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DECAY OF A PLASMA PRODUCED BY A PULSED BEAM OF ACCELERATED ELECTRONS IN AN He—Ne MIXTURE AT HIGH PRESSURE

Yu. D. Korolev and A. P. Khuzeev

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Increased interest has recently been shown in the low-temperature plasma produced by the action of electron beams on dense gases. This interest is due to the possibility of investigating plasma-chemical reactions in highly nonequilibrium conditions [1] and also to the prospect of introducing new methods of pumping gas lasers: by recombination [2], charge transfer [3], the formation of complex compounds [4, 5], etc. The plasma in question is characterized by high rates of reactions involving charged and neutral particles, which largely accounts for the difficulty of investigating such a plasma experimentally. For instance, problems of determining the kind of ions predominating in the plasma, the mechanism of recombination decay, the nature of the luminescence on individual spectral transitions, etc., become nontrivial. In this paper we investigate the decay of a plasma in neon and in an He—Ne mixture at high gas pressure.

§1. Experimental method

The kinetics of recombination processes were investigated by the photoelectric method of recording the spectrum and measuring the decay of electron density in the plasma. The experimental apparatus is shown schematically in Fig. 1. A beam of fast electrons from the accelerator 1 was injected into the gas cuvette through a window sealed with titanium foil 20 μ thick. The beam-current density was 25 A/cm^2, the length of the current pulse at the base was $1.5 \cdot 10^{-7}$ sec, and the electron energy was 200 keV. The time for recombination decay of the plasma greatly exceeded the duration of the beam current and, hence, the electron beam, ionizing the gas in the cuvette, determined the initial density of electrons and ions (n_0).

The photoelectric system for recording of the emission spectrum consisted of an MDR-3 grating monochromator 4, an FÉU-38 photomultiplier 3, and an S8-2 dual-beam recording oscilloscope 2. The resolving time of the spectrum recording channel was 25 nsec.

An electric field was applied to the plasma by connecting a capacitor bank 6, charged from a voltage supply 5, to the anode 7 of the gas cuvette. The anode was a disk 4 cm in diameter. The cathode 8 was a brass grid with 0.2 × 0.2-mm^2 mesh. The interelectrode distance was 4.5 cm. There were two reasons for applying a voltage to the plasma: for stabilization of the electron temperature and for determination of the variation of the electron concentration with time from the current across the gap. In the absence of the field the electron temperature is determined by the balance between the processes leading to heating and to cooling of the electrons [6-8]. The electrons are cooled by elastic and inelastic collisions with gas atoms. Fast electrons can be produced by ionization of the gas by the beam electrons, deexcitation of metastables by electron impact, production of electrons in metastable—metastable collisions, and so on. In the considered conditions (electron concentration $n > 10^{14}$ cm^{-3}, gas pressure ~1 atm) the excess of electron temperature over the gas

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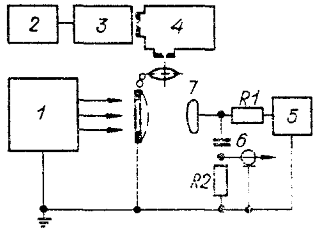

Fig. 1

temperature will be determined by the density of metastables, which varies with time. This introduces indeterminacy into the value of T_e. When an electric field is applied the electron temperature increases so much that the effect of its variation due to reactions involving metastables becomes insignificant. On the other hand, excessive increase in T_e can also lead to incorrect measurements. The decay of the plasma has to be investigated in conditions where the electron concentration exceeds the equilibrium concentration. As an example, we note that when $T_e = 0.6$ eV and p = 1 atm the equilibrium electron density in helium is 10^{12} cm^{-3}, which is much lower than that attained in the experiment.

The electron concentration in relation to time was determined from oscillograms of the current across the gap

$$n(t) = I(t)/ev_{-}S,$$

where e is the electron charge; v_{-} is the drift velocity; and S is the area of the discharge column.

