

*ELECTRON-ION RECOMBINATION AND DIFFUSION IN A CRYOGENIC HELIUM PLASMA*

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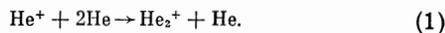
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The kinetics of recombination and diffusion destruction of the electron density  $n$  in a helium plasma is measured by the resonator technique in a broad temperature range from 293 to 10°K and in a pressure range from 1 to 30 mm Hg. The ambipolar diffusion coefficient of a diatomic  $\text{He}_2^+$  ion is measured at room temperature and found to be  $D_2P = 680 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ . A triatomic helium ion  $\text{He}_3^+$  with a coefficient  $D_3P = (280 \pm 15) \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$  has been observed at 100°K. The collision cross sections for polyatomic helium ions colliding with atoms are estimated on the basis of these measurements. With decreasing temperature, the region of recombination plasma destruction broadens considerably and a linear dependence of  $n^{-1}(t)$  is observed in a large range of concentration variation. The two-particle recombination coefficients  $\alpha$  are independent of the pressure and equal  $5.8 \times 10^{-8} \text{ cm}^3 \text{ sec}^{-1}$  at about 100°K and  $1.3 \times 10^{-7} \text{ cm}^3 \text{ sec}^{-1}$  for a plasma cooled by liquid helium. At 10°K the electrons cool off at a very slow rate in a helium plasma. Recombination mechanisms are discussed and it is concluded that the main contribution to such large values of  $\alpha$  is due to collision-radiative recombination with dissociation and also to purely dissociative recombination of triatomic  $\text{He}_3^+$  ions with electrons.

1. INTRODUCTION

THE main processes leading to a decrease of the density of electrons and ions in a decaying plasma are ambipolar diffusion and electron-ion recombination. The destruction of the plasma is accompanied by prolonged and rather intense optical radiation, and therefore plasma decay is frequently called afterglow. In the early afterglow, when the electron and ion concentration is sufficiently large, volume-recombination processes predominate, and in the later afterglow the principal role is played by ambipolar diffusion.

At room temperature and above, a helium plasma contains, besides the atomic  $\text{He}^+$  ions, also diatomic molecular ions  $\text{He}_2^+$ . In the afterglow, the formation of diatomic ions proceeds during the course of the conversion of the atomic  $\text{He}^+$  ions in accordance with the scheme



The conversion frequency  $\nu_c$  is obviously proportional to the square of the density of the normal atoms  $N$ , and amounts at  $T = 293^\circ\text{K}$  to

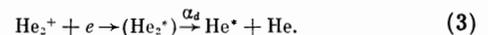
$$\nu_c = 0.93 \cdot 10^{-31} N^2 [\text{sec}^{-1}] = 100 P^2 [\text{sec}^{-1}] \tag{2}$$

where  $P$  is the gas pressure referred to 293°K. The rate of conversion increases with decreasing temperature like  $T^{-3/4}$ . [1-3] At low pressures, atomic  $\text{He}^+$  ions predominate in the plasma, and starting with pressures of several mm Hg, recombination and ambipolar diffusion proceed with predominant participation of the diatomic  $\text{He}_2^+$  ions.

The recombination coefficient  $\alpha$  in a helium plasma at 300°K and above was measured by many workers (see, for example [4, 5] and the results cited therein). Its value turned out to be of the order of  $10^{-9}$ - $10^{-8} \text{ cm}^3 \text{ per sec}^{-1}$  and depended strongly on the conditions of

plasma excitation. The question of the recombination mechanism has remained under discussion to this day. [6-10] For a helium plasma, the most important among the known recombination mechanisms are the following three: shock-radiative, Thomson, and dissociative.

In all the earlier investigations, it was assumed that the recombination of molecular helium ions proceeds with formation of unstable quasimolecules  $\text{He}_2^*$  and subsequent dissociation into an excited atom of helium and a normal one in accordance with the scheme



The necessary condition for dissociative recombination of molecular ions is the presence of repulsive molecular states between the ionization boundary and the lower state of the  $\text{He}_2^*$  molecule. In 1964, Mulliken showed [11] that the excited  $\text{He}_2^*$  molecule is stable against dissociation, since the potential curves of the upper Rydberg states ( $n \geq 4$ ) have deep minima and their asymptotes lie near the molecule ionization boundary. The lower states  $n = 3f\sigma$  and  $4f\sigma$  are stable because of the presence of high maxima on the potential curves. It was concluded on the basis of Mulliken's calculations that dissociative recombination of the  $\text{He}_2^*$  is hindered.

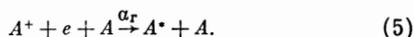
At approximately the same time, theoretical papers were published by D'Angelo, Bates, Kingston, and McWhirter, [12-14] by Gurevich and Pitaevskii, [15] and by a few others. In these papers there was constructed a quantitative theory of shock-radiative recombination with participation of an ion and two electrons, in accordance with the scheme



The coefficients of this recombination are proportional to the electron density, increase strongly with decreasing temperature, and can be large enough to explain the

values of the coefficients at room temperatures. Subsequent experiments<sup>[5-8]</sup> have shown that the features of the recombination decay of a helium plasma can be reconciled with the mechanism (4). However, the possibility of dissociative recombination of  $\text{He}_2^+$  apparently cannot be fully excluded. Hurt and Collins<sup>[16]</sup> and Robertson<sup>[17]</sup> noted that as a result of the radiative transition  $B_{2S}, {}^3\Sigma_g^+ \rightarrow 3d\pi, {}^3\Pi_u$  the  $\text{He}_2^+$  molecule turns out to be in the auto-ionized state beyond the maximum of the long-range repulsion, and dissociates into an atom in the state  $2^3P$  and an unexcited atom. Calculations of the potential curve of the  $3d\pi, {}^3\Pi_u$  state of  $\text{He}_2^+$  performed by Gupta and Matsen<sup>[18]</sup> and the experiments of<sup>[16, 17]</sup> confirm the possibility of such a dissociation.

At high gas pressures and at relatively low electron concentrations, Thomson recombination<sup>[19]</sup> taking place with participation of a neutral atom



may compete with the shock-radiative and dissociative recombinations. The latest calculations of the coefficient  $\alpha_T$  of the triple recombination (5) were performed by Pitaevskii<sup>[20]</sup> and by Bates and Khare.<sup>[21]</sup> The experiments performed by Berlande and his colleagues<sup>[22]</sup> in a wide range of pressures at room temperature gave a value  $\alpha_T = (6.6 \pm 1.6) \times 10^{-11} \text{ P} [\text{cm}^3 \text{ sec}^{-1}]$ , in close agreement with the theory of<sup>[20]</sup>.

There have been practically no investigations of the kinetics of the electron density in a decaying helium plasma at low temperatures. One can cite only the paper of Goldan and Goldstein,<sup>[23]</sup> in which a waveguide method was used to study collisions of electrons with atoms in a helium plasma at 77 and 4°K. Direct measurements of the electron density in a cryogenic plasma are particularly interesting because with decreasing temperature there is an increased probability of formation of new and stable ion-molecular complexes, particularly polyatomic helium ions  $\text{He}_n^+$  ( $n > 2$ ). The appearance of such ions can change the character and magnitude of the recombination and ambipolar-diffusion coefficients. Thus, triatomic helium ions  $\text{He}_3^+$  were observed in the temperature region near 100°K by Paterson in a measurement of the mobility of ions in a drift tube<sup>[24]</sup> and by us in a study of the kinetics of the electron density by a microwave method and of the intensity of the afterglow in the region of diffusion decay of a plasma.<sup>[25, 26]</sup>

In the present paper we present the results of investigations of recombination and diffusion decays of the electron component of a helium plasma at room temperature, in the region of 100°K and when cooled with liquid helium. We analyze the magnitude and temperature dependence of the mobilities of different molecular ions and the cross section of ion-atom diffusion. We discuss the mechanisms of electron-ion recombination and processes of electron heating in a plasma at low temperatures.

