EXPERIMENTAL INVESTIGATION OF THE DESTRUCTION OF METASTABLE ATOMS OF HELIUM IN A PLASMA AT LOW TEMPERATURES

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The kinetics of volume destruction of metastable atoms 2^3S He at high initial concentrations (10^{13}–10^{14} cm^{-3}) is investigated experimentally at temperatures from 6 to 300°K and pressures from 3 to 80 mm Hg. The constant $A_2 = 0.35$ cm^3 sec^{-1} $(mm\ Hg)^{-2}$ of the process of the destruction of the metastable atom in collision with two normal atoms is measured at room temperature. The constant k of the process of the collision of metastable atoms is measured at low temperatures and the value $k \sim T^{1/6}$ is obtained. The conditions under which the process of superelastic collision with a slow electron predominates in the destruction of the 2^3S state of He were determined. The constant of this process is calculated and found to be $k_e = 4.5 \times 10^{-9}$ cm^3 sec^{-1}. A Study of the kinetics has made it possible to determine the rate of impact-radiation recombination and the electron temperature in the plasma. The main mechanisms of heating of electrons in a plasma at low temperatures are discussed.

1. INTRODUCTION

AMONG all the excited states of the helium atom, the metastable state 2^3S has the longest lifetime. With decreasing temperature of the atoms, the rates of any processes whereby this state is destroyed decrease and its lifetime increases. Consequently, the metastable atoms 2^3S He can accumulate in a discharge and their concentration reaches values higher than 10^{13} cm^{-3} at a total density on the order of 10^{17} cm^{-3}. At high concentrations of the metastable atoms, their collisions with one another, with electrons, and with unexcited helium atoms become important. In these collisions, the excited state becomes deactivated and an appreciable fraction of the excitation energy is released (E_{exc} = 19.8 eV), and this exerts a strong influence on the degree of ionization and on the plasma temperature.

We report in this paper the results of an experimental investigation of volume destruction of the 2^3S state in a decaying plasma at temperatures 300–6°K and pressures 3–80 mm Hg. The kinetics of pair collisions of metastable atoms of helium at low temperatures is described in a brief communication.[1]

2. GENERAL DESCRIPTION OF THE PROCESSES OF DESTRUCTION OF METASTABLE ATOMS

The processes of destruction of metastable atoms can be subdivided into surface and volume processes. Processes of surface destruction occur on the walls of the discharge vessel and are due to the diffusion of the atoms. The diffusion of metastable atoms of helium in their own gas plays an important role at low pressures and high temperatures. The rate of diffusion decay of the 2^3S state of He was investigated earlier in a wide temperature range, from 300 to 4°K, and these results are summarized in the review.[2]

Among the different volume processes of destruction, one of the most important is the collision of two metastable atoms of helium. Their interaction with each other is determined by the van der Waals attraction forces and is characterized by a large constant of this attraction C. According to calculations by Dalgarno et al.,[3] $C = 3.29 \times 10^3$ at.un. $= 3.16 \times 10^{-57}$ erg-cm^6. When two metastable atoms collide, mutual capture takes place, with formation of an unstable quasimolecule of helium He_2^* (the asterisk denotes the excited state). Owing to the excessive reserve of excitation energy, this quasimolecule decays immediately in accordance with an autoionization scheme (of the Penning-reaction type). The products of the decay are a "hot" electron, which carries away an appreciable fraction of the excitation energy, the atomic ion He^+, and an unexcited helium atom in the state 1^1S. The reaction scheme can be written in the form

$$He^*(2^3S) + He^*(2^3S) \rightarrow He(1^1S) + He^+ + e. \quad (1)$$

One cannot exclude the possibility that the decay products may be not the atomic helium, but the molecular helium ion H_2^+, although the rate of destruction of the state 2^3S does not depend on the type of the ion in the scheme (1). Experimental data on the change of the concentration of atomic He^+ ions in a decaying plasma[4-6] offer evidence favoring more readily the formation of atomic and not molecular ions in the reaction (1). In addition, in Beatty and Patterson's drift-tube experiments[7] it was shown that the number of He_2^+ ions can be described satisfactorily by conversion of the atomic ions into molecular ones in the process

$$He^+ + 2He \rightarrow He_2^+ + He. \quad (2)$$

It can therefore be assumed that the formation of the atomic ion is more probable as a result of pair interaction of two metastable atoms.

Another inelastic process in which the metastable state is destroyed is the collision of an excited 2^3S atom with a slow electron. The metastable atom goes over into the ground state, and the excitation energy is carried away by the "hot" electron. The scheme of the reaction is in this case

$$He^*(2^3S) + e \rightarrow He(1^1S) + e. \quad (3)$$

Decay in accordance with scheme (3) is a process that is the inverse of the direct excitation process; the reaction (3) is sometimes called also superelastic collision. Unlike reaction (1), the rate of which is proportional to the square of the concentration of the metastable atoms M, the rate of destruction of the state 2^3S in the process (3) is proportional to the product of the concentrations of the electrons and of the metastable atoms.

Finally, at room temperatures (and above) and relatively high pressures (above 10 mm Hg), an important role is played by the triple-collision process, in which a metastable and two unexcited helium atoms take part:

$$He^*(2^3S) + 2He(1^1S) \rightarrow He_2^*(2^3\Sigma_u^+) + He(1^1S). \quad (4)$$

The metastable He_2^* molecule is produced as a result of this reaction. The rate of destruction in the process (4) is obviously proportional to the square of the gas pressure and depends linearly on the concentration M. The process (4) has an activation energy on the order of 0.08 eV, due to the existence of a maximum of the long-range repulsion potential of the interaction of the metastable and unexcited helium atoms (see, for example [2]). Owing to this maximum, the rate of decay in the reaction (4) decreases exponentially with decreasing temperature, and in the region of low temperatures $T \lesssim 150-100°K$ it turns out to be negligibly small compared with the rates of the processes (1) and (3).

Thus, the equation describing the change of the concentration of the metastable atoms in all the volume-destruction reactions listed above can be written in the form

$$-\frac{\partial M}{\partial t} = kM^2 + k_e nM + A_2 P^2 M. \quad (5)$$

Here k is the rate constant of the process (1), k_e is the constant of the process (3), n is the electron concentration, and A_2 characterizes the process (4).