When this method is used the ratio of the field to the pressure on the discharge column (E/p) and the dependence of the drift velocity on E/p have to be known. We measured the distribution of field strength in the plasma by means of two probes at floating potential. The distance between the probes was 1 cm. The probe nearer to the cathode was at a distance of 1 cm from it. The potential difference between the probes, and also between the probes and the cathode, did not change during decay of the plasma. Hence, the cathode potential drop was constant: U_c = 180–200 V. In the treatment of the experimental results we took the ratio of the field on the plasma to the pressure to be $E/p = (U_0 - U_c)/pd$, where U_0 is the voltage applied to the gap, and d is the gap length. The electron-drift velocity in the region of E/p = 0.05–0.8 V/(cm•mm Hg) for pure helium was taken from Table II.3.1 in [9], and that for pure neon was taken from [10]. In the case of helium with neon added we used the same relation $v_{-}(E/p)$ as for pure helium.

We used high-purity helium and neon in the experiments. We also subjected the gases to additional adsorption purification by passage through activated charcoal at liquid-nitrogen temperature. Before being filled with gas the cuvette was evacuated to a pressure of 10^{-6} mm Hg by a diffusion pump with freezing of the oil vapor. After repeated injection of the electron beam into the gas cuvette we observed the release of impurities from the cuvette walls and the insulator. This was revealed by the appearance of additional lines (e.g., of the nitrogen ion N_2^+) in the luminescence spectrum and an increase in the plasma decay rate. Hence, after two or three injections of the beam into the gas the cuvette was evacuated and filled with a new batch of gas.

§2. Discussion of Results

The parameter characterizing the rate of decay of electron density in the plasma is the effective recombination coefficient β. Figure 2 shows the relation β(E/p) at total pressure p = 800 mm Hg in an He + 0.5% Ne mixture (curve 1) and in pure neon (curve 2) with initial electron density $n_0 = 2 \cdot 10^{14}$ and 10^{15} cm^{-3}, respectively. In all cases the reduction of electron concentration was represented by the relation $1/n(t) = 1/n_0 + \beta t$. Oscillograms in the range n_0 to $(0.1–0.15)n_0$ were analyzed. The recombination coefficient in an He–Ne mixture was higher than in pure helium at the same E/p.

We found that with successive increase in the neon content of the mixture the value of β increased steadily at first and then became constant at 0.2–2% neon. The shape of the oscillograms of the current through the plasma accordingly remained the same.

Figure 3 shows plots of the recombination coefficient against gas pressure in an He + 0.5% mixture (curves 1–3) and in pure neon (curves 4, 5). The ratio of the field to pressure

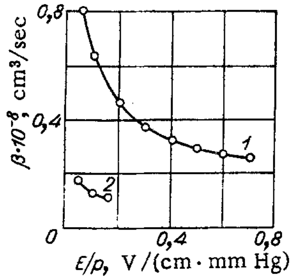

Fig. 2

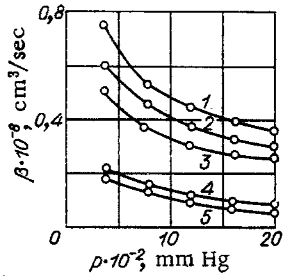

Fig. 3

was E/p = 0.15, 0.2, 0.3, 0.05, and 0.1 V (cm•mm Hg), respectively. Reduction of β with pressure increase was a little faster in neon than in an He—Ne mixture.

Experimental data for recombination of a neon plasma at high gas pressure indicate a predominance of Ne_2^+ ions and dissociative recombination of these ions [1]. A relation between β and pressure was also found in [11]. According to existing ideas of the mechanism of dissociative recombination, excited neon atoms will be formed mainly in the group of 2p states. Observation of the luminescence of the spectral lines on transitions in neon from the group of 2p levels to the 1s group showed that the $2p_1$ level is mainly populated by recombination, and there is intense luminescence on transitions with wavelengths λ = 585.2 ($2p_1$—$1s_2$) and 540.0 nm ($2p_1$—$1s_4$).