## 2. APPARATUS AND EXPERIMENTAL PROCEDURE

The electron density in the plasma was determined with the aid of the well known resonator microwave method (see, for example, <sup>[27]</sup>). Figure 1 shows a block diagram of the experimental setup for measurements at

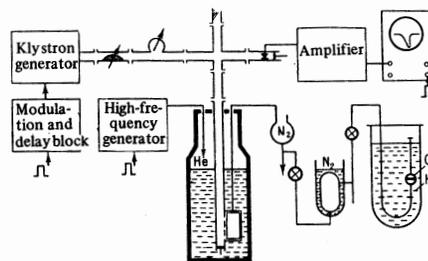


FIG. 1. Block diagram of experimental setup.

low temperatures. The apparatus consists of a vacuum system for clearing and filling the vessel with helium at different pressures, a resonator with a discharge bulb (placed inside a metallic cryostat and cooled down to  $T = 4.2^\circ\text{K}$ ), and microwave apparatus for sounding the plasma. The discharge vessel is a cylindrical quartz bulb of radius  $R_1 = 2 \text{ cm}$  and length  $L_1 = 2.8 \text{ cm}$ , with thin walls (0.8–1 mm), and is placed in a cylindrical copper resonator and connected to the vacuum system.

The glass vacuum system was evacuated and conditioned to a pressure not higher than  $3 \times 10^{-7} \text{ mm Hg}$ ; it was cleaned out by a high frequency discharge. The gaseous helium was gathered directly from the vapor over the liquid helium in the Dewar and passed through a special filter and traps with liquid nitrogen. The pressure was monitored during the course of the measurements with an oil manometer. The temperature of the discharge-vessel walls in the region of  $100^\circ\text{K}$  was measured with a copper-constantan thermocouple. When cooled with liquid helium, the temperature of the gas in the plasma could be estimated from the period of the acoustic oscillations excited in the discharge at the instant of the breakdown (see Fig. 2). The range of helium densities from  $3.3 \times 10^{16}$  to  $10^{18} \text{ cm}^{-3}$  was used, corresponding to the range of gas pressures  $P$  at  $293^\circ\text{K}$  from 1 to 30 mm Hg. For all other temperatures  $T$ , we give the pressures in units referred to  $293^\circ\text{K}$ , namely,  $P = p \times 293/T$ , where  $p$  is the pressure at the temperature  $T$ .

To excite the plasma in the discharge chamber, the resonator was slotted across the axis. One half-cylinder was grounded, and to the other, insulated with a teflon liner, we applied a high-frequency excitation pulse (carrier frequency 10–20 MHz) from a laboratory oscillator based on a GI-30 tube. The excitation-pulse duration  $\tau$  was varied from 10 to 50  $\mu\text{sec}$ . The repetition frequency was 10–25 Hz. The power of the plasma excitation pulse did not exceed 1 kW.

The sounding microwave signal was fed from a klystron generator. The generator and the matched detector section were connected in the arms of a waveguide tee. The signal reflected from the resonator was detected and fed after amplification to the oscilloscope screen. At the instant when the frequency of the resonator with the plasma and that of the klystron coincided, a minimum of the reflected signal was registered on the oscilloscope. At a low rate of change of the electron density, to increase the accuracy with which the position of the resonance was read, frequency modulation of the klystron was used. The modulator was triggered with the aid of an adjustable-delay block synchronized relative to the exciting pulse. The accuracy with which the

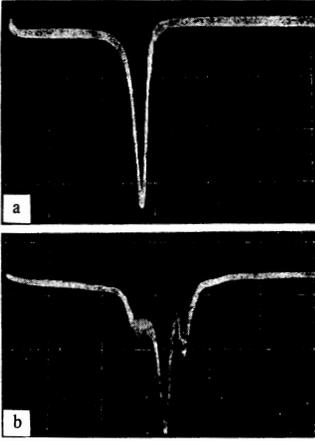


FIG. 2. Oscillograms of resonant signal (one division corresponds to 0.25 msec): a— $T = 293^\circ\text{K}$ , b— $T = 10^\circ\text{K}$ . Additional maxima are seen, caused by low-frequency modulation with period  $\Delta T \approx 180\mu\text{sec}$ .

shift of the resonant frequency could be measured with the wavemeter was 0.5–1 MHz. The power of the sound-ing signal was several microwatts.

The resonator was designed for excitation of the  $\text{TE}_{011}$  mode, which gives a high  $Q$  and makes it possible to measure larger electron concentrations than in the case of other modes. The  $Q$  of the slotted resonator with the quartz bulb was  $Q \approx 10^4$ . The dimensions of the resonator were: radius  $R_0 = 2.25$  cm, length  $L_0 = 3$  cm. The natural frequency of the empty resonator, calculated from its dimensions, was  $f_{00} = 9.6 \times 10^9$  Hz. The frequency of the resonator with the bulb, measured at nitrogen temperature, was  $f_0 = 8.9 \times 10^9$  Hz and the deviations from  $f_0$  did not exceed  $\pm 1\%$  following variation from room to helium temperature. A detailed description of the experimental setup was given in [28].

The excitation of the plasma in the vessel leads to a shift of the resonant frequency, determined by the formula

$$f^2 - f_0^2 = \frac{\langle n \rangle e^2}{\pi m \epsilon}, \quad \langle n \rangle = \int_{\text{pl}} n E^2 dV / \int_{\text{res}} E^2 dV, \quad (6)$$

where  $\epsilon = (f_{00}/f_0)^2 = 1.16 \pm 1\%$  is the effective value of the distributed dielectric constant of the real resonator with the bulb,  $e$  and  $m$  are the charge and mass of the electron,  $E$  is the intensity of the electric field in the resonator,  $\langle n \rangle$  is the measured average electron density, and the index under the integral sign indicates the volume of integration. Relation (6) is valid when it is possible to neglect the change of the resonator  $Q$  as a result of the collisions between the electrons and the atoms, i.e., when  $(\nu_{ea}/2\pi f_0)^2 \ll 1$ . Here  $\nu_{ea} = N \nu_{ea} = 1.08 \times 10^7 P \sqrt{T_e}$  [sec $^{-1}$ ] is the frequency of the collisions between the electrons and the atoms,  $\sigma_{ea} = 5.3 \times 10^{-16}$  cm $^2$  is the corresponding cross section,  $\nu_e = (8\kappa T_e/\pi m)^{1/2}$  and  $T_e$  are the velocity and temperature of the electrons and  $\kappa$  is Boltzmann's constant. Even under the extreme conditions of our experiments, at  $P = 30$  mm Hg and  $T_e = 500^\circ\text{K}$ , the correction  $(\nu_{ea}/2\pi f_0)^2 = 1.7 \times 10^{-2}$  is small and does not play any role in the measurement of the shift of the resonant frequency. For small shifts  $\Delta f = f - f_0 \ll f_0$  formula (6) simplifies and gives

$$\langle n \rangle = \frac{2\pi m \epsilon f_0}{e^2} \Delta f = 2.6 \cdot 10^8 \Delta f \text{ [cm}^{-3}\text{]} \quad (7)$$

( $\Delta f$  is in MHz).

The determination of the true recombination coefficient from the measurement of the kinetics of the averaged density  $\langle n \rangle$  is a rather complicated problem, owing to the inhomogeneous and time-varying distribution of the electrons inside the discharge vessel, and also to the inhomogeneous distribution of the field in the resonator. The values of the recombination coefficients obtained from such measurements depend on how realistic is the method of recalculating  $\langle n \rangle$  to other mean values of  $n$ , for which the inhomogeneities of the plasma and the microwave field play a minimal role. In those cases when the volume of the plasma  $V_{\text{pl}}$  is much smaller than the volume of the resonator  $V_{\text{res}}$ , the inhomogeneity of the field in the plasma is small, and it is meaningful to recalculate the measured quantity  $\langle n \rangle$  to the value averaged over the volume of the plasma

$$\bar{n} = \int_{\text{pl}} n(r) dV / V_{\text{pl}}.$$

Such a method of calculation was described and used by Golant and co-workers. [29, 30] Extensive use is also made of recalculation from  $\langle n \rangle$  to the "microwave mean"  $\bar{n}$ , equal to

$$\bar{n} = \int_{\text{pl}} n E^2 dV / \int_{\text{pl}} E^2 dV = G \langle n \rangle, \quad (8)$$

$$G = \int_{\text{res}} E^2 dV / \int_{\text{pl}} E^2 dV,$$

where  $G$  is a geometric factor that depends on the oscillation mode and on the ratio of the dimensions of the plasma and of the resonator.

