AN INVESTIGATION OF ELECTRON-ION RECOMBINATION IN INERT GASES

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Electron-ion recombination in inert gases is investigated at pressures up to 150 mm Hg. The plasma was produced either by a 600-keV electron beam or by a rapid direct discharge. Both an ultrahigh-frequency and an optical method were used to determine the decrease of electron density with time. It was found that the recombination rate depends on the conditions of plasma production, specifically on the degree of ionization and on the initial electron temperature. In lighter atomic gases the recombination rate was enhanced considerably when an electron beam was used to produce the plasma. Dissociative recombination with molecular ions can evidently be regarded as the principal mechanism responsible for the decrease of electron concentration.

1. INTRODUCTION

THE present article is concerned with an investigation of electron-ion recombination in inert gases. In all previously published experimental articles reporting measurements of recombination rates an identical technique was used to produce the plasma - by high-frequency breakdown of gas in a resonator, followed by measurement of the decay after the field was switched off. The experimental conditions, the degree of ionization, and the electron temperature, were very much alike in all instances.

In the present work we have investigated electron-ion recombination in a plasma produced by passing a strong high-energy electron beam through inert gases. The recombination rate was also investigated in plasmas produced by direct discharges.

APPARATUS

Our work was done with the ÉLIT pulsed electron accelerator[1], been developed at our Institute, under the following operating conditions: $E = 600$ keV, $I = 10$ A, and $\tau = 2 \times 10^{-6}$ sec. The electron beam 1 (Fig. 1) entered the inert-gas atmosphere through a 50-μ titanium foil; a collector 4 was located at the bottom of the hf resonator 3. The open cylindrical resonator was designed for E_{010} waves with $Q \approx 1500$ and resonant frequency $f \approx 10$ GHz. The output of the hf generator was about 10 mW. The resonator was first evacuated to 10^{-5} mm Hg; a very pure inert gas was then introduced. The gases were not subjected to any special purification process since their certified impurity content was below 0.01%.

The decay of electron concentration following the current pulse was measured in the conventional manner on the basis of the resonant frequency shift:[2]

$$\frac{\Delta f}{f} = \frac{1}{2} C_V \frac{n}{n_{cr}} \frac{\omega}{\omega^2 + \nu^2} \frac{V_p}{V_r}.$$

In our case the resonator and plasma volumes were equal, $V_p = V_r$, the collision frequency ν was smaller than the frequency of the probe signal, the coefficient C_V equalled the longitudinal form coefficient C_h, assuming a uniform radial distribution of the plasma,

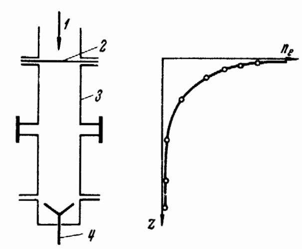

FIG. 1. Experimental scheme. 1 – electron beam, 2 – titanium foil, 3 – resonator, 4 – collector. Right-hand figure – electron concentration distribution along the resonator axis.

and n_{ct} is the critical electron concentration. The variation of plasma concentration along the resonator axis is approximated well by a quadratic parabola (Fig. 1), and we have the form coefficient $C_V \approx 0.2$.

Electron-ion recombination in a plasma produced by a direct discharge was investigated using the same hf apparatus in conjunction with the same system of gas evacuation and admission. The plasma was produced in a quartz tube that was fitted tightly to the resonator. The discharge parameters were $U = 20$ kV, $I = 50$ A, and $\tau = 10^{-7}$ sec. In this case $V_p = V_r$ and $C_V \approx 1$.

The variation of the electron density was measured independently by registering the recombination radiation of the plasma that was emitted as the plasma decayed following the current pulse. Recombination was investigated at gas pressures from 5 to 150 mm Hg.

RESULTS

Electron-ion recombination has been investigated quite frequently. Biondi and Brown were the first to develop and use an uhf technique to measure the rate at which the number of electrons decreases.[3,4] Recombination in gases, especially in inert gases, has been thoroughly investigated in[5-8].

The measurements of the electron-density decrease rate, on the basis of which the recombination coefficient is determined, do not enable us to learn what type of recombination is predominant. It is customarily assumed that when the gas pressure exceeds a few mm Hg the dominant process is dissociative recombination:

$$XY^+ + e \rightarrow XY^* \rightarrow X^* + Y^*$$

where X and Y with asterisks denote atoms that can remain in an excited state following the reaction. This

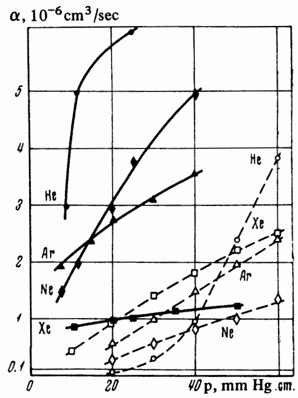

FIG. 2. Recombination coefficient in inert-gas plasmas produced by a direct discharge (dashed curves) and by an electron beam (solid curves).

in the case of plasmas produced by electron beams.