From pure helium excited by an electron beam we obtained a strong recombination spectrum of the He_2 molecule, but with increase in neon concentration the intensity of the molecular bands decreased, while the intensity of the neon lines (585.2 and 540.0 nm) increased. At a neon concentration of 0.2-0.4% the spectrum of helium molecules practically disappeared. Of the 30 resolved transitions from 2p levels to 1s levels the mentioned lines had an intensity an order higher than the rest. We note that the $2p_1$—$1s_4$ transition has the smallest oscillator strength and, hence, observation of the luminescence on this transition indicates high population of the $2p_1$ level. Figure 4 shows the time course of the luminescence of the 585.2-nm line in an He + 0.5% Ne mixture for E/p = 0, 0.35, and 0.7 V/(cm•mm Hg) (curves 1-3, respectively). The shape of the luminescence and the nature of the effect of the electric field indicate the recombination nature of population of the $2p_1$ level. The decay of relative intensity $\varepsilon/\varepsilon_0$ is satisfactorily described by the relation $\sqrt{\varepsilon_0/\varepsilon(t)} = 1 + \beta n_0 t$. The recombination coefficient determined from this equation agrees with that measured from the decay of electron density. This fact, and also the similar nature of the relation β(p) in an He—Ne mixture and in pure neon, enables us to confirm that the decay of a helium plasma containing small amounts of neon in the considered experimental conditions is due to dissociative recombination of Ne_2^+ ions. Figure 5 shows a plot of the recombination coefficient in neon (curve 2) and in an He—Ne mixture (curve 1) against electron temperature, converted from the data in Fig. 2 from the relations $T_e(E/p)$ taken from [12]. The absolute values of the recombination coefficients at the same electron temperatures were close to one another, which also indicates the identity of the recombination mechanism in pure neon and in an He—Ne mixture. The temperature dependence in both cases has the form $T_e^{-0.43}$. Exact agreement of the recombination coefficients for Ne_2^+ ions in a mixture containing helium as a buffer gas and in pure neon can hardly be expected. This is due to the fact that the coefficient of dissociative recombination depends on the distribution of the population of vibrational states of molecular ions [1]. This distribution will obviously be different in different gases. The relation β(p) may also be due to the more rapid relaxation of the vibrational energy of the Ne_2^+ ion with increase in pressure.

To distinguish the channel of formation of molecular-neon ions we analyze the characteristic times of the reactions predominating in the plasma. The most rapid reactions, which may implicate helium ions forming by the passage of the beam of accelerated electrons through the gas, are the conversion of atomic ions to molecular ions (the constant of the process $k_1 \approx 10^{-31}$ cm^6•sec^{-1} [1], characteristic time $\tau_1 = 1/k_1[He]^2 = 10^{-8}$ sec) and charge transfer from the He_2^+ ion to neon atoms ($k_2 = 1.4 \cdot 10^{-10}$ cm^3•sec^{-1} [13], $\tau_2 = 1/k_2[Ne] = 10^{-7}$ sec).

These processes take place in accordance with the scheme

$$He^+ + 2He \rightarrow He_2^+ + He; \tag{2.1}$$

$$He_2^+ + Ne \rightarrow Ne^+ + 2He. \tag{2.2}$$

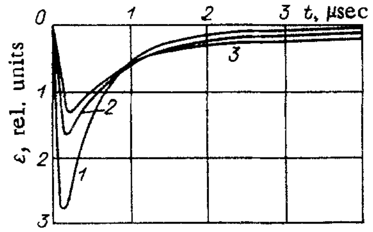

Fig. 4

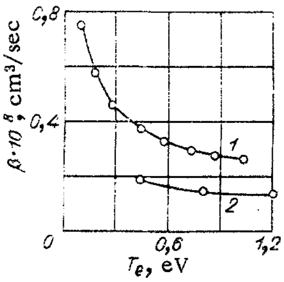

Fig. 5

Since the density of neutral particles is high, there are reactions leading to the formation of complex ions

