

Ion–ion recombination in SF₆ and in SF₆–C₂H₆ mixtures for high values of E/N

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Abstract. Ion–ion recombination coefficients in a decaying SF₆ plasma and in SF₆–C₂H₆ mixtures are measured in the pressure range 15–90 Torr for reduced field strengths 100–250 Td. The charge composition is analysed and dominating ion–ion recombination channels in such plasmas are determined. Relations for estimating the potential drop at the electrodes are obtained for decaying plasma in strongly electronegative gases. The results of measurements are extrapolated for estimating the ion–ion recombination coefficient in SF₆ for nearly critical field strengths. It is concluded that the ion–ion recombination should be taken into account in calculations of the discharge characteristics in non-chain reaction HF lasers.

Keywords: SF₆, ion–ion recombination, HF laser, electronegative gases.

1. Introduction

The possibility of exciting a self-sustained volume discharge (SVD) without preionisation, i.e., self-initiated volume discharge [1] in SF₆ mixtures with hydrocarbons, was discovered in Ref. [2], and raised the problem of scaling non-chain reaction HF lasers to a qualitatively new level, increasing their power and energy by more than an order of magnitude (by a factor of 40). At present, the energy emitted by SVD-initiated non-chain reaction HF lasers exceeds 400 J for an electrical efficiency of more than 4% [3].

In this connection, it is interesting to continue the investigation of SVD in SF₆ and in mixtures of SF₆ with hydrocarbons. Because of its strongly electronegative nature, the SF₆ plasma of such a discharge possesses a number of specific features. For example, the concentration of positive and negative ions in such a plasma is much higher than the electron concentration (by almost two orders of magnitude) [1, 4]. For this reason, processes associated with the ionic component of plasma (destruction of negative ions by

electron impact, dissociative electron–ion recombination and ion–ion recombination) may play a significant role in the discharge kinetics [4]. The latter process affects considerably the ion concentration in SVD and completely determines the charge kinetics in a decaying plasma.

The ion–ion recombination in SF₆ and its mixtures with hydrocarbons in an external electric field has not been adequately studied. Apparently, only two works [5, 6] reported the measurement of the ion–ion recombination coefficient β in SF₆, binary mixtures of SF₆ with the hydrocarbon CH₄ used rarely in HF lasers, and in ternary SF₆–CH₄–Ar/He mixtures at working pressures $p > 100$ Torr and relatively low reduced electric field strengths $E/N < 160$ Td (N is the concentration of neutral particles). The remaining experimental [7] and theoretical (simulation by the Monte Carlo method) [8] studies cover a much wider range of pressures ($\sim 10^2 - 10^4$ Torr), however, in the zero electric field approximation.

To calculate the SVD characteristics in a non-chain reaction HF laser, of main interest is the coefficient β for mixtures of SF₆ with the hydrocarbon C₂H₆ [1–3] for $p = 30 - 90$ Torr (lasers with quite large apertures) and values of E/N close to the critical value $(E/N)_{cr}$ in SF₆. The aim of this paper is to measure the ion–ion recombination coefficient in pure SF₆ and its mixtures with C₂H₆ in the above-mentioned pressure range for values of E/N up to 250 Td. In particular, this makes it possible to obtain a reasonable value of the coefficient β for $E/N \sim (E/N)_{cr}$ characteristic of the self-sustained discharge.

2. Experimental

Ion plasma for measuring the coefficient β was induced by a pulsed SVD. The scheme of the experimental setup is shown in Fig. 1. The SVD was ignited between an anisotropically resistive cathode K of size 5 cm × 5 cm (similar to the one used in Ref. [9]) and a disc-shaped anode A of diameter 12 cm, rounded off at the edge to a radius 1 cm, by commutating the voltage with a gap P1. The interelectrode distance was varied from 2 to 8 cm. The anisotropically resistive cathode made it possible to obtain a discharge distributed uniformly over the cathode surface, which is necessary for the applicability of comparatively simpler relations that we will use below for determining ion concentration.

