P_o = 430 \pm 20$ cm^2 sec^{-1} Torr and a conversion frequency $\nu = (80 \pm 3) p_0^2$ sec^{-1} for process (8) at a gas temperature of 300 °K. The measurements were performed for neon concentrations in helium varying from 10^{-3}% to 0.1%. These values are in excellent agreement with values obtained in pure helium.[4,9,15]

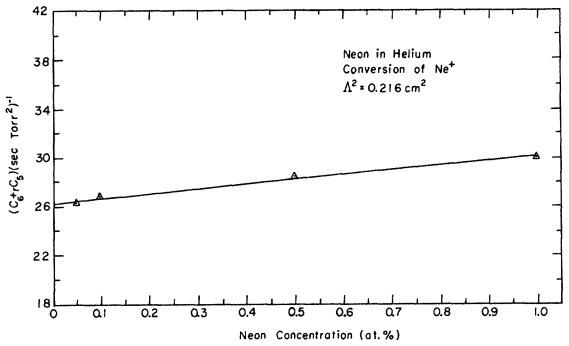

FIG. 2. Measured values of the conversion constants of the Ne^+ ion as a function of neon concentration at a gas temperature of 300 °K. The data points are obtained from the best straight-line fit to the measured p_0/τ values of Ne^+ versus p_0^3. Each plot consisted of at least 10 measured values for p_0/τ of Ne^+. In addition, each straight line was extrapolated to the accepted value of the diffusion coefficient of Ne^+ in helium.

Similarly, the measurements of the decay rate of the density of He_2^+ ions as a function of pressure in the helium-neon mixtures resulted in an ambipolar diffusion coefficient $D_a p_0 = 650 \pm 20$ $cm^2\,sec^{-1}$ Torr for He_2^+ in helium. The measured conversion frequency for process (4) was consistent with the rate coefficient of 1.5×10^{-10} $cm^3\,sec^{-1}$ at a gas temperature of 300 °K reported by Oskam.[3] The loss processes of He_2^+ could be studied only at low neon concentrations ($\leq 10^{-3}\%$) since for larger concentrations the time dependence of the He_2^+-ion density is controlled by the production of He^+ by process (7), which in turn produces He_2^+ by process (8).

The loss processes of Ne^+ can be measured for sufficiently high neon concentrations. Then the volume destruction of $He^m(2^3S)$ by collisions with neon atoms is efficient enough to neglect their influence on the time dependence of the Ne^+-ion density. The time constant related to the loss of Ne^+ ions is given by

$$p_0/\tau = D_a p_0/\Lambda^2 + (\nu_5 + \nu_6)p_0\ , \tag{13}$$

where ν_5 and ν_6 are the ion-conversion frequencies related to processes (5) and (6), respectively, and p_0 is the reduced helium pressure. According to process (5), $\nu_5 = c_5 r p_0^2$, where c_5 is a constant and $r \equiv p_0(\mathrm{Ne})/p_0(\mathrm{He})$. Process (6) implies that $\nu_6 = c_6 p_0^2$. Equation (13) can thus be written as

$$p_0/\tau = D_a p_0/\Lambda^2 + (c_6 + r c_5)p_0^3\,. \tag{14}$$

By plotting the measured value of p_0/τ for Ne^+ as a function of p_0^3, the values of $(c_6 + r c_5)$ as a function of r yield both c_5 and c_6. The value obtained at four different neon concentrations for the ambipolar diffusion coefficient of Ne^+ in helium is $D_a P_0 = 790 \pm 10$ $cm^2\,sec^{-1}$, which is in excellent agreement with values reported previously.[3] Figure 2 shows the measured value of $(c_6 + r c_5)$ as a function of neon concentration. From these data it follows that $\nu_{c5} = (370 \pm 40) r p_0^2$ sec^{-1} and $\nu_{c6} = (26 \pm 1) p_0^2\,sec^{-1}$.[21] These conversion frequencies correspond to rate constants of 3.0×10^{-31} $cm^6\,sec^{-1}$ and 2.1×10^{-32} $cm^6\,sec^{-1}$ for processes (5) and (6), respectively (gas temperature of 300 °K).