Grey and Kerr [31] and later Frommhold and Biondi [32] carried out detailed numerical calculations of the nonlinear equation describing the kinetics of the electrons  $n(\mathbf{r}, t)$  in two-particle recombination and ambipolar diffusion for different initial distributions  $n(\mathbf{r}, 0)$ , oscillation modes, and geometrical relations between the dimensions of the resonator and the discharge vessel. They present plots of the corrections  $\Delta$ , with the aid of which it is possible to obtain the "true" recombination coefficients

$$\alpha = a(1 - \Delta) \quad (9)$$

from the coefficients  $a$  determined from the kinetics of the "microwave mean"  $\bar{n}$ ,

$$\frac{1}{\bar{n}(t)} = \frac{1}{\bar{n}(t_0)} + a(t - t_0). \quad (10)$$

The magnitude of the corrections  $\Delta$  depends on the length of the interval of the linear variation of  $\bar{n}^{-1}(t)$ , and for a given geometry and oscillation mode it is determined by the quantity  $F$ , which equals the ratio of the electron concentrations on the boundaries of this interval.

In our experimental setup, the geometric factor turns out to be  $G = 1 \pm 0.01$ , and therefore the measured mean value  $\langle n \rangle$  coincides with the "microwave mean"  $\bar{n}$ . Subsequently, the measurements of the shift of the resonant frequency  $\Delta f$  were reconverted into concentrations  $\bar{n}$  by means of formula (7), and the results of [32] were used in the calculation of the recombination coefficients  $\alpha$ . It must be borne in mind, however, that the applicability of these results to real experiments is limited. The numerical analysis in [31, 32] does not take into ac-

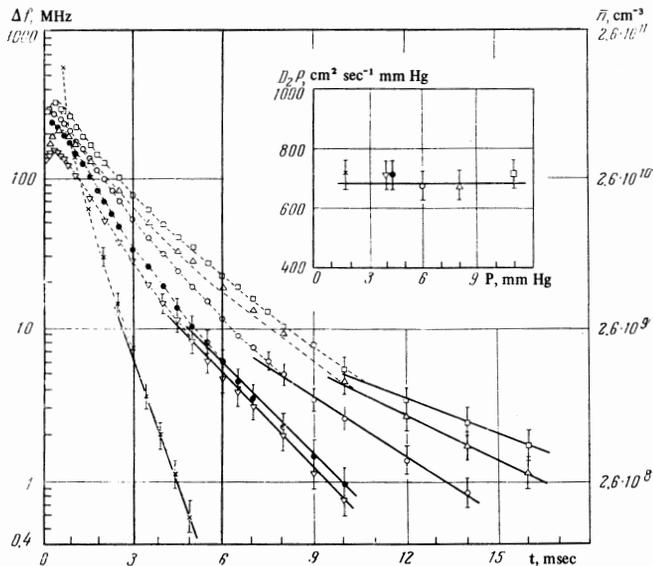


FIG. 3. Change of the shift of the resonant frequency  $\Delta f$  (of the electron density  $\bar{n} = 2.6 \times 10^8 \Delta f$ , where  $\Delta f$  is in MHz) in the afterglow of a helium plasma at 300°K;  $\lambda = 0.61$  cm. The experimental conditions are:

P, mm Hg	$\tau$ , msec	$\nu$ , sec <sup>-1</sup>
×	1.7	1150 ± 80
∇	4	477 ± 30
●	4.3	450 ± 30
○	6	303 ± 20
△	8	228 ± 15
□	11	176 ± 10

The figure in the upper right shows a plot of  $D_2P$  against  $P$ , obtained from the relation  $D_2P = \nu P \lambda^2$ . The straight line corresponds to  $D_2P = 680 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ .

count many factors, such as the change of the electron temperature in the afterglow and the additional ionization, and was performed only for two-particle recombination. Therefore even the introduction of the correction  $\Delta$  does not make it possible to obtain the exact value of the "true" recombination coefficient. One can only assume that when the parameter  $F \gtrsim 10$  and the correction  $\Delta$  is small we obtain a reasonable estimate of the coefficient  $\alpha$ . In this respect, it is much simpler to obtain the rate of diffusion variation of the electron density in the late afterglow, since it is not necessary here to know the absolute concentration and it suffices to determine the argument of the exponential function in  $\Delta f(t)$ .

The range of measured mean electron concentrations is limited from above by the critical density  $n_c = \pi m \epsilon_0^2 / e^2 = 1.1 \times 10^{12} \text{ cm}^{-3}$  and from below by the accuracy with which the shift of the resonant frequency is measured, and amounts to  $\bar{n} \approx (1-2) \times 10^8 \text{ cm}^{-3}$ . In the case of diffusion distribution of the electrons, the maximum concentration at the center of the vessel is larger by 2.5 times than the mean density  $\bar{n}$ .<sup>[28]</sup>

Let us point out some features of measurements at low temperatures. If the cryogenic liquid falls between the discharge bulb and the resonator (especially in the case of liquid nitrogen), the structure of the microwave field becomes distorted and harmful modes appear. The measurements at nitrogen temperatures were therefore performed in the vapor of the nitrogen over the liquid. The measured temperature of the bulb walls was  $100 \pm 5^\circ \text{K}$ . At helium temperatures, the resonator was im-

mersed directly in the liquid helium, but no liquid helium entered the resonator. The estimate of the gas temperature from the periods of the acoustic oscillations gave a value  $T = 10 \pm 3^\circ \text{K}$ .

### 3. EXPERIMENTAL RESULTS

1. Room temperature ( $T = 293^\circ \text{K}$ ). Figure 3 shows on a semilog scale the results of measurements of the shift of the resonant frequency of the resonator with the plasma at room temperature of the gas and at different pressures. The experimental conditions are given in the caption of the figure. In the region of the late afterglow, starting with a concentration  $\bar{n} = (2.5-1.3) \times 10^8 \text{ cm}^{-3}$ , the  $\bar{n}(t)$  curves become exponential, pointing to a diffusion character of the decay of the electron component. This is also confirmed by the fact that the slopes of the straight lines in the semilog scale  $\nu = -\partial \ln n / \partial t$  are inversely proportional to the pressure. In the right corner of Fig. 3 are shown the constants of the ambipolar diffusion  $\nu P \lambda^2 = DP$ . We see that the points lie near the straight line  $D_2P = 680 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ , which corresponds to the known value of the coefficient of ambipolar diffusion of the diatomic helium ion  $\text{He}_2^+$  at 293°K.<sup>[33]</sup>

At the initial instants of time, in the region of 0.2–0.4 msec after the termination of the excitation pulse, one can see clearly the maxima of the electron density connected with the processes of additional ionization. Figure 4 shows on a large scale this and the adjacent region of recombination decay in coordinates  $\bar{n}^{-1}$  and  $t$ . The interval of the linear time dependence of  $\bar{n}^{-1}$  is small here. A near-linear  $\bar{n}^{-1}(t)$  dependence is observed no farther than 2–2.5 msec, and in this case the electron concentration decreases by a factor 2–3 (the parameter  $F = 2-3$ ). At high pressures,  $P = 12-16 \text{ mm Hg}$  there exists an extensive region, up to the start of the diffusion breakdown, in which  $\bar{n}^{-1}(t)$  increases with time more rapidly than linearly. If we attempt nevertheless to determine the recombination coefficients directly beyond the maximum of the electron density, then it turns out that the observed values of these coefficients are of the order of a  $\approx (0.8-1) \times 10^{-8} \text{ cm}^3 \text{ sec}^{-1}$ . The

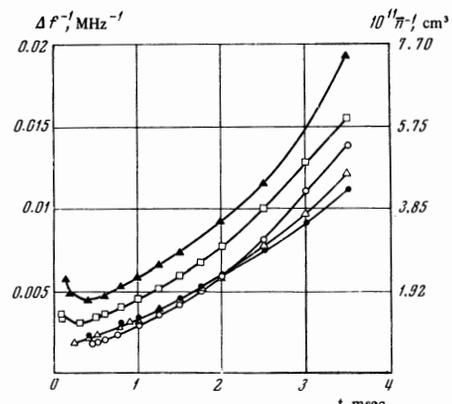


FIG. 4. Dependence of the reciprocal electron density on the time in the afterglow of a helium plasma at 300°K. Excitation-pulse duration  $\tau = 15 \text{ msec}$ . Experimental points: ○— $P = 6$ ,  $u = 100$ ; ▲— $P = 8$ ,  $u = 40$ ; □— $P = 11$ ,  $u = 60$ ; △— $P = 12$ ,  $u = 100$ ; ●— $P = 16$ ,  $u = 100$ . The voltages  $u$  are given in relative units, and the pressure  $P$  in millimeters of mercury.