The resistance $R_c \sim 1 \Omega$ of the anisotropically resistive cathode is much smaller than the resistance $R_p \sim 1 \text{ k}\Omega$ of the ion plasma, and hence does not introduce any noticeable distortions in the measured values of ion current. The capacitance 2–8 nF of capacitor C₁ was determined from the

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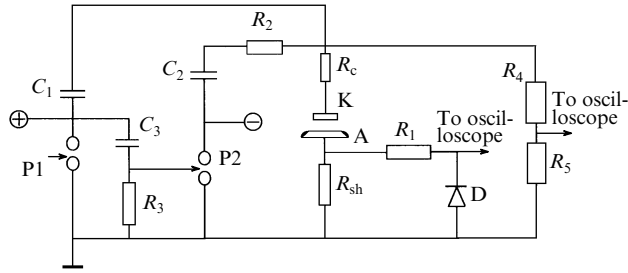


Figure 1. Scheme of the setup for measuring the ion–ion recombination coefficient.

conditions of the SVD stability and homogeneity. The capacitor $C_2 = 174$ nF connected to the gap P2 was used to maintain a constant voltage across the discharge gap during the measurement of the ion current (up to 20 μ s). The resistance $R_2 = 50$ Ω , connected in the discharge circuit of this capacitor, minimises its influence on the energy released in the SVD plasma. The gap P2 was triggered automatically upon activation of the gap P1 and closure of the capacitor C_3 circuit through the resistor R_3 . The current was detected by the shunt resistance R_{sh} .

Because the ion current of the decaying plasma was much weaker (by several orders of magnitude) than the maximum SVD current, to improve the accuracy of measurements, the signal from the shunt was cut off at the level of 1 V by the $R_1 - D$ diode limiter. Calibration of the ion current measuring circuit and verification of its linearity were performed by shunting a capacitor (through the gap) charged to a voltage ~ 4 kV through a resistance ~ 1 k Ω . Fig. 2a shows typical oscillograms of the current and voltage across the discharge gap recorded in this way. The negative spikes on the oscillograms correspond to the SVD current and voltage.

The ion–ion recombination coefficient β was calculated from the current oscillogram $I(t)$ for a fixed voltage U across the discharge gap with the help of the relations

$$n_i(t) = \frac{I(t)}{Se(b_i^+ + b_i^-)E}, \quad (1)$$

$$n_i(t) = \frac{n_i(0)}{1 + n_i(0)\beta t}. \quad (2)$$

Here, $n_i(t)$ is the ion concentration; S is the cathode area; e is the electron charge; $E = U/d$; $n_i(0)$ is the initial ion concentration. Relations (1) and (2) are written under the assumption that there are only one type of positive and one type of negative ions with mobilities b_i^+ and b_i^- , respectively. The possibility of such an assumption is discussed below. The quantities $n_i(0)$ and β were determined from relation (2) from the values of $n_i(t)$ reconstructed from (1) by the method of least squares (Fig. 2b).

3. Results of measurements

Fig. 3 shows the dependences of the ion–ion recombination coefficient β on the parameter E/N in pure SF₆ under a pressure $p = 15 - 90$ Torr. One can see that β strongly decreases with increasing E/N over the entire pressure range. This result is consistent with the data presented in Ref. [5], where measurements were performed by the

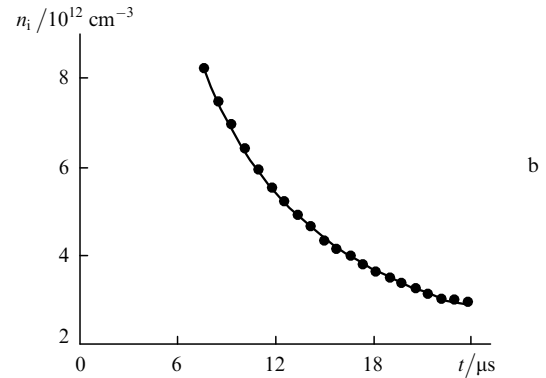
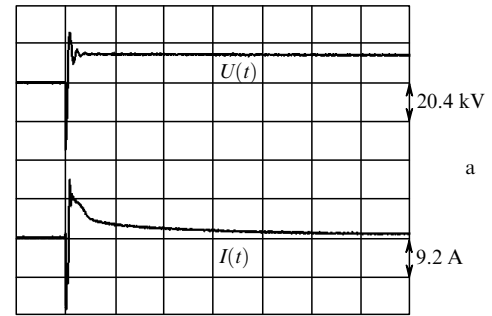