In the experimental investigation of the kinetics of the decay of the helium plasma, the triple-collision process (4) can easily be separated from the other two processes by means of its pressure dependence. The constant A_0 was measured at room temperature by several groups of workers.[8-10] The most complicated matter is the separation of the first two processes in (5), since both, as a rule, appear under identical conditions. The situation is complicated also by the fact that the electron concentration frequently turns out to be comparable with the concentration of the metastable atoms (or may even exceed it), and little is known concerning the value of k_e. Direct measurement of the constant k_e is extremely difficult, since it is necessary to determine simultaneously the local absolute values of the concentrations n and M. We know likewise of no reliable theoretical calculations of the constant k_e. One can expect the constant k_e to be of the same order of magnitude as k. The process of paired interaction (1) is separated from (3) if the following condition is satisfied:

$$kM > k_e n, \quad (6)$$

which reduces to the fact that the number of metastable atoms in the plasma should be much larger than the number of electrons. When this inequality is satisfied, the kinetics of the decrease of the density of the metastable atoms in the afterglow is described by the first term of Eq. (5), and obeys the hyperbolic law

$$M^{-1}(t) = M_0^{-1} + kt. \quad (7)$$

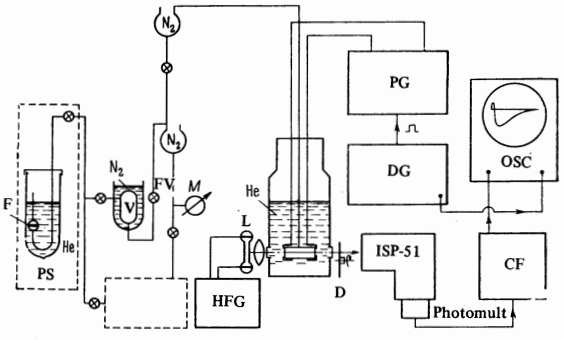

FIG. 1. Experimental setup: PS–helium purifying system, F–filter, V–vessel for gaseous helium, FV–fine inlet valve, M–manometer, PG–pulse generator, DG–delayed-pulse generator, CF–cathode follower, L–helium lamp, D–disk modulator.

Experimental data on the value of k at room temperature were obtained by Phelps and Molnar;[8] measurements at T = 77°K were performed by Ludlum et al.[10], and also in [11]. In a wide temperature interval from 5 to 120°K, the values of the constant k are given in our paper.[1] The value of k changes from 2×10^{-9} cm^3 sec^{-1} at 300°K to 1.1×10^{-9} cm^3 sec^{-1} at 6--10°K.

3. EXPERIMENTAL SETUP AND MEASUREMENT PROCEDURE

The kinetics of destruction of metastable atoms in a decaying helium plasma was investigated by a spectroscopic method in the temperature interval from 300 to 6°K. The experimental setup is shown in Fig. 1. The measured quantity is the intensity of the 3889 Å resonance line (3^3P-2^3S) passing through a discharge vessel with the plasma. The vessels were cylindrical in form and were placed inside an optical cryostat. The experiments were performed on two glass tubes with dimensions (in mm) $d_1 = 8$, $l_1 = 35$ and $d_2 = 22$, $l_2 = 39$; the dimensions of the tubes were limited by the cryostat. The diffusion lengths λ of these vessels correspond to $\lambda_1^{-2} = 37$ cm^{-2} and $\lambda_2^{-2} = 5.4$ cm^{-2}. The discharge tubes were connected to a glass purification and filling system, which was subject to careful prior conditioning. The preliminary vacuum was not worse than 10^{-7} mm Hg. The pure gas entering the discharge vessel was tapped from the vapor over the liquid helium and p passed through a U-tube with a micron filter immersed in liquid helium. The gas was then fed into the flask V, which was cooled with liquid nitrogen, and from there to a "fine inlet" valve FV into the discharge vessel.

The plasma was produced by a high-frequency discharge (f ≈ 18 MHz). The shapes of the external electrodes were chosen to ensure homogeneity of glow in the investigated pressure range from 1 to 80 mm Hg. The pressures are given throughout in units reduced to room temperature, $P = P_T 293/T$, and characterize the gas density. The HF generator produced rectangular voltage pulses of duration τ from 2 to 20 μsec and at voltages u from 0.4 to 2 kV. Special measurements were made of the plasma resistance by simulating it with an equivalent resistor and a capacitor. A high-

frequency detector was used to measure the HF voltage u directly across the plasma, and the beat method was used to measure the change of the pulse carrier frequency relative to vacuum when the plasma was excited. The characteristic currents through the plasma were i ~ 0.2–0.5 A. The plasma resistance increased with decreasing gas temperature, from several dozen ohms to several hundred ohms. The energy released per pulse was approximately 3.3×10^{-4} J when liquid helium was used for cooling and at P = 10 mm Hg, leading to heating of the gas by 3–4°K. The temperature of the atoms in the plasma was also determined from the acoustic oscillations accompanying the excitation at low temperatures.[12] By measuring the frequency ω of the fundamental mode of the standing acoustic oscillations in a cylinder of radius r, it was possible to determine the speed of the sound s and the temperature T by means of the formulas

$$s = \frac{\omega r}{3.83}, \quad T = \left(\frac{280\, r}{\tau_s}\right)^2, \tag{8}$$

where 3.83 is the first root of the Bessel function $J_1(x)$, $\tau_s = 2\pi/\omega$ is the period of the acoustic oscillations in μsec. For a bulb with r = 1.1 cm at a wall temperature 4°K, the characteristic periods τ_s were 130–100 μsec, corresponding to an atom temperature T = 5–10°K. The temperature of the walls of the discharge vessel were measured with a capacitive pickup.

The external radiation source was a capillary lamp L filled with helium. The temperature of the radiating atoms T_r was determined by interferometry, using the Doppler broadening of the lines. In most measurements we used a lamp excitation regime corresponding to T_r = 1000°K. The light intensity J passing from the external source through the plasma was measured with the aid of an ISP-21 spectrograph and a ChÉU 64 photomultiplier with a cathode follower CF on an S1-8 or S1-16 oscilloscope. The generator DG made it possible to trigger the pulse generator PG and the oscilloscope synchronously at any instant of time after the end of the pulse. This made it possible to measure the intensity of the transmitted signal with increasing gain and with good time resolution. Figure 2 shows by way of illustration one of the oscillograms of the radiation intensity J after the end of a pulse. In measurements of J, we took into account the additional intrinsic radiation of the plasma at the initial instants of the afterglow and, where necessary, suitable corrections were introduced in the absorption $\Delta J = J_0 - J$. Figure 3 shows the time variation of the measured relative absorption curves $\Delta J/J_0$ for three different temperatures.