The present value of the rate coefficient for process (5) is seven times smaller and the measured rate coefficient for process (6) is 26 times larger than the values reported by Oskam.[3] The reason for this apparent discrepancy between the two results is that Oskam did not consider the production of He^+ by mutual collisions between helium triplet metastable atoms due to process (7). Thus the measured electron density decay corresponded to the time dependence of the He^+ density, which was governed by the production of He^+ by reaction (7). For this condition the time constant is given by $\frac{1}{2}\tau_m$, where τ_m refers to the loss of the metastable particles. Indeed, the time constant measured by Oskam[3] for the electron density decay is in very good agreement with the value of $\frac{1}{2}\tau_m$. Sauter *et al.*[8] reported also a value for the rate constant for process (6). Their value is about a factor of 3 smaller than the present value, which is believed to be more reliable. The previous measurements were performed at only one neon concentration, while using gases which had not been purified separately by the cataphoretic segregation process. Moreover, no consideration was given to the possible influence of process (7) on the decay properties of the plasma.

The p_0/τ values obtained for the time dependence of the $(HeNe)^+$-ion density are shown in Fig. 3 for a neon concentration of $10^{-3}\%$ in helium. The extrapolated $p_0 = 0$ intercept gives an ambipolar diffusion coefficient $D_a p_0 = 810 \pm 40$ $cm^2\,sec^{-1}$ Torr. The linear dependence of p_0/τ on p_0^2 is consistent with the conversion of $(HeNe)^+$ into Ne_2^+ by process (9). The relevant rate constant was found to be

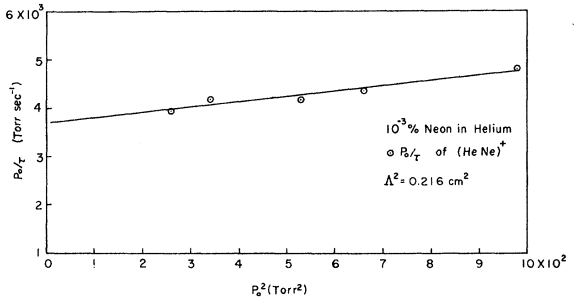

FIG. 3. Measured values of p_0/τ for $(HeNe)^+$ as a function of p_0^2 for a fixed neon concentration in helium of $10^{-3}\%$ (gas temperature 300 °K).

$(3\pm1)\times10^{-11}$ $cm^3\,sec^{-1}$ at a gas temperature of 300 °K. Bohme *et al.*[16] reported a value of 1.4×10^{-10} $cm^3\,sec^{-1}$ for this rate coefficient at a gas temperature of 200 °K.

Because of the large reaction rate for reaction (9), the number density of the $(HeNe)^+$ ions does not increase with neon concentration for concentrations larger than about 5×10^{-3}%. For these conditions the number density of Ne_2^+ ion increases with increasing neon concentration and the recombination coefficient attributed by Oskam[3] to the recombination of $(HeNe)^+$ with electrons was most probably the recombination coefficient of Ne_2^+ with electrons in the helium-neon mixture studied.

B. Light-Emission Studies

The time dependence of the intensities of various neon spectral lines in the visible spectrum from 3000 to 7000 Å has been measured during the decay period of plasmas produced in helium-neon mixtures. Three types of processes can be expected to be responsible during the decay period for the population of energy levels of neon atoms; i.e., (a) population by a recombination process involving Ne^+, (b) population by the dissociative recombination of Ne_2^+ with electrons, and (c) excitation of neon by metastable helium atoms.

No relation was found between the time dependence of the intensity of spectral lines originating

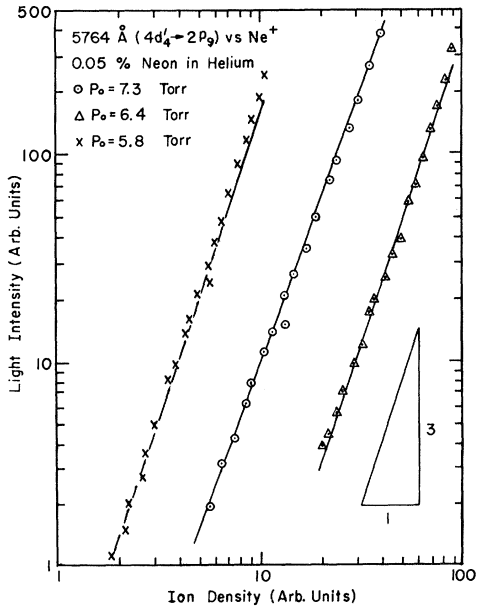

FIG. 4. The intensity of the 5764-Å line of neon versus the Ne^+-ion density during the decay period. Each data point corresponds to the values of the ion density and light intensity at a given time in the afterglow.