Figure 2. (a) Oscillograms of voltage U across the discharge gap (upper trace) and the current I in a decaying plasma (lower trace), and (b) dependence of the ion concentration n_i on time t , calculated from the oscillograms of Fig. 2a (dark circles) and by the method of least squares (solid curve) for $p = 30$ Torr and $E/N = 230$ Td.

standard technique using an electron beam for producing the ion plasma. In the region of small E/N , the coefficient β increases with pressure p almost linearly. In the range $E/N > 200$ Td, this law is violated and the dependence of β on p becomes weaker. This fact does not contradict the results obtained in Ref. [10], where it was shown by calculations that the ion–ion recombination coefficient depends not only on the parameter E/N , but also on the absolute value of the electric field strength E , decreasing with increasing E .

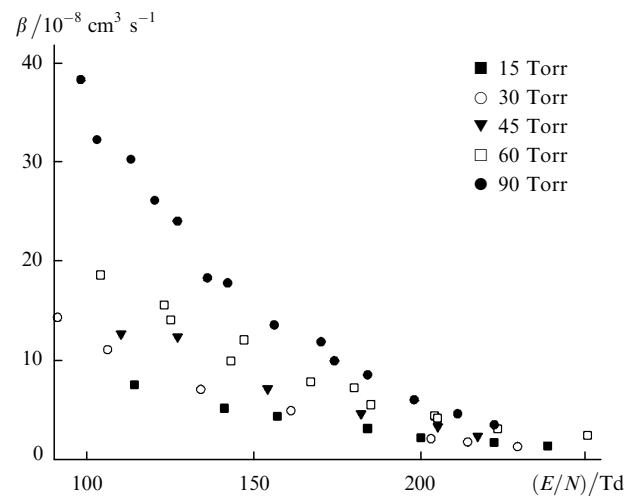


Figure 3. Dependences of the ion–ion recombination coefficient β on the parameter E/N for SF₆ under different pressures.

Note also that in the region of low values of $E/N = 100 - 160$ Td, the values of the coefficient β measured in our experiments for $p = 90$ Torr are close to the corresponding values from Ref. [5]. In the SF₆:C₂H₆ = 10:1 mixture, the values of β obtained for $E/N = 250$ Td and $p = 60$ and 90 Torr are 4.3×10^{-8} and 6.5×10^{-8} cm³ s⁻¹, respectively. No changes β were observed upon variation of the SVD parameters, such as the specific deposited energy and the discharge-current duration over a rather wide range.

4. Discussion of results

Consider the ion composition and the nature of ion-ion recombination in SF₆ and its mixtures with C₂H₆. We are not aware of any experimental facts confirming the presence of positive SF₆⁺ ions in the plasma of self-sustained discharge in SF₆. According to the results of direct mass-spectrometric measurements (see, for example, Refs [11, 12]), the SF₅⁺ ion dominates in the plasma. This is consistent with the results presented in paper [13], in which it was shown that the SF₆⁺ ion formed in the plasma is in predissociative state and dissociates into the SF₅⁺ ion during a time period much smaller than any characteristic discharge times. As for the negative ions, several kinds of such ions should be considered in general.

In the range of values of E/N close to $(E/N)_{cr}$ and below, in which we are interested, the dissociative attachment of electrons to SF₆ molecules results in the formation of predominantly negative SF₆⁻ ions [14]. The rates of generation of the SF₅⁻ and F⁻ ions are about half the rate of formation of SF₆⁻ ions [14]. The mobilities of the SF₆⁻ and SF₅⁻ ions are virtually identical and are close to those obtained in the Langevin (polarisation) approximation [13]. The mobility of the F⁻ ions can also be estimated quite accurately in the same approximation.

The dissociative charge exchange of the SF₆⁻ and SF₅⁻ ions with SF₆ molecules, resulting in the formation of negative ions of another kind (mainly F⁻ ions) does not have time to occur even under the maximum pressures ~ 90 Torr used in our experiments. For example, according to the data presented in Ref. [15], the minimum time τ_{min} for the reaction SF₆⁻ + SF₆ → SF₆ + SF₅ + F⁻ at $p = 90$ Torr and for $E/N \sim (E/N)_{cr}$ corresponding to the self-sustained discharge stage exceeds 1 μ s, while the duration of the SVD itself in these experiments is $\sim 100 - 300$ ns.