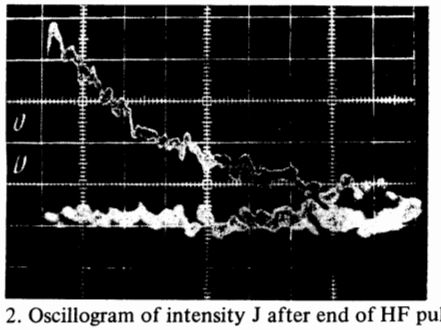

FIG. 2. Oscillogram of intensity J after end of HF pulse.

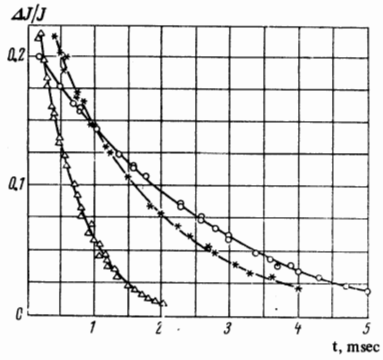

FIG. 3. Dependence of the relative integral absorption on the time for the 3889Å line. P = 5 mm Hg, $\lambda^{-2} = 5.4$ cm^{-2}, Δ–at room temperature, *–after cooling with liquid nitrogen, O–after cooling with liquid helium.

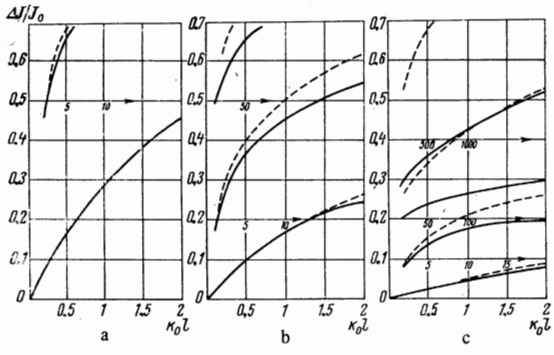

FIG. 4. Calculated plots of A against $k_0 l$ for different temperatures of the excited atoms. $T_r = 1000°K$, $b = (T_r/T)^{1/2}$, a–voigt parameter, P = 5 mm Hg (solid curves), P = 60 mm Hg (dashed),

a — $T = 293°$ K, $b = 1.82$, $a_5 = 0.69 \cdot 10^{-2}$ and $a_{60} = 9.6 \cdot 10^{-2}$;
b — $T = 80°$ K, $b = 3.54$, $a_5 = 1.07 \cdot 10^{-2}$ and $a_{60} = 12.1 \cdot 10^{-2}$;
c — $T = 5°$ K, $b = 14.1$, $a_5 = 1.3 \cdot 10^{-2}$ and $a_{60} = 13 \cdot 10^{-2}$

Some of the curves for large $k_0 l$ are shown in the upper parts of the figure; the ordinate scale remains unchanged, and the abscissa scale is multiplied by a factor of 10.

The relative integral absorption $A = \Delta J/J_0$ depends on the optical thickness $x = k_0 l$, where k_0 is the coefficient of absorption at the center of the Doppler contour of the absorption line. The function A(x) at low temperatures and high concentrations of the metastable atoms is nonlinear and quite complicated. It was tabulated with a computer with account taken of the triplet structure of the emission and absorption lines. The components of the absorption line were described by Voigt contours, and the components of the emission line by Doppler contours. The function A(x) for the 3889 Å triplet helium line was tabulated up to values x = 500 in the pressure interval 1–80 mm Hg, and for temperatures of the absorption layer from 5 to 300°K and the temperatures of the emitting atoms T_r = 600, 800, 1000, and 1200°K. The numerical calculations are discussed in greater detail in [13, 14]. Figure 4 shows plots of the function $A(k_0 l)$ for the two extreme pressures and three temperatures: 293, 80, and 5°K. It is seen from these figures that with decreasing temperature T, the $A(k_0 l)$ dependence becomes less steep, and a given value of A is reached at much higher optical thicknesses. The dependence on the pressure is weak at small $k_0 l$ and becomes

manifest at sufficiently large $k_0 l$. The calculations have shown that for correct determination of the relative absorption, it is important to take into account the multiplet structure of the line, especially at low temperatures. The absolute concentration of the metastable atoms M can be determined from the experimentally measured relative absorption $A = \Delta J/J_0$ and the corresponding theoretical $A(k_0 l)$ curve, using the formula

$$M = k_0 mc\Delta_a / \pi^{1/2} e^2 f, \tag{9}$$

where Δ_a is the Doppler width of the absorption line divided by $2\sqrt{\ln 2}$ ($\Delta_a = 1.66 \times 10^3\ T^{1/2}\ sec^{-1}$), f = 0.65 is the integral oscillator strength of the 2^3S-3^3P transition, m and e are the mass and charge of the electron, and c is the speed of light.

4. QUALITATIVE ANALYSIS OF THE KINETICS OF THE DESTRUCTION OF METASTABLE ATOMS

1. The experimental results described below show that the kinetics of the destruction of the metastable state 2^3S, especially at low temperatures, is determined to a considerable degree by the magnitude and by the rate of change of the electron density in the decaying helium plasma. Therefore it is first necessary to know the coefficient $k_e = \langle v_e \sigma_{se} \rangle$ in the process of the superelastic collision of the metastable atom with the slow electron. The cross section $\sigma_{se}(\epsilon)$ of the process (3) is expressed in terms of the cross section $\sigma_{exc}(E_{exc} + \epsilon)$ of the direct excitation of the metastable atom by a fast electron of energy $E_{exc} + \epsilon$, with the aid of the detailed balancing principle

$$\sigma_{se}(\varepsilon) = \frac{E_{exc} + \varepsilon}{\varepsilon} \sigma_{exc}(E_{exc} + \varepsilon). \tag{10}$$

Here ϵ is the energy of the slow electron, and E_{exc} = 19.79 eV is the threshold of the excited state 2^3S. Because the cross section $\sigma_{exc}(E)$ has been thoroughly investigated, there is a possibility of finding $\sigma_{se}(\epsilon)$ and $k_e(T_e)$. The most exact measurements of the dependence of σ_{exc} on the energy were carried out by Schultz and Fox.[15] A distinguishing feature of this process is the closeness of the threshold of the direct excitation, near which σ_{exc} is proportional to $(E - E_{exc})^{1/2}$, to the resonant level corresponding to the capture of the electron and the formation of the negative He^- ion. The difficulties in the interpretation and the discussions concerning the experimental data of Schultz and Fox are connected precisely with this circumstance. As shown by Baranger and Guerjuoy,[16] the results of Schultz and Fox can be described by the formula

$$\sigma_{exc}(E) = \frac{B(E - E_{exc})^{1/2}}{(E - E_c)^2 + {}^1/_4\Gamma^2}, \tag{11}$$

where E_c = 20.19 eV is the position of the resonant level and Γ = 1 eV is its width; the constant B is expressed in terms of the cross section at the maximum $\sigma_{max} = 4 \times 10^{-18}\ cm^2$ by the relation $B = \sigma_{max}\Gamma^2/4(E_c - E_{exc})^{1/2}$. If the excess of the energy of the electron above the threshold $\epsilon = E - E_{exc}$ is much smaller than 0.5 eV, then expression (11) can be expanded in powers of ϵ, and only the first two terms need be retained:

$$\sigma_{exc}(E_{exc} + \varepsilon) = \frac{B\varepsilon^{1/2}}{(E_c - E_{exc})^2 + {}^1/_4\Gamma^2}\left[1 + \frac{2\varepsilon(E_c - E_{exc})}{(E_c - E_{exc})^2 + {}^1/_4\Gamma^2}\right]. \tag{12}$$