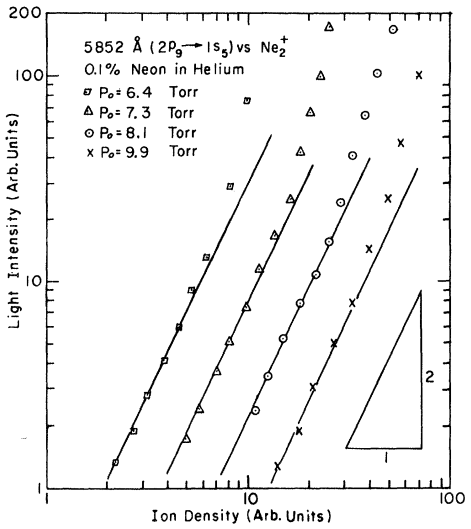

FIG. 5. The intensity of the 5852-Å line of neon versus Ne_2^+-ion density during the decay period obtained in the same way as the data curve in Fig. 4.

from the *d* levels of neon[22] and that of the density of the Ne_2^+ ions. However, the intensity of these spectral lines was found to be proportional to the third power of the Ne^+ density for experimental conditions during which the electron density was nearly equal to the Ne^+ density. Figure 4 shows this relation at different pressures in helium containing 0.05% neon for the 5764-Å ($4d_4' \rightarrow 2p_9$) spectral line of neon. The log-log plot is obtained by measuring the time dependences of the spectral-line intensity and that of the Ne^+ density under identical experimental conditions. Each measured point in Fig. 4 gives the line intensity and the Ne^+ density at the same time in the decay period.

The functional dependence between the emission intensity and the Ne^+ density establishes that the recombination process involving the atomic ion is, for the present experimental conditions, the collisional recombination process (3) with a recombination coefficient proportional to the electron density.

The dissociative recombination process (1) was found to contribute significantly to the population of the $2p$ and $3p$ levels of the neon atom. This is demonstrated in Fig. 5, which shows the relation measured between the 5852-Å ($2p_9 \rightarrow 1s_5$) neon line and the Ne_2^+-ion density. During the late afterglow period the Ne_2^+ ion was the dominant ion. The intensity of the spectral line is then clearly proportional to the square of the Ne_2^+ density. This confirms that the recombination process involving the Ne_2^+ ion is the dissociative recombination process (1). The deviation of the measured data from the straight solid line in Fig. 5 during the early decay

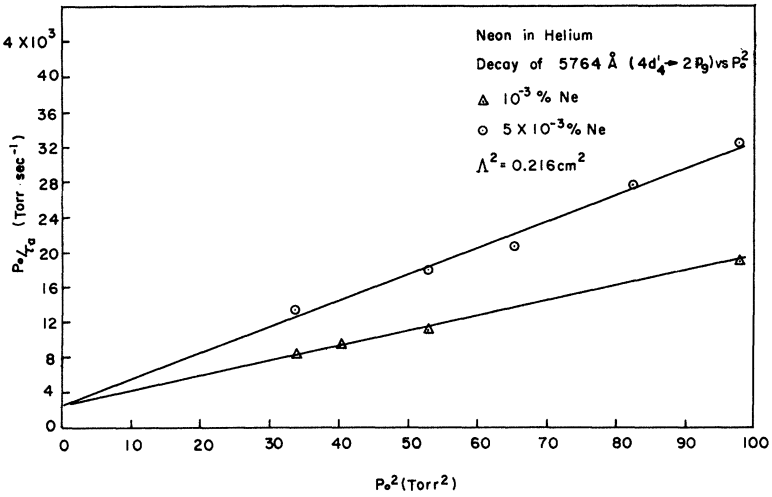

FIG. 6. Measured values of p_0/τ_a of the 5764-Å line of neon as a function of p_0^2 under conditions where the excitation transfer process from the helium singlet metastable is dominant in populating the $4d'_4$ level.