Figure 3a shows the pressure dependence of the recombination coefficient for He, Ne, Ar, and Xe in the case of a plasma produced by a direct discharge at higher pressures; Fig. 3b shows the analogous curves for a plasma produced by an electron beam. It is seen that here the recombination rates as functions of pressure do not differ so much as they do at lower pressures, although the dependence of the recombination coefficient on atomic number differs with the method of plasma production.

It must be noted that the mentioned values of the recombination coefficients in the electron beam case can differ from their true values because in calculating

	Recombination coefficient α, cm^3/sec				
	He	Ne	Ar	Kr	Xe
Published results	$1.7 \cdot 10^{-8}$ [3] $4 \cdot 10^{-9}$ [5]	$3.4 \cdot 10^{-7}$ [4] $2.3 \cdot 10^{-7}$ [5] $1.7 \cdot 10^{-7}$ [6]	$8.8 \cdot 10^{-7}$ [4] $6.7 \cdot 10^{-7}$ [5]	$1.2 \cdot 10^{-6}$ [5] $1.2 \cdot 10^{-6}$ [7]	$1.4 \cdot 10^{-6}$ [5]
Our data					
In the case of a discharge	$1 \cdot 10^{-7}$	$4 \cdot 10^{-7}$	$8 \cdot 10^{-7}$		$1.2 \cdot 10^{-6}$
In the case of an electron beam	$5{,}6 \cdot 10^{-6}$	$4 \cdot 10^{-6}$	$3 \cdot 10^{-6}$		$1 \cdot 10^{-6}$

is the principal type of recombination included in the measured rates of recombination in weakly ionized gases.

As already stated, in our experiments the plasma was produced in two different ways, and the initial state of the plasma following the current pulse was different in each instance. A direct discharge at $p < 40$ mm Hg produced a highly ionized gas with hot electrons. With increasing pressure ($p > 40$ mm Hg) the degree of ionization was considerably reduced and the electron temperature was relatively low.

When fast electrons traversed a gas the degree of ionization was low, the secondary electrons cooled down quickly, and the latter were at nearly room temperature very soon after the current pulse was terminated. A linear relation was found between the inverse electron density at different gas pressures and the time elapsed after the passage of a fast electron beam. It is thus shown that the decay of electron concentration results from recombination.

It was found that the method of producing a plasma has an essential influence on the recombination rate. Figure 2 shows the pressure dependence of the recombination coefficient α for He, Ne, Ar, and Xe when the plasma was produced by an electron beam (solid curves) and by a direct discharge (dashed curves). In the case of a direct discharge for $p < 40$ mm Hg the recombination coefficient increases with the atomic number of the gas. For $p > 40$ mm Hg the recombination rate increases steeply in He. In the case of the electron beam technique the recombination coefficient increases for almost all the gases, but the dependence on atomic number is altogether different (solid curves).

Earlier published results and our data at 25 mm Hg are given in the accompanying table. From Fig. 2 and the table we learn that the recombination coefficient for xenon does not depend on the initial conditions; for the other gases the coefficient is considerably larger

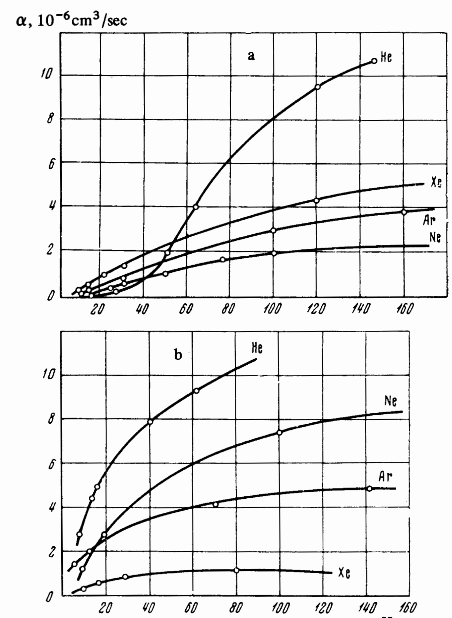

FIG. 3. Recombination coefficients for plasmas produced by (a) a direct discharge and (b) an electron beam.

the electron concentration the form coefficient C_V was taken to be identical for all gases. In actuality neither the specific ionization nor the elastic scattering cross section has identical values for He, Ne, Ar, and Xe. Consequently, the form coefficient will vary somewhat and will obviously be smaller for heavier gases. When the effect is taken into account the recombination coefficients in lighter gases can only increase.