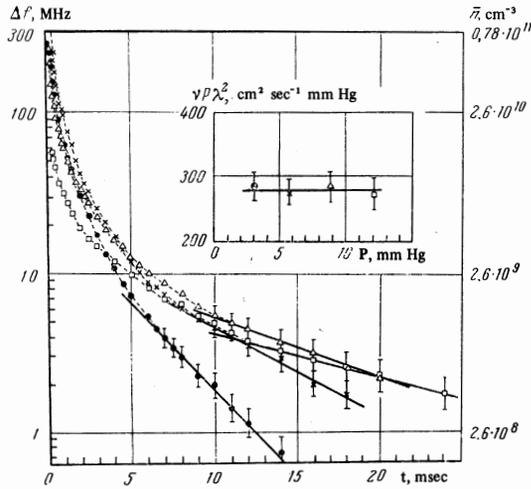


FIG. 5. Change of the shift of the resonant frequency (electron density) in the afterglow of a helium plasma cooled with liquid-nitrogen vapor ( $T = 100 \pm 5^\circ\text{K}$ ) under different experimental conditions:

P, mm Hg	u, rel. un.	$\tau$ , msec	$\nu$ , sec <sup>-1</sup>
● 3.1	40	35	$250 \pm 14$
× 5.7	100	35	$130 \pm 10$
△ 8.8	100	35	$87 \pm 8$
□ 12	40	10	$61.5 \pm 7$

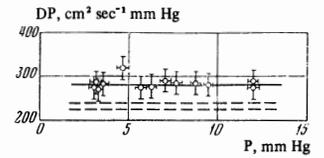
In the upper right is shown the dependence of  $\nu P \lambda^2$  on P. The straight line corresponds to  $\nu P \lambda^2 = 280 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ ,  $\lambda^{-2} = 2.7 \text{ cm}^{-2}$ .

corrections  $\Delta$ , according to [32], reach 40–60% for the initial diffusion distribution of the electron density. Such an initial distribution  $n(r, 0)$  is realized because of the high electron temperatures in the discharge ( $T_e \sim 5\text{--}10 \text{ eV}$ ). Consequently, the corrected values of the recombination coefficients can be estimated at  $\alpha \approx (4\text{--}5) \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$ . We note that these values are close to the value  $k_e = 4.5 \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$ —the coefficient of disintegration of the metastable  $2^3\text{SHe}$  atoms in the case of superelastic collision with electrons. [34]

2. Nitrogen temperatures ( $T = 100^\circ\text{K}$ ). Figure 5 shows the time variation of the electron density when a helium plasma is cooled with liquid-nitrogen vapor. The region of diffusion decay shifts towards longer times and the time interval of recombination disintegration broadens accordingly. The ambipolar diffusion begins in the region of concentrations  $\bar{n} \approx 10^9 \text{ cm}^{-3}$  and is noticeably slower than at room temperature. In the right corner of Fig. 5 is shown the ambipolar diffusion constant  $\nu P \lambda^2$  as a function of the pressure. We see that the slopes  $\nu$  are inversely proportional to P.

In Fig. 6 are gathered the results of all the measurements of the diffusion coefficient. The experimental points fit with good accuracy the straight line  $D_3P = 280 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ . As shown in our paper, [25] this value of the diffusion coefficient corresponds to the triatomic molecular helium ion  $\text{He}_3^+$ . The dashed lines  $DP = 238$  and  $DP = 225 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ , between which the probable value of  $D_2P$  for the diatomic  $\text{He}_2^+$  ion is located, lie noticeably below and beyond the limits of the maximum error of the values of DP measured by us. In order to assign a value  $DP = 280 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$  to the  $\text{He}^{2+}$  ion, it would be necessary to assume that the electron temperature  $T_e$  is 1.5 times higher than the gas temperature  $T = 100^\circ\text{K}$  and remains constant

FIG. 6. Coefficient of ambipolar diffusion of helium ions in a plasma at  $T_e = 100^\circ\text{K}$ . The solid straight line corresponds to  $D_3P = 280 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ . The dashed lines correspond to  $DP = 238$  (upper) and  $DP = 225$  (lower) (in  $\text{cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ ).



even 10–15 msec after the termination of the excitation pulse. We consider such an assumption to be unlikely at nitrogen temperatures.

Figure 7 illustrates the dependence of the reciprocal electron density on the time at different pressures. One can clearly see linear  $\bar{n}^{-1}(t)$  dependences. At high pressures  $P \gtrsim 10 \text{ mm Hg}$ , the electron concentration changes by 10–20 times within the limits of the linear law (10). This makes it possible to determine the recombination coefficient with satisfactory accuracy.

In Table I are gathered the results of several series of measurements in the pressure interval from 4 to 30 mm Hg under different excitation conditions, characterized by initial concentrations  $\bar{n}_0$  (at  $t_0 = 200 \mu\text{sec}$ ). The table lists the observed recombination coefficients  $a'$  (in  $\text{MHz}^{-1} \text{ sec}^{-1}$ ) and  $a$  (in  $\text{cm}^3 \text{ sec}^{-1}$ ), calculated with the aid of (7) from  $a'$ . In the last column of Table I are given the recombination coefficients corrected in accordance with formula (9). It is seen that in the investigated band there is no dependence of  $\alpha$  on the pressure. The average of all the measurements is  $\alpha_{av} = (5.8 \pm 1.5) \times 10^{-8} \text{ cm}^3 \text{ sec}^{-1}$ .

3. Helium temperatures. At these temperatures in the early afterglow, up to 2–3 msec, one observes low-frequency modulation of the reflected signal. In Fig. 2b one sees on both sides of the resonant curve the lateral maxima connected with the oscillations of the electron density. The periods of the oscillations, depending on the excitation conditions, have a value  $\Delta T = 150\text{--}$

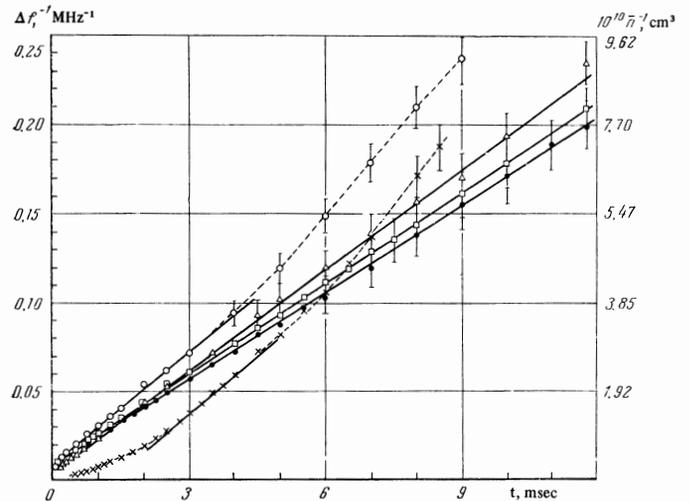


FIG. 7. Dependence of the reciprocal shift of the resonant frequency  $\Delta f^{-1}$  (of the electron density  $\bar{n}^{-1}$ ) on the time in the afterglow of a helium plasma cooled with liquid-nitrogen vapor ( $T = 100^\circ\text{K}$ ):

P, mm Hg	u, rel. un.	$\tau$ , msec
× 6.3	100	40
○ 9.5	40	20
△ 16.4	100	40
● 22.8	100	20
□ 27.3	100	40

Table I. Recombination coefficient  $\alpha$  for a plasma cooled to  $T = 100^\circ\text{K}$