In this case the main term in $\sigma_{exc}(E_{exc} + \epsilon)$ is proportional to $\epsilon^{1/2}$, and in $\sigma_{se}(\epsilon)$ it is inversely proportional to $\epsilon^{1/2}$. Consequently, the coefficient k_e at small electron energies turns out to be practically constant. Using formulas (10) and (12) and substituting the numerical values of the parameters, we obtain[1] after averaging over the Maxwellian distribution, for the region $\epsilon \ll 0.5$ eV

$$k_e = 4.5 \cdot 10^{-9}\left(1 + \frac{T_e^\circ}{4000^\circ}\right) cm^3\text{-}sec^{-1} \tag{13}$$

It can be shown that when T_e is increased k_e increases, passes through a maximum located near 0.5 eV, and then decreases in proportion to $T_e^{-3/2}$. The obtained value $k_e = 4.5 \times 10^{-9}\ cm^3\ sec^{-1}$ for slow electrons is in accord with the experimental data described below.

2. We now proceed to a qualitative discussion of the kinetics of the destruction of metastable atoms in the case of low gas temperatures and high concentrations n and M. In this case we can retain in Eq. (5) only the first two terms:

$$-\partial M/\partial t = kM^2 + k_e nM. \tag{14}$$

When the plasma is excited by short pulses, the concentration of the metastable atoms at the initial instant of the afterglow, M_0, can reach values on the order of or larger than $10^{13}\ cm^{-3}$, and usually exceeds the concentration of the electrons. Therefore immediately after the end of the excitation pulse the inequality (6) is satisfied, and the concentration M decreases in accordance with the hyperbolic law (7). The characteristic time of the decrease of M is of the order of $1/kM_0$, and amounts to several tenths of a millisecond at $M_0 \sim 10^{13}\ cm^{-3}$. The larger the initial concentration M_0, the shorter this time. At high concentrations M, electrons are produced in the paired collision process (1), and if the electron concentration is low at the initial instant of time, it increases and reaches values comparable with M. The time during which we get $n \sim M$ and the condition (6) is violated amounts to $(2-3)/kM_0$. Thus, in order to observe the region of hyperbolic dependence during one or more milliseconds, the initial concentrations should be of the order of $10^{12}\ cm^{-3}$. Subsequently, destruction in collisions with electrons begins to predominate. When

$$k_e n > kM, \tag{15}$$

the term kM^2 in (14) can be neglected. As will be shown below, the inequality (15) corresponds to a slower variation of the electron density $|dn/dt|$ compared with $|dM/dt|$. Consequently it is possible to integrate Eq. (14) approximately by replacing n with a certain mean value $\bar{n}$:

$$M(t) = M_1 \exp(-k_e \bar{n} t). \tag{16}$$

The interval of hyperbolic dependence should therefore be followed by a region in which M(t) decreases approximately exponentially, with an exponent determined by the average electron density.

3. We can attempt to refine the laws governing the

[1] The independence of k_e of T_e was also pointed out in Smirnov's book [17], but the value given by him, $k_e = 1.7 \times 10^{-10}\ cm^3\ sec^{-1}$, differs from ours.

decrease of M(t) in the region (15), by analyzing in greater detail the kinetics of the variation of the electron concentration n(t) together with (14). The equation for n(t) will be written in the form

$$\partial n / \partial t = {}^1/_2 kM^2 - cn^3 - (\alpha_0 + \alpha_1 P) n^2 - D_a \lambda^{-2} n. \qquad (17)$$

The first term in the right-hand side of (17) describes the production of an electron upon collision of two metastable atoms. The factor $^1/_2$ is a result of the fact that the loss of two metastable atoms is accompanied by the production of one electron. The succeeding two terms correspond to the decrease of the electron concentration upon recombination. The term cn^2 is the rate of the impact-radiative recombination with participation of an ion and two electrons.[2)] The recombination coefficient $\alpha = \alpha_0 + \alpha_1 P$ includes both the possible dissociative recombination α_0 and the recombination in triple collision with participation of a neutral atom $\alpha_1 P$. Finally, the last term describes the loss of electrons because of ambipolar diffusion, with a diffusion coefficient D_a.

The relative role of the terms determining the decrease of the number of electrons n depends on the value of the electron density, on the pressure, and on the coefficients c, α, and $D_a \lambda^{-2}$. The theoretical calculation of the coefficient of impact-radiative recombination cn has been the subject of a large number of papers.[17-19]. According to calculations by A. Gurevich and Pitaevskiĭ,[19] subsequently confirmed by Veselovskiĭ,[19] the coefficient c for singly-charged ions is equal to

$$c = \frac{\pi \ln 2}{2^{5/2}} \frac{e^{10}}{m^{1/2} (\varkappa T_e)^{9/2}} = 0.62 \cdot 10^{-8} T_e^{-9/2} \text{ cm}^6\text{-sec}^{-1}, \qquad (18)$$

where $\varkappa$ is the Boltzmann constant. Numerical calculations by Bates et al.[18] give a somewhat larger numerical factor in formula (18).

The coefficient of electron-ion recombination in triple collision with participation of the neutral atom $\alpha_1 P$ can be estimated from the known Thomson formula (see also [20]). It turns out that at $T_e \lesssim 400°K$ and $P \lesssim 60$ mm Hg $\alpha_1 P$ is smaller by one order of magnitude than cn even at $n \gtrsim 10^{11}$ cm^{-3}, and therefore the Thomson recombination can be neglected.