$$Ne^+ + 2He \rightarrow (HeNe)^+ + He; \tag{2.3}$$

$$Ne^+ + Ne + He \rightarrow Ne_2^+ + He; \tag{2.4}$$

$$Ne^+ + He + Ne \rightarrow (HeNe)^+ + Ne. \tag{2.5}$$

The constants of the processes (2.3) and (2.4) [1] are $k_3 = 0.2 \cdot 10^{-31}$ $cm^6 \cdot sec^{-1}$ and $k_4 = 3 \cdot 10^{-31}$ $cm^6 \cdot sec^{-1}$, respectively. Since the complex $(HeNe)^+$ ions are characterized by a weak bond (dissociation energy ~0.034 eV [14]), their effect on plasma decay, as estimates showed, could be neglected. Thus, molecular Ne_2^+ ions, which are responsible for the recombination decay of an He–Ne plasma, are formed as a result of the chain of reactions (2.1), (2.2), and (2.4).

The luminescence on transitions whose upper level is colonized by dissociative recombination of Ne_2^+ ions is the most intense in the spectrum of an He–Ne plasma produced by an electron beam. Since the generally accepted mechanism of luminescence of neon in He–Ne mixtures at low pressures and low electron concentrations involves the transfer of exictation from helium metastables to neon [15], this process must be analyzed in application to the considered conditions.

It is known that when helium is ionized by fast electrons the electron energy expended on the production of charged particles is approximately equal to the energy expended on excitation. This means that after the cessation of action of the beam the density of atomic-helium metastables $n_M \approx n_0$. When $n_0 \approx 10^{14}$ cm^{-3} the transition of He (2^1S) metastables to He (2^3S) metastables in quenching collisions with electrons occurs in characteristic time ~10^{-7} sec [1]; i.e., in excitation transfer reactions only the 2^3S metastable needs to be taken into account. The absence of emission on the 632.8-nm transition confirms this statement. The transfer of excitation from the 2^3S metastable to the 2s level can be detected experimentally by measuring the emission of 2p–1s transitions. The rate constant of excitation transfer from He (2^3S) ($k = 4 \cdot 10^{-12}$ $cm^3 \cdot sec^{-1}$) is an order less than for the 2^1S metastable [15]. Comparing the rate of pumping of the $2p_1$ level by recombination ($\beta n_0^2 \approx 3 \cdot 10^{20}$ $cm^{-3} \cdot sec^{-1}$) and by excitation transfer ($[He_M][Ne]k \approx 10^{20}$ $cm^{-3} \cdot sec^{-1}$) we obtain commensurable values. In the experiments, however, the intensity of the recombination emission greatly exceeded the intensity on the transitions 703.5, 640.2, 724.5 nm, etc. This is probably due, first, to the selective population of the $2p_1$ level, and second, to subsidiary processes leading to the destruction of metastables. Such processes are Penning ionization by metastable–metastable collisions ($k = 1.5 \cdot 10^{-9}$ $cm^3 \cdot sec^{-1}$, $\tau = 3 \cdot 10^{-6}$ sec [16]), quenching of metastables by collisions with electrons ($k \geq 0.5 \cdot 10^{-10}$ $cm^3 \cdot sec^{-1}$, $\tau \leq 10^{-6}$ sec [16]), and conversion of the atomic 2^3S metastables to molecular $2^3\Sigma$ ($k = 0.3 \cdot 10^{-33}$ $cm^6 \cdot sec^{-1}$, $\tau = 5 \cdot 10^{-6}$ sec [1]).

In conclusion, we note that the possibility of using dissociative-recombination processes for the obtention of inversion in lasers has been discussed already [15]. The selective excitation of the neon $2p_1$ level, discovered in our investigation, may lead in particular conditions to inversion on the $2p_1$–$1s_4$ and $2p_1$–$1s_2$ transitions. Favorable factors for this are the high concentration of a plasma produced by an electron beam and the high recombination coefficient, which can ensure adequate pumping levels. For instance, when $n = 5 \cdot 10^{14}$ cm^{-3}, $\beta = 10^{-8}$ $cm^3 \cdot sec^{-1}$ (an estimate of the amplification coefficient of the stimulated radiation in self-bounded conditions) gives $\varkappa \approx 1.5 \cdot 10^{-3}$ cm^{-1}.

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