No.	P, mm Hg	$n_0 \cdot 10^{-10}$ ( $t_0 = 0.2$ msec)	$\alpha'$ , MHz <sup>-1</sup> sec <sup>-1</sup>	$\alpha \cdot 10^8$ , cm <sup>3</sup> sec <sup>-1</sup>	F	A, %	$\alpha \cdot 10^8$ , cm <sup>3</sup> sec <sup>-1</sup>
1	4.7	3.0	20.5	7.5	3.7	27	5.5
2	6.3	10.0	22	8.5	4.1	26	6.3
3	7.1	2.2	23	8.75	9	15	7.4
4	7.7	8.7	19	7.3	3	30	5.1
5	8.8	6.8	13.8	5.3	5	22	4.2
6	9.5	2.6	20	7.7	11	13	6.8
7	12	6.0	17	6.5	75	17	5.4
8	12	2.2	18.7	7.25	22	11	6.5
9	12.5	3.3	17.4	6.7	10	15	5.7
10	15.5	2.6	18	6.9	22	11	6.2
11	16.4	2.9	18.9	7.25	10	15	6.2
12	17.6	2.5	15.5	6	11	13	5.2
13	19.5	2.6	18.4	7	17.5	12	6.2
14	22	1.6	16.2	6.25	10	15	5.3
15	23.5	1.8	17	6.5	20	11	5.8
16	27.3	2.6	17.1	6.6	23	10	5.9
17	28	2.1	16	6.25	33	8	5.75
18	29	2.2	15.7	6.0	33	8	5.5

$$\alpha_{av} = (5.8 \pm 1.5) \cdot 10^{-8}$$

200  $\mu\text{sec}$ . These oscillations are connected with the participation of the electrons in the acoustic oscillations of the neutral gas, excited at the instant of the breakdown in the cryogenic plasma.<sup>[35]</sup> From the periods  $\Delta T$  ( $\mu\text{sec}$ ) = 560  $T^{-1/2}$  [28] it is possible to determine the gas temperature and to estimate the rate of its cooling. Such estimates give a value  $T = 10 \pm 3^\circ\text{K}$  several milliseconds after the termination of the pulse.

Figure 8 shows the dependence of the shift of the resonant frequency on the time in a semilogarithmic scale. The pressures are referred to 293°K, starting with  $T = 10^\circ\text{K}$ . The inaccuracy in the determination of  $T$ , of course, leads to an appreciable error (on the order of 30%) in the values of the reduced pressures (gas densities). The initial electron concentrations are lower here than at 100°K. In the early afterglow, the recombination decay of the plasma is more rapid, and in the later afterglow the diffusion is slower than at 100°K. At low pressures, the decrease of the electron density in the low-concentration region can be approximately described by an exponential whose argument is assumed

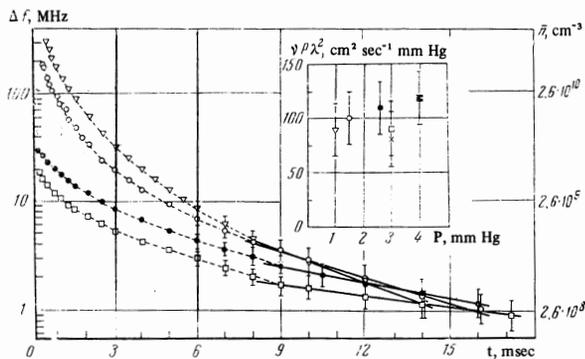


FIG. 8. Change of the shift of the resonant frequency (electron density) in the afterglow of a helium plasma upon cooling with liquid helium ( $T = 10 \pm 3^\circ\text{K}$ ; the pressures are referred to 293°K):

P, mm Hg	u, rel. un.	$\nu$ , sec <sup>-1</sup>
▽	1	240
○	1.5	60
●	2.6	100
□	3	40

In the upper right corner is given the dependence of  $\nu P \lambda^2$  on  $P$ . The average value is  $\nu P \lambda^2 \approx 90 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ .

to vary slowly with time. This approximation corresponds to very slow cooling of the plasma electrons when the plasma is cooled with liquid helium. Figure 8 shows also the values of  $\nu P \lambda^2$  at several pressures from 1 to 3 mm Hg. The approximate average is  $\nu P \lambda^2 \approx 90 \pm 30 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$ .

Figure 9 shows the region of recombination variation of the electron density in coordinates  $\bar{n}^{-1}$  and  $t$  at 10°K. We see that the curves for  $P = 1$  and 3 mm Hg deviate from straight lines much earlier than at higher pres-

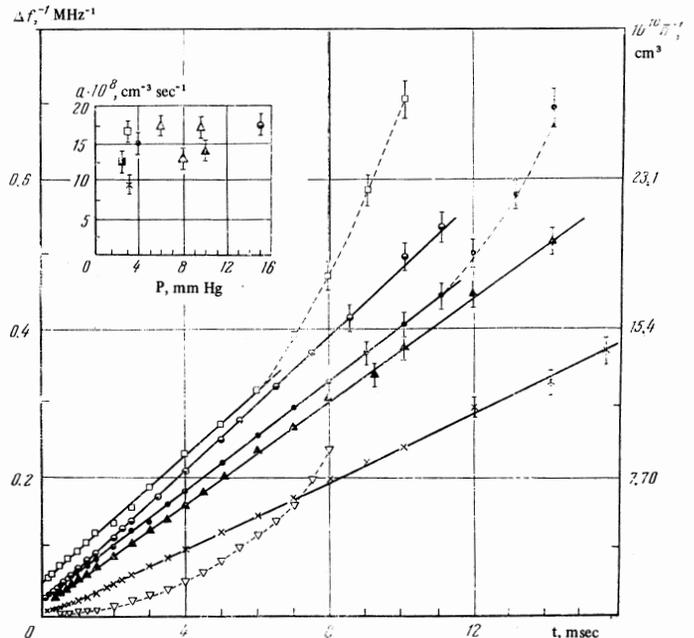


FIG. 9. Time dependence of the reciprocal shift of the resonant frequency  $\Delta f^{-1}$  (of the electron density  $\bar{n}^{-1} = 3.85 \times 10^{-10} \Delta f^{-1}$ ) in the afterglow of a helium plasma cooled with liquid helium ( $T = 10 \pm 3^\circ\text{K}$ ).

P, mm Hg	u, rel. un.	$\tau$ , msec
▽	1	40
×	3	100
□	3	40
●	4	60
○	10	100
▲	15	80

The pressures are referred to 293°K. In the left corner is given the dependence of the observed recombination coefficient  $\alpha$  on the pressure.

**Table II.** Recombination coefficient  $\alpha$  for a plasma cooled with liquid helium

No.	$P$ , mm Hg	$\bar{n}_e \cdot 10^{-10}$ ( $t_0 =$ $= 0.2$ msec)	$\alpha'$ , MHz <sup>-1</sup> sec <sup>-1</sup>	$\alpha \cdot 10^8$ , cm <sup>3</sup> sec <sup>-1</sup>	$F$	$\Delta$ , %	$\alpha \cdot 10^8$ , cm <sup>3</sup> sec <sup>-1</sup>
1	2.6	0.8	32.5	12.5	5.3	21	10
2	3	0.50	43.5	16.7	6.5	18	15.7
3	3	2.6	24	9.25	10	10	8.3
4	4	0.8	39	15	10	15	12.7
5	6.3	0.6	45	17.3	6.5	18	14.2
6	8	0.8	34	13	20	11	11.6
7	9.6	0.6	45	17.3	16	12	15.2
8	10	1.6	36.6	14	23	10	12.6
9	15	0.8	45.5	17.5	20	11	15.5

$$\alpha_{av} \approx (1.3 \pm 0.3) \cdot 10^{-7}$$

tures, corresponding to a transition to the diffusion disintegration of the plasma at low pressures. The higher the pressure and the higher the initial electron concentration, the broader the interval of the observed linear dependence (10). Figure 9 also gives the observed recombination rates  $\alpha$  as functions of  $P$ . No dependence of  $\alpha$  on the pressure is revealed.

In Table II are summarized the results of the measurements of  $\alpha$  and the coefficients  $\alpha$  corrected in accordance with (9) in the pressure range from 2 to 15 mm Hg. Owing to the large value of the parameter  $F$ , the corrections to the observed quantities amount to 10–15%. The average over all the experimental data is  $\alpha_{av} = (1.3 \pm 0.3) \times 10^{-7}$  cm<sup>3</sup> sec<sup>-1</sup>.

#### 4. ANALYSIS OF DIFFUSION DECAY OF PLASMA

The exponential decrease of the electron density with time observed in the region of small concentrations  $\bar{n}$  should be regarded as a result of ambipolar diffusion. The diffusion rate  $\nu$  depends on the type of the helium ions that predominate in the plasma at the given pressure and temperature of the neutral gas.