The question of the coefficient of dissociative recombination for helium is much more complicated. For the diatomic molecular ion He_2^+, recombination with dissociation occurs exceedingly slowly. At low temperatures, however, there exists a possibility of dissociative recombination for polyatomic ions of the type He_3^+. In a helium plasma at low temperatures, the presence of such ions is very probable, as is evidenced by the data of [21], and also by our preliminary results on the measurements of the electron density at low temperatures.[3)] However, the exact value of α_0 is unknown. If we assume that $\alpha_0 \approx 10^{-9}$ cm^3 sec^{-1}, then impact-radiative recombination dominates at $n \gtrsim 5 \times 10^{11}$ cm^{-3}. This assumption apparently does not contradict the experimental results described below.

Estimates show that in the same region of electron densities and at pressures $P \gtrsim 5$ mm Hg, ambipolar diffusion does not play a noticeable role compared with recombination. Thus, at low temperatures and high concentrations n, the principal role in (17) is played by impact-radiative recombination, and possibly by dissociative recombination, in addition to the production term $kM^2/2$.

Owing to the competition between the additional ionization $kM^2/2$ and the recombination, the electron density n(t) changes more slowly in the region (15) than the density of the metastable atoms M(t). Therefore the quasistationarity condition is approximately satisfied for n(t):

$$dn / dt = {}^1/_2 kM^2 - (cn + \alpha_0) n^2 \approx 0, \qquad (19)$$

and the exponential law (16) is valid to the degree that this condition is satisfied.

In the region intermediate between the hyperbolic (7) and the exponential (16) relations, when $cn > \alpha_0$, the quasistationarity condition (19) yields

$$kM^2 / 2 = cn^3. \qquad (20)$$

If we determine n from this equation and substitute it in the equation $dM/dt = -k_e nM$, then M(t) will obey the "two-thirds law":

$$M^{-2/3}(t) = M_1^{-2/3} + \beta t, \quad \beta = {}^2/_3 k_e (k/2c)^{1/3}. \qquad (21)$$

The existence of an interval where the "two-thirds law" holds makes it possible to determine the constant β from the experimental curves and to determine, for the known values of k and k_e, the recombination constant c. It must be borne in mind here that the interval of the approximate law (21) is relatively small ($\sim$ 1–2 msec) and usually corresponds to a slight change of the electron temperature T_e.

5. EXPERIMENTAL RESULTS

Our results cover the range of concentrations M from 10^{13} to 10^{10} cm^{-3}. The lower limit is connected with the short length of the discharge vessel, which must be placed inside the cryostat. A decrease of the density of the metastable atoms within these limits occurs within several milliseconds. The initial concentrations M_0 and the rate of change of M(t) depend on the gas temperature. We present first the experimental data at T = 300°K. Figure 5 shows by way of an example the kinetics M(t) in a semilogarithmic scale for four values: 5, 11, 20, and 60 mm Hg. The excitation conditions are given in the captions in the figure. It is seen that starting with 0.2 msec and approximately up to 2 msec, M(t) decreases exponentially. Depending on the pressure and on the input power, the arguments of the exponential ν range from 1 to 5 msec^{-1}. When the pressure changes, starting with 3 mm Hg, the slope of ν first decreases, reaches a minimum at 10–20 mm Hg, and then increases. The position of the minimum depends on the dimensions of the discharge vessel. The dependence of ν on P is seen in Fig. 5; the values of ν at 50 and 60 mm Hg, as well as the slopes at 11 and

[2)] Since the coefficient of impact-radiative recombination c does not depend on the ion mass, in the presence of several species of helium ions N_i this term can have the form $cn^2 \Sigma N_i$, and reduces to cn^3 as a result of the quasineutrality of the plasma.

[3)] We note that since molecular helium ions predominate at low temperature, the excited molecule He_2^* is produced even as a result of dissociative recombination, rather than the atom He*. Therefore Eq. (17) does not contain a term corresponding to the production of metastable atoms in the recombination process.

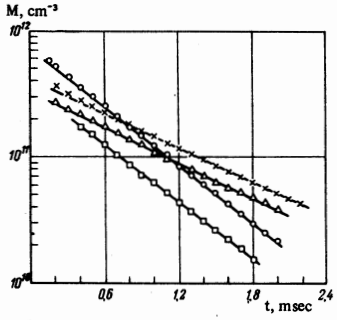

FIG. 5. Change of concentration of metastable atoms in the afterglow at room temperature. $\lambda^{-2} = 5.4$ cm^{-3}; $\tau = 6$ μsec; u = 0.6 kV.

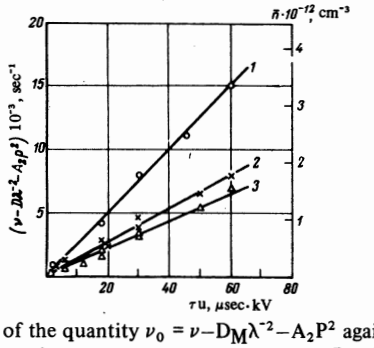

FIG. 6. Plots of the quantity $\nu_0 = \nu - D_M\lambda^{-2} - A_2P^2$ against the pulse duration τ and the voltage u at room temperature: 1—P = 10, 2—P = 20, 3—P = 40 mm Hg. The right-hand scale pertains to $n = \nu/k_e$ ($k_e = 4.5 \cdot 10^{-9}$ cm^3 sec^{-1}).

P, mm Hg		$10^{-2}\nu$, sec^{-1}
○:	5	18
×:	11	10.6
△:	20	10.7
□:	60	17.3

20 mm Hg, are pairwise equal. The decrease in ν with increasing P in the region of small pressures is due to diffusion destruction of the metastable atoms and gives a coefficient $D_M = (470/P)$ cm^2 sec^{-1}. The increase in ν in the region of large pressures is in proportion to P^2, in accordance with the three-body process (4). The proportionality coefficient measured by us turned out to be $A_2 = (0.35 \pm 0.05)$ cm^3 sec^{-1} (mm Hg)$^{-2}$. The values of the coefficients D_MP and A_2 agree with the known data.[2, 8-10] The argument of the exponential ν in the general case is a sum of three terms

$$\nu = D_M\lambda^{-2} + A_2P^2 + \nu_0, \qquad (22)$$

the last term being strongly dependent on the pulse duration τ and on the voltage amplitude u. Figure 6 shows the dependence of ν_0 on τu for pressures 10, 20, and 40 mm Hg. This is essentially the dependence on the number of charges passing through the plasma during the time of the pulse. From the fact that ν_0 varies linearly with τu and can be extrapolated to the origin it follows that ν_0 is determined entirely by the electrons. In accordance with the results of the preceding section, we can assume that $\nu_0 = k_e\bar{n}$. We note that these mean values of the concentration $\bar{n}$ in the region of small τu are of the same order of magnitude as the concentrations of the metastable atoms after 0.2–0.3 msec following the cessation of the pulse (compare with Fig. 5).