In a decaying plasma with much smaller values of the ratio E/N , the time τ_{min} increases by several orders of magnitude. Thus, the SF₆⁻ and SF₅⁻ ions vanish in the decaying SF₆ plasma only as a result of recombination with the SF₅⁺ ions. The detachment of an electron from the F⁻ ion can occur only in the layers in the vicinity of the electrode (see below). As a result, the decrease in the concentration of F⁻ ions in the plasma is also determined only by their recombination with SF₅⁺ ions.

In the pressure range under study, a three-body mechanism of ion-ion recombination is realised. According to the preliminary estimates obtained by using the well-known three-body recombination models [16] and assuming the polarisation nature of interaction of ions with gas molecules, it can be expected that the coefficients of recombination of SF₆⁻ and SF₅⁻ ions with SF₅⁺ ions are close, while the coefficients of recombination of F⁻ and SF₅⁺ ions are much higher. In this connection, we consider a decaying ion plasma with one type of positive ions with the concentration

$n^+(t)$ and two types of negative ions with concentrations $n_1^-(t)$ и $n_2^-(t)$.

Suppose that the ion-ion recombination coefficients are equal to β_1 and β_2 , respectively. For the sake of definiteness, we assume that $\beta_2 > \beta_1$. Taking into account the quasi-neutrality of the plasma, we obtain the following integro-differential equation for the density of positive ions:

$$\frac{d\varphi}{d\tau} = q_1 \exp(-\varphi) + q_2 \exp(-\sigma\varphi),$$

$$\varphi = \int_0^\tau y d\tau, \quad y = \frac{n^+(t)}{n^+(0)}, \quad (3)$$

$$\tau = \beta_1 n_1^-(0)t, \quad \sigma = \frac{\beta_2}{\beta_1}, \quad q_1 = \frac{n_1^-(0)}{n^+(0)}, \quad q_2 = \frac{n_2^-(0)}{n^+(0)}.$$

Here, $n^+(0)$, $n_1^-(0)$ and $n_2^-(0)$ are the initial concentrations of positive and negative ions.

Because $\sigma > 1$ and $\varphi(\tau) \rightarrow \infty$ for $\tau \rightarrow \infty$, the second term in Eqn (3) can be neglected starting from a certain instant of time. Taking into account that $n_1^-(t) = n_1^-(0) \times \exp(-\varphi)$, this equation is transformed into the recombination equation only for negative ions with the smaller recombination coefficient β_1 . As applied to the SF₆ plasma, this means that within a certain time interval after the recombination onset, negative ions SF₆⁻ and SF₅⁻, which have close values of the mobility and recombination coefficient (see above), start dominating in the plasma along with the positive SF₅⁺ ions. This justifies the use of Eqns (1) and (2). This is clearly shown in Fig. 2b. One can see that for $t > 8$ μ s, the time decay $n_i(t)$ of the ion concentration calculated from (1) strictly follows the recombination relation (2). The values of ion mobilities used in calculations were borrowed from Ref. [13].

The complex SF₆⁻(SF₆) ion was considered as the main negative ion in Refs [5, 6, 8], but the gas pressures were 5–10 times higher in these works than in our experiments. Taking into account that the rate of clusterisation of the SF₆⁻ ions quadratically depends on p , we can expect that the fraction of the complex SF₆⁻(SF₆) ions in the investigated plasma is insignificant. Moreover, according to Ref. [13], the mobilities of ions SF₆⁻ and SF₆⁻(SF₆) differ by just a few percent in the range $E/N \sim 100 - 250$ Td, so that the formation of complex ions under the conditions of our experiments is insignificant at all.

In addition to the SF₅⁺ ions, the SF₆:C₂H₆ = 10:1 mixture also contains positive ions formed upon the electron impact ionisation of C₂H₆. According to Refs [17, 18], the mechanism of dissociative ionisation with the formation of C₂H₄⁺ ions and H₂ molecules dominates in this case. Using the Langevin approximation and the Blank law [19], we find that the mobility of SF₅⁺ and C₂H₄⁺ ions in the SF₆:C₂H₆ = 10:1 mixture is determined by their interaction with the SF₆ molecules. Despite the fact that the mobility of C₂H₄⁺ ions (estimated in the polarisation limit) is about 1.8 times higher than the mobility of SF₅⁺ ions, the C₂H₄⁺ ions do not make a significant contribution to the total current $I(t)$ for the above-mentioned ratio of the concentrations of SF₆ and C₂H₆. As a result, the set of negative ions remains unchanged. Taking into account that positive SF₅⁺ ions dominate in the plasma under study, expression (2) describing the recombination kinetics remains applicable for the mixture as well.