At room temperature and low pressures, the positive component of the plasma consists mainly of the atomic ions He<sup>+</sup>. These ions become molecular in accordance with the scheme (1) at a rate  $\nu_C = \gamma P^2$ , and at high pressures the molecular ions play the predominant role in the later afterglow. The critical value of  $P_0$  at which the attenuation of the atomic ions in the ambipolar diffusion and conversion ( $\nu_1 + \gamma P^2$ ) becomes comparable with the rate of diffusion  $\nu_2$  of the molecular ions is determined by the formula

$$P_0 = \left( \frac{\nu_2 P - \nu_1 P}{\gamma} \right)^{1/2} = \left( \frac{D_2 P - D_1 P}{\gamma \lambda^2} \right)^{1/2}. \quad (11)$$

The value of  $DP$  measured by us in the pressure range from 1.7 to 11 mm Hg is  $680 \pm 35$  cm<sup>2</sup> sec<sup>-1</sup> mm Hg and agrees with the published constant of ambipolar diffusion for the diatomic ion. If we use this value of  $D_2 P$  in (11) and also the ambipolar-diffusion constant of the He<sup>+</sup> ion  $D_1 P = 465$ ,  $\gamma = 100$  sec<sup>-1</sup> (mm-Hg)<sup>-2</sup> and  $\lambda = 0.61$  cm we obtain  $P_0 = 1.8$  mm Hg. Such a value of the critical pressure shows that under the conditions of our experiment the diatomic ions He<sub>2</sub><sup>+</sup> actually predominate in the later decay of the plasma at room temperatures.

The ambipolar diffusion coefficient  $DP$  is connected with the ion mobility by the Einstein relation

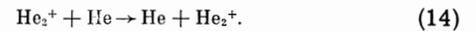
$$K = \frac{e}{\kappa} \frac{DP}{(T_e + T)} = 15.27 \frac{(DP)_{273}}{T_e + T} = 14.2 \frac{DP}{T_e + T}, \quad (12)$$

where  $K$  (cm<sup>2</sup> V<sup>-1</sup> sec<sup>-1</sup>) is the ion mobility reduced to normal conditions ( $T = 273^\circ$  K,  $P = 760$  mm Hg),  $(DP)_{273}$  is reduced to  $T = 273^\circ$  K, and  $\kappa$  is Boltzmann's constant. For a diatomic ion with  $D_2 P = 680 \pm 35$  we obtain  $K_2 = 16.3 \pm 0.7$  cm<sup>2</sup> V<sup>-1</sup> sec<sup>-1</sup>.

From the value of  $DP$  or from the corresponding mobility  $K$  we can determine the coefficient of mutual ion-atomic diffusion  $D_{id} = TD/(T + T_e)$  and the corresponding cross section  $Q_{id}$  averaged over the Maxwellian distribution of the atom velocities. Using the well-known expression of Chapman and Enskog<sup>[33]</sup> for  $D_{id}$  and formula (12), we obtain

$$Q_{id} = \frac{3\pi}{32} \left( \frac{8\kappa T}{\pi\mu} \right)^{1/2} \frac{293}{T} \frac{e}{K} = \frac{2.38 \cdot 10^{-21}}{K(\mu T)^{1/2}}, \quad (13)$$

where  $Q_{id}$  is in cm<sup>2</sup> and  $\mu$  is the reduced mass of the ion and atom. At  $T = 293^\circ$  K we obtain for the diatomic ion He<sub>2</sub><sup>+</sup> from our experimental data  $Q_{2d} = (4.05 \pm 0.25) \times 10^{-15}$  cm<sup>2</sup>. We present also the cross section  $Q_{1d}$  for a monatomic ion He<sup>+</sup> with mobility  $K = 11.3$ , (corresponding to  $DP = 465$ ), namely,  $Q_{1d} = (6.75 \pm 0.25) \times 10^{-15}$  cm<sup>2</sup>. The fact that the cross sections for the mutual ion-atomic diffusion for He<sup>+</sup> and He<sub>2</sub><sup>+</sup> differ from each other, with  $Q_{1d} > Q_{2d}$ , is connected with the fact that these cross sections are determined by two processes. One is the polarization attraction between the ion and the atom, which is proportional to  $R^{-4}$ , and the other is the process of resonant charge exchange of the atomic ion on the atom, He<sup>+</sup> + He = He + He<sup>+</sup>, or the process of resonant ion-atomic exchange of the type



The cross section of the polarization interaction  $Q^*$  does not depend on the mass of the ion and is determined only by the polarizability  $\beta$  of the gas atoms and by the temperature:<sup>[3]</sup>

$$Q^* = 5.15 \cdot 10^{-14} (\beta / T)^{1/2} [\text{cm}^2]. \quad (15)$$

For helium,  $\beta = 1.39$  atomic units. At  $T = 293^\circ$  K, the polarization cross section is  $Q^* = 3.55 \times 10^{-15}$  cm<sup>2</sup>, and at  $T = 100^\circ$  K we have  $Q^* = 6.1 \times 10^{-15}$  cm<sup>2</sup>.

The cross section of resonant charge exchange  $Q_{tr}$  of the atomic ion He<sup>+</sup> can be calculated from an interpolation formula on the basis of the approximate representation of Firsov<sup>[36]</sup> for  $Q_{tr} = (a - b \log v)^2$ . The entire aggregate of the experimental data on the charge exchange in helium<sup>[3, 33]</sup> can be described by using the values  $a = 12.6 \times 10^{-8}$  cm and  $b = 1.33 \times 10^{-8}$  cm. For  $T = 293^\circ$  K and a relative velocity  $v = 1.7 \times 10^5$  cm/sec we get  $Q_{tr} = 3 \times 10^{-15}$  cm<sup>2</sup>. In the quasiclassical limit,

the diffusion cross section in the process of resonant charge exchange is determined by double the cross section  $Q_{tr}$  and should be of the order of  $2Q_{tr} = 6 \times 10^{-15} \text{ cm}^2$ . We see that for a monatomic ion the contribution made to the diffusion by the process of resonant charge exchange is almost twice as large as the contribution from the polarization interaction. Therefore the process of diffusion scattering of monatomic ions at 293°K with an experimental cross section  $Q_{1d} = 6.75 \times 10^{-15} \text{ cm}^2$  can be attributed mainly to charge exchange.

The resonant cross section (14) for the molecular ion  $\text{He}_2^+$  has unfortunately not been measured. It is quite obvious that this cross section should be much smaller than for monatomic ions. In the case of  $\text{He}_2^+$  a comparison of the experimental cross section ( $Q_{2d} = 4.05 \times 10^{-15} \text{ cm}^2$ ) with the polarization cross section ( $Q^* = 3.55 \times 10^{-15} \text{ cm}^2$ ) shows that the main contribution to the diffusion cross section is made here by the polarization interaction. Since  $Q^* \propto T^{-1/2}$  and the cross section  $Q_{tr}$  increases very slowly with decreasing temperature, the relative role of the polarization interaction and the process of diffusion scattering should even increase at low temperatures. Therefore the mobility of the molecular ions  $\text{He}_2^+$  should not change in practice with changing temperature. This is confirmed by experimental measurements of the mobility of the ions  $\text{He}_2^+$ , carried out by Beatty and Patterson.<sup>[37, 24]</sup> In the later paper<sup>[24]</sup> it is stated that the mobility of  $\text{He}_2^+$  remains constant in the entire temperature interval from 300 to 76°K, and the values cited (in  $\text{cm}^2 \text{ V}^{-1} \text{ sec}^{-1}$ ) are 16.7<sup>[37]</sup> and 16.0.<sup>[24]</sup> These values are close to the value  $K_2 = 16.3 \pm 0.7$  measured by us at 293°K and to the estimate given above,  $K_2 = 16.9$ . The dashed lines  $DP = 238$  and  $225 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$  in Fig. 6 correspond to the extreme values of the mobilities 16.9 and 16.0.