At room temperature we were unable to observe a clearly pronounced hyperbolic M(t) dependence. The

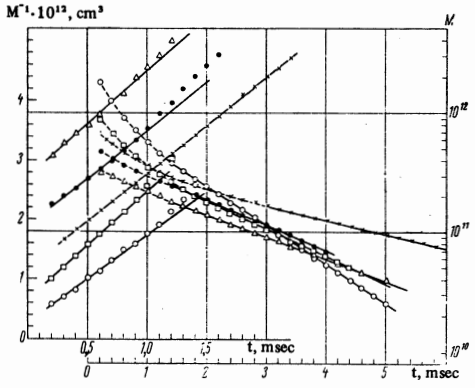

FIG. 7. Change of the concentration of the metastable atoms in the afterglow when cooled with liquid nitrogen (T = 80°K), $\lambda^{-2} = 5.4$ cm^{-2}. The left ordinate scale and the upper abscissa scale correspond to $M^{-1}(t)$; the right logarithmic ordinate scale and the lower abscissa scale describe the M(t) dependence.

P, mm Hg		τ, μsec	u, kV	10^9 cm^3·sec^{-1}	$10^{-3}\nu$, sec^{-1}
●:	5	3	0.4	1.6	4.7
×:	10	3	0.5	1.6	3
○:	10	9	0.4	1.5	7.6
△:	31	5	0.4	1.68	4.2
□:	60	3	1	1.8	4.9

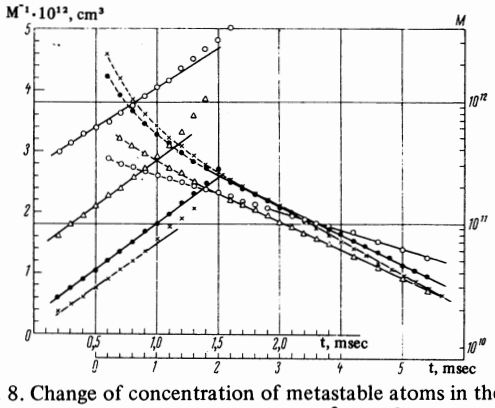

FIG. 8. Change of concentration of metastable atoms in the afterglow upon cooling with liquid helium (T ≈ 9°K), $\lambda^{-2} = 5.4$ cm^{-2}, $\tau = 6$ μsec, u = 0.5 kV. The left ordinate scale and the upper abscissa scale correspond to $M^{-1}(t)$; the right ordinate scale and the lower abscissa scale describe the dependence of lnM on t.

P, mm Hg		$10^9 k$, cm^3·sec^{-1}	$10^{-3}\nu$, cm^{-1}
●:	3	1.4	5.8
×:	10	1.4	6.45
△:	20	1.3	5.5
○:	45	1.2	3.5

reason is that the initial concentrations n_0 and M_0 at the instant when the excitation pulse terminates have the same order of magnitude, and in some cases we even have $n_0 > M_0$. Therefore practically immediately after the termination of the pulse the condition (15) is realized, and M(t) decreases exponentially. In general, it should be noted that at room temperature the initial concentrations of the metastable atoms M_0 are usually smaller by several times, and the initial electron concentrations n_0 are noticeably higher than the corresponding values of M_0 and n_0 at low temperatures under the same excitation conditions. The decrease of the initial concentrations n_0 with decreasing temperature of the atoms was verified by measuring the plasma resistance during the time of the pulse. It turned out that the plasma resistance R_{p1} increased by 10–15 times

Table 1. Rate constant of the process of collision of two metastable atoms of He 2^3S.

T, °K	$k \cdot 10^9$, $cm^3 \cdot sec^{-1}$	
	Experiment	Calculation by formula (25)
10	1.3±0.2	1.1
80	1.65±0.2	1.48
120	1.7±0.2	1.58
300	2 [8]	1.83

when the temperature was lowered from room temperature to that of liquid helium, and the characteristic initial concentrations were $n_0(300°) \sim (2-3) \times 10^{12}$ cm^{-3} and $n_0(10°) \sim (1-2) \times 10^{11}$ cm^{-3}.

Owing to the fact that the initial concentrations of the electrons decrease with decreasing gas temperature, one can observe the hyperbolic dependence (7) at low temperatures. Figures 7 and 8 (the curves on the left side) illustrate the presence of such dependences from 0.2 to 1.5 msec after the termination of the pulse, when cooled with liquid nitrogen and helium, for different pressures and excitation conditions. The initial sections of the curves follow a linear dependence of M^{-1} on t, and their slopes give the value of the constant k, averaged values of which for different temperatures are given in Table 1 below. As the temperature is decreased, the constant k decreases. Starting with approximately 2 msec, the $M^{-1}(t)$ dependence in our experiments deviates from linear. The larger the power fed to the discharge, the sooner this deviation takes place. Reduction of more than 100 experimental curves has shown that the constant k does not depend on the pressure, whereas the degree of deviation of $M^{-1}(t)$ from linear and the characteristic time at which this deviation begins depend on the pressure. The dependence of the effective coefficient k* on P given by us earlier in [11] was obtained under conditions of still more powerful excitation, when an important role was played not only by the process (1), but also by destruction in collisions with slow electrons.

Further destruction of the density M is satisfactorily approximated by the exponential relation (16) (see the curves on the right of Figs. 7 and 8). The exponents depend more strongly on the voltage and on the duration of the excitation pulse than on the pressure. Only in the case of a small vessel with $\lambda^{-2} = 37$ at low pressures P ~ 3–20 mm Hg does diffusion appear. The dependence of ν on P at high pressures, owing to the three-body process (4) with participation of two unexcited helium atoms, is not observed. This corresponds to a sharp decrease of the constant A_2 on going from room temperature to the lower temperatures of the gas. A characteristic feature of the exponential relations is their faster decrease at higher initial concentrations. This also confirms the important role played by the additional ionization as a result of paired collisions of metastable atoms. From ν we can determine the values of $\bar{n}$, averaged over the exponential-dependence interval 2–5 msec. These values of $\bar{n}$ amount to $(1-3) \times 10^{11}$ cm^{-3} and coincide in order of magnitude with the average concentrations of the metastable atoms in this interval. We note that sometimes small deviations from the exponential

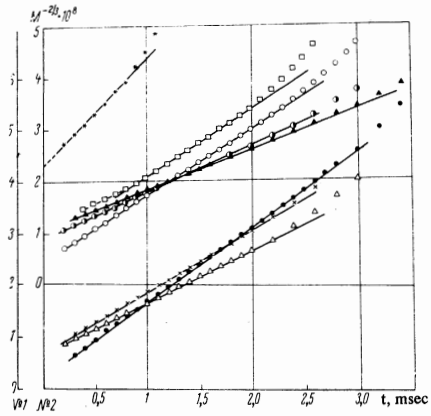

FIG. 9. Dependence of $M^{-2/3}$ on t for the different excitation conditions, temperatures, and pressures. The three lower curves pertain to scale No. 1, which is shifted for convenience relative to scale No. 2. Discharge vessel $\lambda^{-2} = 5.4$ cm^{-2}. The six lower curves—cooling with liquid helium (T ≈ 6°K). The values of M_0 correspond to t = 0.2 msec after the end of the pulse.