The growth of secondary-electron concentration during the passage of a fast-electron beam through a gas and the decay of the concentration immediately following the current pulse were investigated by the

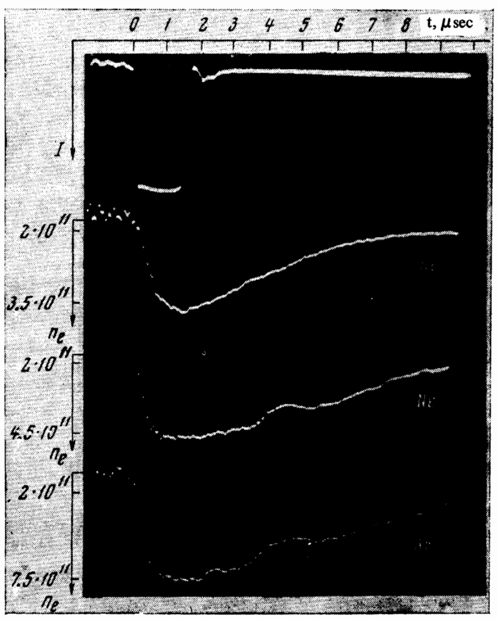

FIG. 4. Top oscillogram: fast-electron beam current. Lower oscillograms: electron concentrations in gasses at 25 mm Hg.

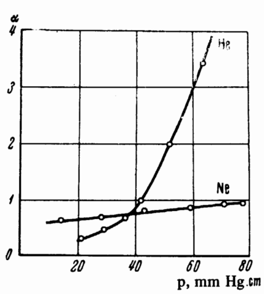

FIG. 5

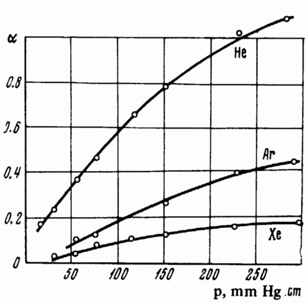

FIG. 6

FIG. 5. Recombination coefficients in He and Ne, determined from the afterglow intensity in plasmas produced by a direct discharge, as a function of pressure. (α in relative units).

FIG. 6. Recombination coefficients in He, Ar, and Xe, determined from the afterglow intensity in plasmas produced by an electron beam, as a function of pressure. (α in relative units).

wave method with transverse probing of the plasma[2] Figure 4 shows oscillograms of the electron concentration for three gases at 25 mm Hg. We observe that recombination in He proceeds more rapidly than in Ne and Ar immediately following the termination of the current pulse. The fluctuations on the oscillograms for the decay of electron concentration in Ne and Ar resulted from inadequate matching of the uhf system.

We investigated recombination on the basis of plasma radiation decay immediately following the current pulse. During this stage the radiation intensity accompanying dissociative recombination is proportional to the square of the electron density.[4] At the same time we should observe a linear time dependence of the inverse electron concentration $[1/n_e \sim 1/\sqrt{J} = f(t)$, where J is the intensity of recombination radiation]. In our experiments this dependence was ob-

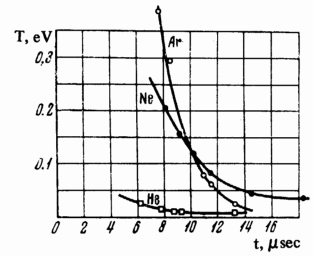

FIG. 7

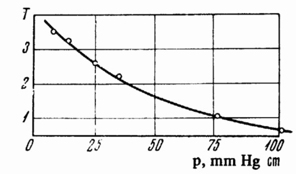

FIG. 8

FIG. 7. "Cooling" of secondary electrons following the passage of a fast-electron beam through a gas (p = 30 mm Hg).

FIG. 8. Decrease of electron temperature with increase of helium pressure following a direct discharge. The temperature (in relative units) was determined from the intensity ratio of the 4713-Å and 4921-Å lines [9].

served practically immediately following the pulse (after ~1 μsec). Figure 5 shows the pressure dependence of the recombination coefficient for He and Ne. Relative curves of the recombination coefficients for He, Ar, and Xe were obtained from the oscillograms of integral plasma radiation. Both Fig. 5 and Fig. 2 reveal a steep increase of the recombination rate in He for $p > 40$ mm Hg.

Recombination coefficients were measured similarly for plasmas produced by the passage of fast electrons; Fig. 6 represents the results in relative units. We observe good agreement between the results obtained by the optical and uhf resonator methods.