The experimental value of DP measured at 100°K and for pressures from 3 to 12 mm Hg turns out to be  $280 \pm 15 \text{ cm}^2 \text{ sec}^{-1} \text{ mm Hg}$  and is much higher than the value expected for the diatomic ion, the difference exceeding the maximum measurement error. This means that the diatomic  $\text{He}_2^+$  ions do not predominate in a helium plasma at 100°K. Taking into account the large probability of production of polyatomic ion-molecular complexes with low binding energy with decreasing temperature, we ascribe this value to the triatomic ion  $\text{He}_3^+$ .<sup>[25]</sup> The mobility corresponding to the experimental value  $D_3P = 280$  is  $K_3 = 19.8 \pm 1$ , and the cross section is  $Q_{3d} = (5.4 \pm 0.25) \times 10^{-15} \text{ cm}^2$ . Ions with a mobility close to this were observed recently by Patterson in the temperature region from 200 to 135°K; these ions were observed simultaneously with the diatomic ions  $\text{He}_2^+$  and were in equilibrium with the latter. Patterson regards this new type of ion as triatomic and obtains for it a dissociation energy  $0.17 \pm 0.03 \text{ eV}$ . The cross section for ion-atomic diffusion for  $\text{He}_3^+$  should not contain an increment due to the resonant exchange process (14), since the probability of such a reaction is even lower for  $\text{He}_3^+$  than for  $\text{He}_2^+$ . The pure polarization cross section at 100°K is  $Q^* = 6.1 \times 10^{-15} \text{ cm}^2$ , and the  $\text{He}_3^+$  mobility determined by this cross section is  $K_3 = 17.6$ . For the triatomic ion, unlike  $\text{He}_2^+$ , the experimental cross section turns out to be smaller than the calculated cross section  $Q^*$  (by a factor 1.13). Such differ-

ences are typical of the cross sections for the diffusion of ions in a foreign gas.<sup>[37]</sup> This means that besides attraction there is also a short-range repulsion in the energy of interaction between the  $\text{He}_3^+$  ion and the atom.

Let us proceed to discuss the data in the region of helium temperatures. Here the probability of conversion of the  $\text{He}_2^+$  into  $\text{He}_3^+$  is even higher than at 100°K. Moreover, the formation of even heavier molecular ions is quite probable. The difference between the mobilities of these ions is small and decreases with increasing ion mass. Whereas the mobility of the  $\text{He}_3^+$  ion in the case of pure polarization interaction is  $K_3^* = 17.6$ , for  $\text{He}_4^+$  it amounts to  $K_4^* = 17$ , and for the infinitely heavy ion  $\text{He}_\infty^+$  it amounts to  $K_\infty^* = 15.1$ . This is connected with the fact that the mobility of the heavy ions depends only on the reduced mass of the atom and the ion. Since the repulsion increases K somewhat, it is difficult to differentiate experimentally between different heavy ions by their mobilities or by the values of DP. Therefore in the following analysis of the data we shall use the values of K and DP measured for the triatomic ion.

It was indicated in the preceding section that the later decay of the plasma at 10°K can be described by ambipolar diffusion with a slowly varying exponent  $\nu$ . The measured value is  $D_3P = 90$ , corresponding to  $T_e + T \sim 60^\circ \text{ K}$  and  $T_e \sim 50^\circ \text{ K}$ . The diffusion character of the plasma decay at low concentrations (see Fig. 8) is confirmed by the following estimate. The recombination disintegration is less than the diffusion disintegration at  $n < \nu/\alpha$ . Using the experimental value  $\alpha = 1.3 \times 10^{-7}$  and  $\nu P = 270$ , we have  $n < 2.1 \times 10^9/P$ , corresponding to  $\Delta f < 8P^{-1} [\text{MHz}]$  and coinciding with the observed diffusion region  $\Delta f < 3 \text{ MHz}$  on Fig. 8 (for pressures 1–3 mm Hg). Thus, even 15–20 msec after the termination of the excitation pulse the electron temperature in the plasma exceeds the gas temperature and amounts to several times 10°. The noticeable heating of the electron component in the later afterglow can be attributed to the production of "hot" electrons in superelastic collisions with metastable  $2^3\text{S}$  atoms, the concentration M of which decreases exceedingly slowly. The main role in the disintegration of the metastable atoms is played by diffusion, since at small electron concentrations  $n \lesssim 10^9 \text{ cm}^{-3}$  the rate of superelastic disintegration  $k_e n = 4.5 \text{ sec}^{-1}$  is much smaller than the rate of the diffusion disintegration of the metastable atoms  $D_M \lambda^{-2} \sim 80/P [\text{sec}^{-1}]$ . ( $k_e = 4.5 \times 10^{-9} \text{ cm}^2 \text{ sec}^{-1}$  is the superelastic collision constant,<sup>[34]</sup>  $D_M = 30/P [\text{cm}^2 \text{ sec}^{-1}]$  is the diffusion coefficient of the metastable atoms in the region of 10°K.<sup>[38]</sup>) Therefore the characteristic time of the decrease of M amounts to several times 10 milliseconds for pressures 1–3 mm Hg, and consequently the concentration M remains approximately constant in the interval 5–10 msec.

The electron heating was analyzed in<sup>[34]</sup>, where it was shown that their cooling is due mainly to collisions with atoms, and the most significant heating is connected with superelastic collisions of the electrons with the  $2^3\text{S}$  atoms of helium, in which an excitation energy  $E_{exc} = 19.8 \text{ eV}$  is released. Therefore the equation for the electron temperature is

$$\nu_{ea}^*(T_e - T) = k_e M E_n, \quad (16)$$

where  $\nu_{ea}^* = (2m/M)\nu_{ea} = 6.7 \times 10^3 P T_e^{1/2} \text{ sec}^{-1}$  is the averaged "energy" electron-atom collision frequency.

If we substitute in (16) the numerical values of the parameters,  $T_e = 50^\circ\text{K}$ , then we obtain for the concentration  $M$  a value of the order of  $(2-5) \times 10^9 \text{ cm}^{-3}$  for pressures 1-3 mm Hg. Such values of the concentration of the metastable atoms in the late afterglow are reasonable and agree with our measurements of the absolute concentrations  $M$ , which usually turn out to be several times larger than the electron density at helium temperature.<sup>[34]</sup>

A similar estimate of the heating of the electrons at  $100^\circ\text{K}$  shows that in the region of ambipolar diffusion from  $P = 3$  to  $P = 12$  mm Hg (Fig. 5)  $T_e$  exceeds  $T$  by not more than  $10^\circ$ . The diffusion coefficient  $D_M$  is here approximately seven times higher than at  $10^\circ\text{K}$ ; the concentration  $M$  decreases much more rapidly, and as a rule is of the order of or not much higher than  $n$ . In other words, in the late afterglow at  $100^\circ\text{K}$  the concentration is  $M \lesssim 10^9 \text{ cm}^{-3}$ . Assuming  $M = 10^9 \text{ cm}^{-3}$ , we get from (16)  $T_e - T \approx 15/P \sim 5-2^\circ\text{K}$ .

## 5. DISCUSSION OF RECOMBINATION PROCESSES AT LOW TEMPERATURES

Let us discuss briefly three basic experimental facts pertaining to the early afterglow: 1) a linear dependence  $n^{-1}(t)$  in a wide concentration interval, which is evidence of an apparent two-particle character of the recombination, 2) the large value of the observed recombination coefficients, and 3) the independence of  $\alpha$  of  $P$  in the reduced-pressure interval 3-30 mm Hg.

As indicated in the Introduction, the recombination coefficient is a sum of three terms

$$\alpha = \alpha_e + \alpha_r + \alpha_d \quad (17)$$

The coefficient of shock-radiative recombination in accordance with scheme (4), according to Gurevich and Pitaevskii,<sup>[15]</sup> is equal to

$$\alpha_e = cn, \quad c = \frac{\pi^2 \ln 2}{2^{1/2}} \frac{e^{10}}{m^{1/2} (\chi T_e)^{3/2}} = 0.62 \cdot 10^{-8} T_e^{-3/2} \quad (18)$$

The value of  $\alpha_e$  obtained by Bates et al.<sup>[13, 14]</sup> with the aid of computer calculations gives approximately the same dependence on  $T_e$ , but a somewhat larger numerical factor.

The coefficient of Thomson recombination (5) was found by Pitaevskii,<sup>[20]</sup> and his formula is

$$\alpha_r = \frac{32 (2\pi m)^{1/2} e^6 \sigma_{ea} N}{3 (\chi T_e)^{3/2} M_a} = 1.15 \cdot 10^{-4} P T_e^{-3/2}, \quad (19)$$

where  $M_a$  is the mass of the atom. The numerical factor in (19) is 6.75 times larger than that given by the well-known Thomson formula.<sup>[19]</sup>

The coefficient of dissociative recombination  $\alpha_d$  cannot be calculated at present. Even the character of the temperature dependence of  $\alpha_d(T_e)$  has not been sufficiently reliably established. In a number of theoretical and experimental papers<sup>[39, 40, 3, 9]</sup> it is indicated that  $\alpha_d \propto T_e^{-\delta}$  ( $\delta = 3/2$  or  $1/2$ ). The absolute values of  $\alpha_d$  measured in plasmas of heavy atoms and molecules reach large values on the order of  $10^{-7} \text{ cm}^3 \text{ sec}^{-1}$ .