period is due to the presence of a considerable number of Ne^+ ions during this period and the contribution of the Ne^+ recombination process to the emission intensity. This phenomenon was also observed during studies of decaying plasmas produced in helium-argon mixtures.[10] It was not possible to obtain experimental conditions such that the spectral emission due to recombination of Ne_2^+ with electrons was small compared to the emission caused by other processes. This is a consequence of the very effective production of Ne_2^+ by process (9) in helium-neon mixtures.

For small neon concentration ($\leq 5\times10^{-3}\%$) in helium the time dependence of the emission intensity of spectral lines originating from the $4d$ levels of neon was not consistent with any electron-ion recombination process. The p_0/τ values, where τ is the time constant of the measured exponential decay of the spectral line intensity, for the 5764-Å ($4d'_4 \rightarrow 2p_9$) line are shown in Fig. 6 as a function of pressure for two neon concentrations. These values are consistent with the population of the $4d$ levels by the excitation transfer process

$$\mathrm{He}\ (2^1S) + \mathrm{Ne} \rightarrow \mathrm{He} + \mathrm{Ne}\ (4d)\ . \qquad (15)$$

The diffusion coefficient of He (2^1S) following from the present studies is within 20% of the value reported by Phelps.[19] Moreover, both the measured volume-destruction frequencies of He(2^1S) due to collision with helium atoms and with neon atoms agree within the experimental error with the values reported by Phelps[19] and by Benton *et al.*,[23] respectively. Thus, for small neon concentrations in helium, the $4d$ levels seem to be populated by the excitation transfer process (15).

Finally, the relation between the intensity of the 4650-Å emission band of He_2 and the electron and He_2^+-ion densities was measured for small neon concentrations ($\leq 5\times10^{-3}\%$). The same relation was measured as reported by Gerber *et al.*[9] for pure helium. These authors postulated as a consequence of their studies that the recombination of He_2^+ with electrons occurred by the reaction

$$He_2^+ + e + X \rightarrow He_2^* + X \rightarrow 2He + X + h\nu\ , \qquad (16)$$

where X could be a metastable helium molecule or an electron. If the third particle X is an electron, their results would imply that the recombination coefficient for collisional recombination process (16) is proportional to the square root of the electron density. The present studies seem to confirm this, since the measurements in the helium-neon mixture appear to exclude the metastable molecule as a candidate for X in the recombination process (16).[24]

V. CONCLUSIONS

The studies presented relate to the simultaneous measurement of the time dependences of the number density of He^+, He_2^+, Ne^+, $(HeNe^+)$, and Ne_2^+ ions and that of the intensity of spectral lines during the decay period of plasmas produced in helium-neon mixtures.

Improved rate constants were found for the conversion of the atomic neon ion into Ne_2^+ and $HeNe^+$ ions. These rate constants differ appreciably from previous measurements.[3,8] A new value was found for the two-body conversion of $(HeNe)^+$ ions into Ne_2^+ ions at a gas temperature of 300 °K.

Studies of the neon atomic emission have verified that the $2p$ and $3p$ levels are populated mainly by the dissociative recombination of the Ne_2^+ ion. The d levels, however, are populated by a process not related to the Ne_2^+ electron-ion recombination process. When excitation transfer is negligible, the d levels of the neon atom have been found for

the first time to be populated by the collisional recombination of Ne^+ with a recombination coefficient proportional to the electron density.

Excitation transfer to the $4d$ levels of the neon atom from the helium singlet metastable atom has been observed for small neon concentrations ($\leq 5 \times 10^{-3}$% neon in helium).

Finally, measurement of the relation between the He_2-band emission and the He_2^+-ion density in the helium-neon mixture indicates that the third body involved in the collisional recombination of He_2^+ is not a helium metastable molecule.

ACKNOWLEDGMENT

The discussions with other members of the study group on collision processes have been very helpful.

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*Work supported by the Air Force Research Laboratories, Office of Aerospace Research, under Contract No. AF 19(628)-4794 and by the National Science Foundation, under Grant No. GK-10395.

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