While the formation of positive ions in the $\text{SF}_6 - \text{C}_2\text{H}_6$ mixture is also possible upon the charge exchange of the SF_5^+ ions with the C_2H_6 molecules (although, in our opinion, the situation is not quite clear), estimates show that the charge exchange times are $\sim 10^{-8} - 10^{-7}$ s even for the lowest C_2H_6 concentrations of $\sim 10^{17}$ cm^{-3} . Therefore, only one positive ion will dominate on the time scale exceeding 1 μs of interest to us.

The polarisation approximation was used several times while estimating the ion mobility, although this cannot be always substantiated rigorously under the conditions of our analysis. However, it is known that, as a rule, the values of mobility obtained by using the Langevin formula do not differ significantly from the measured values. This is clearly demonstrated in Ref. [13] for the SF_6 molecules. Thus, the above arguments concerning the ion composition of the working medium and the nature of ion recombination under the investigated conditions are justified, at least in principle. The above-mentioned constancy of the ion-ion recombination coefficient upon a considerable variation of the discharge parameters is another indirect evidence in favour of this conclusion, since it follows from what has been stated above that the set of negative ions in the recombination plasma depends weakly on the initial discharge conditions.

The field strength E_p in the plasma was determined in our experiments by dividing the voltage across the discharge gap by the interelectrode distance. It is well known, however, that the voltage drop at the electrode may be very high in the case of strongly electronegative gases. Therefore, it should be interesting to estimate the error introduced in the value of E_p by this method.

To estimate the voltage drop U_c across the cathode, we will use the one-dimensional approximation and assume as in Ref. [20], that the field $E_c(x)$ in the cathode layer is independent of the longitudinal coordinate x , i.e., $E_c(x) = E_c$. The estimates made below show that under the conditions under study, this quantity greatly exceeds not only E_p , but also the critical field strength E_{cr} , so that the formation of negative ions in the layer can be neglected. In this case, the distributions of the current density $j_e(x)$ of electrons and $j_+(x)$ of positive ions in the cathode region are described by the same equations as in the case of an electropositive gas. Beyond the cathode layer ($x > d_c$), the electron component of the total current density J_t in a decaying plasma rapidly vanishes because of an intense attachment of electrons. As a result, the boundary conditions are somewhat different from those considered normally for the electropositive gas:

$$j_e(0) = \gamma j_+(0), \quad j_e(d_c) + j_+(d_c) = J_t, \quad (4)$$

where γ is the secondary electron emission coefficient. The second condition in (4) can also be formulated as $j_e(d_c) = j_-(d_c)$, where $j_-(x)$ is the current density of negative ions. Using the standard technique (see, for example, Ref. [20]), the continuity equations for $j_e(x)$ and $j_+(x)$ taking into account Eqns (4) and the fact that $\gamma \ll 1$, we can easily obtain the following expression in the reduced variables accepted in the theory of near-electrode layers [21]:

$$\frac{\alpha(E_c/p)}{p} p d_c = B_1, \quad B_1 = \ln \left(\frac{1 + \gamma}{\gamma} \frac{b_1^-}{b_1^+ + b_1^-} \right), \quad (5)$$

where α is the Townsend coefficient.

Proceeding from the Poisson equation, we obtain a relation between parameters E_c and d_c [20]:

$$\left(\frac{E_c}{p} \right)^2 = \frac{(J_t/p^2) p d_c}{\varepsilon_0 b_1^+ p}, \quad (6)$$

where ε_0 is the dielectric constant of vacuum. Then, we obtain from Eqns (5) and (6) the following relations for determining U_c :

$$\frac{(E_c/p)^2 [\alpha(E_c/p)/p] \varepsilon_0 b_1^+ p}{J_t/p^2} = B_1, \quad U_c = \frac{E_c}{p} p d_c. \quad (7)$$

To estimate the potential drop in the anode region, we should consider impact ionisation as well as the formation and neutralisation of negative ions. The latter process leads to the emergence of seed electrons in the anode region because the electron current coming to this region from the decaying plasma is vanishingly small. In analogy with the above conclusion, we assume that the field E_a in the anode layer is constant.