P, mm Hg		τ, μsec	u, kV	$10^{-12} M_0$, cm^{-3}	$10^5 \beta$	T_e, °K
●:	5	4	1.2	3.5	1.45	470
×:	5	7	0.4	1.1	1.19	430
△:	5	5	0.4	1.2	0.97	370
○:	20	9	0.8	1.7	1.26	440
◑:	20	7	0.4	0.8	0.9	350
▲:	20	5	0.4	0.75	0.74	306
When cooled with liquid nitrogen (T ≈ 80°K)						
□:	20	4	0.8	0.6	1.26	410
At room temperature						
∗:	20	5	0.4	0.2	2.1	550

relations are observed—a somewhat slower decrease of M(t) at the end of the exponential region.

In a number of cases, principally at large initial concentrations of the metastable atoms, a section of linear dependence of $M^{-2/3}$ on t is observed; this section sometimes precedes and sometimes overlaps somewhat the exponential region. This region is most clearly pronounced when the interval of the hyperbolic dependence (7) is short. Figures 9 and 10 illustrate the existence of the "two-thirds law." This law is satisfied best at helium temperatures. The characteristic slopes of $M^{-2/3}(t)$ are of the order of 10^{-5} cm^2 sec^{-1} and are given in the captions of Figs. 9 and 10. Usually the deviation from this dependence begins at 2–2.5 msec in concentrations $M \sim (3-4) \times 10^{11}$ cm^{-3}. Knowing the experimental slope β, we can determine with the aid of (21) the impact-radiative recombination constant, which in this region turns out to be of the order of $c \sim (0.5-1.5) \times 10^{-20}$ cm^6 sec^{-1}. According to formula (18) this corresponds to electron temperatures $T_e \sim 350-450°K$. The values of T_e for the curves of Figs. 9 and 10 are given in the captions. The values of c turn out to be smaller, and those of T_e correspondingly larger, in the region where the concentration of the metastable atoms is higher. These data show that in the region up to 2–2.5 msec there is still no complete cooling of the electrons. This means that in this region there exist processes whereby the plasma electrons are heated. We note that a tendency is observed for decreasing T_e with increasing pressure, although the dependence of T_e on P is weak.

6. DISCUSSION OF RESULTS

1. In this paper we have measured for the first time the temperature dependence of the paired-collision constant of metastable atoms, and it is therefore of interest to compare it with the theory. The probability of collision of two metastable atoms is due to the van der Waals attraction forces and is determined by the cross section of their mutual capture σ. According to classical scattering theory,[22] the capture cross section for a potential $V = -Cr^{-s}$ is

$$\sigma(E) = \pi s (s-2)^{(2-s)/s} \left(\frac{C}{2E}\right)^{2/s}. \quad (23)$$

The classical value of the constant of the process (1) at s = 6, averaged over the atomic velocity v, is given by the expression

$$\langle \sigma v \rangle = 6 \cdot 2^{1/2} \pi^{1/2} \Gamma(5/3) \frac{C^{1/3}}{\mu^{1/2}} (\varkappa T)^{1/6}, \quad (24)$$

where $\Gamma(5/3) = 0.903$ and μ is the mass of the helium atom. According to the Wigner rule, the decay of a helium quasimolecule in process (1) proceeds with conservation of the total spin. This decreases the number of allowed decays and leads to the appearance of the factor $4/9$ in the classical cross section, to account for the quantum forbiddenness in the spin. Using the value $C = 3.29 \times 10^3$ at.un. given in Sec. 2, we obtain (T is in degrees K)

$$k = {}^4/_9 \langle \sigma v \rangle = 0{,}71 \cdot 10^{-9} T^{1/6} \text{ cm}^3\text{-sec}^{-1}. \quad (25)$$

Table 1 gives the values of k calculated from formula (25). We see that there is satisfactory agreement between the experimental and theoretical values of k. Experiment confirms the weak temperature dependence $k \sim T^{1/6}$ predicted by the theory. From the fact that the average experimental value is even slightly higher than the theoretical one, it follows that the ionization probability P_i in capture that is introduced in certain papers (e.g., [23]) is equal to unity in the case of collision between two metastable helium atoms. This means that such a collision must result in additional ionization.

2. Volume processes of destruction of metastable atoms of He lead to heating of the electrons in the plasma. We shall show that the high electron temperatures, $T_e \sim 350$–$450°K$, observed in the experiment (Figs. 9 and 10) up to 1.5–2 msec, can be attributed to transfer of energy from the excited atoms to the electrons from processes (1) and (3). The electrons are also heated by recombination. Owing to the high rate of energy transfer from the electrons to the atoms and ions, it can be assumed that at each instant of time there exists equilibrium between the heating and cooling of the electrons

$$^3/_2 n (\nu_{ea} + \nu_{ei})(T_e - T) = k_e n M E_{\text{exc}} + {}^1/_2 k M^2 \varepsilon_1 + cn^3 \varepsilon_2. \quad (26)$$

Here $E_{exc} = 19.8$ eV is the energy transferred to the electron in the super-elastic collision (3); $\epsilon_1 = 9$ eV is the average energy of the electron produced in the process (1),[24] $\epsilon_2 = 1$ eV is the energy released in impact-radiative recombination,[25] ν_{ea} and ν_{ei} are the frequencies of the collisions of the electrons with the atoms and ions with energy transfer,

$$\nu_{ea} = \frac{2m}{\mu} \sigma_{ea} N \left(\frac{8\varkappa T_e}{\pi m}\right)^{1/2} = 3 \cdot 10^3 P T_e^{1/2}, \quad (27)$$

$$\nu_{ei} = \frac{4e^4 n}{3(\varkappa T_e)^{3/2}} \frac{(2\pi m)^{1/2}}{\mu} L = 2{,}8 \cdot 10^{-3} n T_e^{-3/2}, \quad (28)$$