In the present work we measured the decrease rate of the mean energy possessed, immediately following a pulse, by secondary electrons produced through the passage of an electron beam. We measured the electron collision frequency according to the ratio between the variation of the reciprocal of resonator Q and the frequency shift:[2]

$$\omega \frac{\Delta(1/Q)}{\Delta\omega} = 2\frac{\nu}{\omega}.$$

In the electron beam case we observed low secondary-electron energy and a small degree of plasma ionization. The effective frequency of electron-ion collisions is equal in order of magnitude to the frequency of collisions between electrons and neutral atoms. Accordingly, the measured collision frequency can be used to calculate the mean velocity of secondary electrons as the plasma "temperature." Figure 7 shows the decrease of the secondary-electron mean velocity following the current pulse in He, Ne, and Ar at 30 mm Hg. The electrons are thermalized most rapidly in He, and a steeper gradient of the "cooling" rate is observed in Ar than in Ne. We know that the elastic interaction cross section in He at electron energies below one volt (up to the Ramsauer effect) is of the same order as in Ar and is considerably greater than in Ne.[8] It is also known that the mean energy of secondary electrons increases with the weight of the gas. These considerations enable us to interpret the results easily.

As has already been mentioned, the recombination rate in He is strongly dependent on pressure in a plasma produced by a direct discharge. It was therefore of interest to learn the pressure dependence of the plasma's electron temperature. Since the gas is not completely ionized in a discharge and the electron concentration is not very high, it is possible to determine the mean electron temperature by measuring the intensity ratio between He singlet and triplet lines.[9] Figure 8 shows the pressure dependence of the electron temperature (in relative units); it is observed that the plasma temperature is lowered considerably in the 50–70 mm Hg range.

DISCUSSION OF RESULTS

It is known[3-7] that high values of recombination coefficients are accounted for by the dissociative recombination mechanism of molecular ions formed in collisions between neutral and excited atoms. The production rates of molecular ions are known[8] for He and Ne: $(10.8 \pm 0.8) \times 10^{-32}$ cm^6/sec for He and $(5.8 \pm 0.8) \times 10^{-32}$ cm^6/sec for Ne. On the basis of scattering data the binding energies of the He_2^+ and Ne_2^+ ions are 2.6 eV and 0.3–1.0 eV, respectively. Similar investigations have yielded 0.055 eV as the upper limit of the Ar_2^+ binding energy, while that of Xe_2^+ is even lower.

When electrons recombine with molecular ions the reaction rate obviously depends on the formation rate of the molecular ions; this, in turn, depends on the state of the plasma, particularly the electron temperature (a decrease is observed as the temperature rises). Figure 8 shows that the electron temperature is lowered with increasing gas pressure. This effect results in an enhanced total recombination rate, as would be expected (Figs. 2, 3).

Similar considerations apply to the explanation advanced for the marked increase of the recombination coefficients in the case of plasmas produced by electron beams (Fig. 2), because the secondary electrons then possess considerably less energy than that which is represented by the electron temperature in the case of a direct discharge (Fig. 7).

It has already been noted that the recombination rate of Xe, for which the dissociation energy is comparable with room temperature, remains invariant for the two cases. On the other hand, the most rapid increase of the recombination rate is observed for He, which exhibits the highest dissociation energy of the molecular ion, the molecular ion He_2^+ being produced more rapidly than ions of the other gases.

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[1] E. A. Abramyan and S. B. Vasserman, Atomnaya ênergiya 23, 44 (1967) [Sov. Atomic Energy 23, 709 (1967)].

[2] V. E. Golant, Sverkhvysokochastotnye metody is sledovaniya plazmy (Ultrahigh-frequency Methods of Investigating Plasmas), Nauka, 1968.

[3] M. A. Biondi and S. C. Brown, Phys. Rev. 75, 1700 (1949) and 76, 1697 (1949).

[4] M. A. Biondi, Phys. Rev. 129, 1181 (1963).

[5] H. J. Oskam and V. R. Mittelstadt, Phys. Rev. 132, 1445 (1963).

[6] W. H. Kasner, Phys. Rev. 167, 148 (1968).

[7] V. B. Brodskiĭ and S. E. Zagik, in Nizkotemperaturnaya plasma (Low-temperature Plasma), Mir, 1967, p. 55.

[8] E. W. McDaniel, Collision Phenomena in Ionized Gases, John Wiley and Sons, New York, 1946 (Russ. transl., Mir, 1967).

[9] R. H. Huddlestone and S. L. Leonard (Ed.) Plasma Diagnostic Techniques, Academic Press, New York, 1965 (Russ. transl., Mir, 1967).

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