The complexity of the analysis of the early afterglow is due not only to the many diverse recombination processes, but also to the presence of additional ionization, as well as to the variation of the electron temperature

in the decaying plasma. Therefore, in order to calculate theoretically the kinetics of the electron density it is actually necessary to solve simultaneously a system of nonlinear equations for the concentrations of the electrons and metastable atoms, and for the temperature  $T_e$ . Since this system cannot be solved analytically, we can attempt only to analyze qualitatively the kinetics in different limiting cases.

Most experiments described above are characterized by relatively small metastable-atom and electron concentrations, and we can therefore neglect pair collisions of the metastable atoms. In this case the change of the electron density is described by the free recombination decay, the rate of which depends on the electron temperature. In the same approximation, the quantity  $T_e$ , as indicated above, is determined by the concentration of the metastable atoms and is described by Eq. (16), which after substitution of the numerical coefficients takes the form

$$T_e^{1/2} (T_e - T) = 1.54 \cdot 10^{-7} M/P. \quad (20)$$

It should be borne in mind here that the dependence of  $T_e$  on  $P$  is actually different from (20), and weaker, since the rate of disintegration of the metastable atoms decreases with increasing pressure.

Table III presents the results of calculations of  $c$  and  $M/P$  at different  $T_e$  with the aid of (18) and (20). For clarity, the table lists also the products  $cM/P$ . In the last column, for  $T = 100^\circ\text{K}$ , are given the values of  $cn$  at  $P = 10$  mm Hg, starting from the crude model assumption that  $n = M$ . In the  $T_e$  interval from 200 to  $110^\circ\text{K}$  the increase of  $c$  with decreasing  $T_e$  is offset by the corresponding decrease of  $n$ . At  $10^\circ\text{K}$ , the analogous region of slowing down of  $cn$  is shifted towards lower temperatures in the region  $20-12^\circ\text{K}$ . In general, if the electron temperature decreases sufficiently slowly, then  $cn$  changes little with time and by the same token imitates the two-body character of electron-ion recombination.

As to the quantity  $\alpha_e$ , if we assume  $T_e = 120^\circ\text{K}$  and a density  $n \approx 10^{10} \text{ cm}^{-3}$ , then  $\alpha_e = 2.6 \times 10^{-8} \text{ cm}^3 \text{ sec}^{-1}$ , which is somewhat lower than the observed values of  $\alpha$  in the region of  $T = 100^\circ\text{K}$ . In the case of plasma cooling with liquid helium, it was established that in the diffusion region the values of  $T_e$  were  $50^\circ\text{K}$ . In the recombination region the temperatures should be higher. If we assume as an estimate  $T_e = 64^\circ\text{K}$  and  $n \sim 2 \times 10^9 \text{ cm}^{-3}$ , then we obtain  $cn \sim 10^{-7} \text{ cm}^3$ . In a cryogenic plasma at  $T \approx 10^\circ\text{K}$ , the coefficients  $\alpha_e$  should remain approximately constant for  $\sim 5$  msec, since the metastable-atom concentration and the electron temperature change little during that time.

It can thus be concluded that the mechanism of shock radiative recombination should make an appreciable contribution to the total observed coefficient  $\alpha$ , and, in principle, does not contradict the extended region of linear time dependence of  $n^{-1}$ , which is observed at low temperatures.

The slow variation of  $T_e$  in the afterglow also makes it possible to use dissociative recombination for the explanation of the experimental data. At  $100^\circ\text{K}$ , the existence of dissociative recombination with probable values of the coefficients  $\alpha_d \times 10^8$  on the order of several units

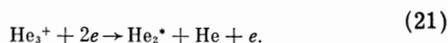
Table III. Estimate of the electron temperature  $T_e$  and of the coefficients of shock-radiative  $\alpha_e = \alpha_n$ 

$T = 100^\circ \text{K}$					$T = 10^\circ \text{K}$				
$T_e$ , deg	$10^{12} c$ , $\text{cm}^6 \text{sec}^{-1}$	$10^{-3} M/P$ , $\frac{\text{cm}^3}{\text{mm Hg}}$	$10^3 cM/P$ , $\frac{\text{cm}^3}{\text{sec mm Hg}}$	$10^3 cn^*$ , $\text{cm}^3 \text{sec}^{-1}$	$T_e$ , deg	$10^{12} c$ , $\text{cm}^6 \text{sec}^{-1}$	$10^{-3} M/P$ , $\frac{\text{cm}^3}{\text{mm Hg}}$	$10^3 cM/P$ , $\frac{\text{cm}^3}{\text{sec mm Hg}}$	$10^3 cn^{**}$ , $\text{cm}^3 \text{sec}^{-1}$
400	0.01	39.	0.47	0.47	256	0.091	25	0.23	0.29
324	0.31	26.	0.82	0.82	196	0.30	17	0.51	0.64
256	0.91	16.	1.5	1.5	169	0.58	13	0.78	0.98
196	3.0	8.8	2.7	2.7	144	1.2	10	1.2	1.5
169	5.8	5.9	3.4	3.4	121	2.6	8.0	2.1	2.6
144	12	3.5	4.2	4.2	100	6.2	5.8	3.6	4.5
121	26	1.5	3.9	3.9	81	16	4.1	6.6	8.2
110	40	0.69	2.8	2.8	64	46	2.8	13	16
105	50	0.34	1.7	1.7	49	150	1.8	27	33
101	59	0.067	0.4	0.4	36	3200	1.0	61	76

\*Assuming  $n = M$  for  $P = 10 \text{ mm Hg}$ .\*\*Assuming  $n = M/4$  for  $P = 5 \text{ mm Hg}$ .

is necessary for the explanation of the absolute value of the measured coefficient. Although the peculiarities of the recombination and the value of its coefficient at  $T = 10^\circ \text{K}$  can be described on the basis of shock-radiative recombination, the model of the afterglow will be reconciled in the entire region of low temperatures only in the presence of dissociative recombination in this case.

The probability of dissociative recombination with large  $\alpha_d$  in a cryogenic plasma is high because the triatomic ions  $\text{He}_3^+$  predominate. Since the molecular glow of only the excited states of  $\text{He}_2^*$  is observed in the optical emission of the plasma,<sup>[25]</sup> the recombination of  $\text{He}_3^+$  is accompanied by dissociation. Moreover, shock-radiative recombination with participation of the  $\text{He}_3^+$  ion and two electrons should also be accompanied by dissociation into an excited  $\text{He}_2^*$  molecule and an atom in a normal state, in accordance with the scheme



Such a reaction can be called shock-dissociative recombination.

Let us discuss now the behavior of the coefficients  $\alpha$  as functions of the pressure. The data of Tables I and II show that when  $P$  changes by almost one order of magnitude the values of  $\alpha$  remain constant within a spread not exceeding  $\pm 25\%$ . The deviations from the mean are connected more readily with different excitation conditions and with the initial concentration  $n_0$  than with the influence of the pressure. In their nature, the coefficients  $\alpha_e$  and  $\alpha_d$  do not depend explicitly on  $P$ , and this dependence can become manifest only to the extent that the pressure has a weak influence on  $T_e$ . Both recombination mechanisms are in full agreement with the experimental fact that  $\alpha$  is independent of  $P$ . To the contrary, triple Thomson recombination leads to a linear dependence on the pressure. According to (19),  $\alpha_T = 1.9 \times 10^{-8}$  at  $T_e = 125^\circ \text{K}$  and  $P = 30 \text{ mm Hg}$ , and  $\alpha_T = 6.2 \times 10^{-8}$  at  $T_e = 60^\circ \text{K}$  and  $P = 15 \text{ mm Hg}$ . These values of  $\alpha_T$  correspond to the characteristic  $T_e$  and to the maximal  $P$  in our experiments. Their values amount to  $\sim 35\%$  of the measured value of  $\alpha$  at  $100^\circ \text{K}$  and  $\sim 50\%$

at  $10^\circ \text{K}$ , i.e., are 1.5–2 times larger than the maximum experimental scatter. The contribution made to  $\alpha$  by such coefficients  $\alpha_T$  could be observed. The independence of  $\alpha$  of  $P$  would be reconciled with the theoretical estimates if the numerical coefficient in (19) were to be smaller by a factor of at least 1.5–2.

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