$\sigma_{ea} = 5.3 \times 10^{-16}$ cm^2 is the cross section for elastic scattering of electrons by the He atom, N is the density of the unexcited He atoms, L = 5.7 is the Coulomb logarithm. Expression (28) is the well-known Landau formula.[26]

An example of calculation of heating of electrons in accordance with (26)–(28), for P = 10 mm Hg and for different temperatures T, is given in Table 2. For sim-

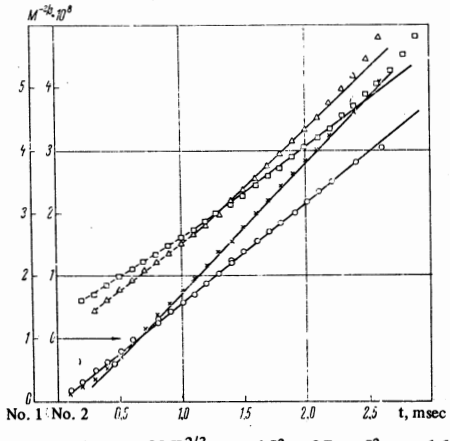

FIG. 10. Dependence of $M^{-2/3}$ on t. $\lambda^{-2} = 37$ cm^{-2}; $\tau = 16$ μsec; u = 0.6 kV. Lower pair of curves–cooling with liquid helium–corresponds to scale No. 1. The values of M_0 are given at t = 0.2 msec after the end of the pulse. Upper pair of curves–cooling with liquid-nitrogen vapor ($T \approx 120°K$).

	P, mm Hg	$10^{-12} M_0$, cm^{-3}	$10^5 \beta$	T_e, °K
×	10	10	2	600
□	60	7	1.6	510
△	20	4	1.82	520
○	60	2,5	1.47	450

Table 2. Analysis of electron heating in a helium plasma.

P = 10 mm Hg					T = 300° K		T = 100 ° K			T = 10° K		
1	2	3	4	5	6	7	8	9	10	11	12	13
n = M, cm^{-3}	$k_e M E_{exc}$ sec^{-1}, g	$\frac{kM^2}{2n}\varepsilon_1$, sec^{-1}, g	Summary heating	$T_e^{1/2}(T_e - T)$, deg$^{3/2}$	T_e, deg	$\frac{\nu_{ea}}{\nu_{ei}}$	T_e, deg	$cn^2\varepsilon_2$, sec^{-1}, g	$\frac{\nu_{ea}}{\nu_{ei}}$	T_e, deg	$cn^2\varepsilon_2$, sec^{-1}, g	$\frac{\nu_{ea}}{\nu_{ei}}$
$3 \cdot 10^{12}$	$3 \cdot 10^9$	$0{,}3 \cdot 10^9$	$3{,}3 \cdot 10^9$	$7{,}65 \cdot 10^4$	2000	13.3	1870		11.7	1800		10,7
$1 \cdot 10^{12}$	10^9	$0{,}1 \cdot 10^9$	$1.1 \cdot 10^9$	$2.55 \cdot 10^4$	1080	11.8	935		8,7	875		7.65
$3 \cdot 10^{11}$	$3 \cdot 10^8$	$0.3 \cdot 10^8$	$3{,}3 \cdot 10^8$	$7.65 \cdot 10^3$	610	12.4	460		7.1	395	$1.4 \cdot 10^7$	5.2
$1 \cdot 10^{11}$	10^8	$0{,}1 \cdot 10^8$	$1{,}1 \cdot 10^8$	$2.55 \cdot 10^3$	425	18	260	$1 \cdot 10^7$	6.75	193	$0.5 \cdot 10^8$	3,75
$3 \cdot 10^{10}$	$3 \cdot 10^7$	$0.3 \cdot 10^7$	$3.3 \cdot 10^7$	$7{,}65 \cdot 10^2$	340	38,6	160	$0{,}9 \cdot 10^7$	8,5	91	$1.1 \cdot 10^8$	2.8

In columns 9 and 12, the empty spaces correspond to quantities that are negligibly small compared with those given in column 4.

plicity we have confined ourselves here to the case $n = M$. The values of T_e were calculated with allowance for only the first two heating terms in (26) and without taking ν_{ei} into account. From the ratios ν_{ea}/ν_{ei} given in columns 7, 10, and 13 we see that cooling of the electrons by the atoms is much faster than cooling by ions. The largest heating is obtained from superelastic collision of an electron with a metastable atom (column 2). However, when the concentration M is decreased and T_e is lowered, it is also necessary to take the recombination heating into account. For the data of Table 2, it becomes significant at low cooling temperatures and at values $T_e \sim 250$–$200°K$. The increase of the recombination heating with decreasing T_e is due to the sharp growth of the impact-radiative recombination due to the strong dependence of c on T_e (18). This mechanism slows down the decrease of T_e at the level 200–250°K for the example in question. Consequently, the simultaneous action of all three heating processes from (26) leads to "self-maintenance" of the electron temperature in a certain interval of variation of n and M.

Under real experimental conditions, the role of the recombination heating in all probability increases. This is connected with the fact that in the region of concentrations $M \sim (5-1) \times 10^{11}$, corresponding to the quasistationarity condition (19), the electron density exceeds M, and the value of T_e should be larger than in the considered example. Those relatively high $T_e \sim 350$–$450°K$ determined from the "two-thirds law" and observed in the region of concentrations M and n where the quasistationarity and increased value of T_e is maintained, are therefore not surprising. The same mechanism of "self-maintenance" of T_e explains the very weak dependence of T_e on the pressure that was observed in the experiment.

3. The exponential dependence $M(t) \sim \exp(-\nu t)$ found in the experiment at large values of the time ($t \gtrsim 2$ msec) enables us to determine the average concentration of the electrons $\bar{n} = \nu/k_e$. The use of the value $k_e = 4.5 \times 10^{-9}$ cm^3 sec^{-1} calculated in Sec. 4 also gives values of $\bar{n}$ that agree reasonably with other experimental data. The fact that the exponential is observed in the final interval and that collisions have been noted at large times should signify that the exponential dependence is only an approximation of some other law. In this region, an important role may be played by dissociative recombination of the electrons with polyatomic ions. If this is so, then we should have $M(t) \sim (1 + \alpha n_1 t) \exp(-k_e/\alpha)$. Processing of the plots of log M against log t at times from 3 to 5–6 msec gave an exponent in the range from 1.8 to 2.5, corresponding to $\alpha \sim (2-2.5) \times 10^{-9}$ cm^3 sec^{-1}. To verify and confirm these preliminary data, further experiments will be performed at low metastable-atom concentrations.

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