In the approximation of a planar layer, the system of corresponding continuity equations can be written in the form

$$\begin{aligned} \frac{dj_e(x)}{dx} &= -\alpha j_e(x) - \delta j_-(x), & \frac{dj_-(x)}{dx} &= -\eta j_e(x) + \delta j_-(x), \\ \frac{dj_+(x)}{dx} &= \alpha j_e(x). \end{aligned} \quad (8)$$

Here, η is the electron attachment coefficient; δ is the coefficient of detachment of electrons from negative ions in collisions with gas molecules. The coordinate x is measured from the anode ($x = 0$) into the depth of the discharge gap.

The decrease in the electron current caused by the attachment to the SF_6 molecules can be neglected for the same reason as in the case of the cathode layer ($E_a \gg E_{cr}$). Taking this into account, the boundary conditions at the anode and at the boundary between the anode layer and the plasma ($x = d_a$) have the form

$$j_+(0) = 0, \quad j_e(d_a) = 0, \quad j_-(d_a) + j_+(d_a) = J_t. \quad (9)$$

As a result, we arrive at the relation

$$(\lambda_1 + \lambda_2) d_a = B_2, \quad (10)$$

$$B_2 = \ln \left\{ \frac{b_1^+ \lambda_2}{b_1^- \lambda_1} + [1 + (\lambda_1 + \lambda_2) d_a] \frac{b_1^+ + b_1^-}{b_1^-} \right\},$$

where $\lambda_{1,2} = \{\pm(\delta - \alpha) + [(\alpha - \delta)^2 + 4\alpha\delta]^{1/2}\}/2$. Since $E_a \gg E_{cr}$, we have $\alpha \gg \delta$, $\lambda_1 \simeq \delta$, $\lambda_2 \simeq \alpha$ and expression (10) is considerably simplified:

$$\frac{\alpha(E_a/p)}{p} p d_a \approx \ln \left[\frac{\alpha(E_a/p)}{\delta(E_a/p)} \right]. \quad (11)$$

Using a relation between E_a and d_a analogous to (6), and neglecting the difference in the mobilities of positive and negative ions ($b_1^+ \approx b_1^- \approx b_1$), we obtain in the approximation $\alpha \gg \delta$ the following expression for E_a :

$$\left(\frac{E_a}{p} \right)^2 \frac{[\alpha(E_a/p)/p] \varepsilon_0 b_1 p}{J_t/p^2} \approx \ln \left[\frac{\alpha(E_a/p)}{\delta(E_a/p)} \right], \quad (12)$$

which is used, together with (11), for finding d_a and, hence, the potential drop U_a across the anode.

Using the results obtained, we can now easily estimate the relative error $\xi = (E - E_p)/E$ in determining the field E_p in a decaying plasma:

$$\xi = \frac{B_1/\eta_c + B_2/\eta_a}{U}, \quad \eta_{c,a}(E_{c,a}/N) = \frac{k_i(E_{c,a}/N)}{u_e(E_{c,a}/N)(E_{c,a}/N)}, \quad (13)$$

where $k_i(E_{c,a}/N)$ and $u_e(E_{c,a}/N)$ are the impact ionisation constant and the drift velocity of electrons, respectively.

As an example, consider the SF₆ plasma under a pressure $p = 30$ Torr with a characteristic current density $J_t \sim 1$ A cm⁻². Using the data of Refs [14, 15, 22] for k_i and u_e and expressions (7), (11), (12) and (13), we obtain $\xi \approx 0.1$. The same relative error is typical for all other plasma decay regimes considered in this work.

We have made a number of assumptions while estimating the quantities U_c and U_a (for example, the possibility of formation of double layers in the electrode region was also not considered). Nevertheless, the estimates obtained approximate well the real values. An indirect confirmation of this is provided, for example, by the fact that the estimates of the quantities $\Delta U = U_c + U_a$ are in satisfactory agreement with the corresponding values obtained by us by extrapolating the experimental dependence $U_{st}(pd)$ [1] for SF₆ and mixtures of SF₆ with C₂H₆ in the region $pd \rightarrow 0$ (U_{st} is the voltage in the quasi-stationary phase of the SVD).

The order of magnitude of the electrode layer relaxation time coincides with the drift time $\tau_{c,a} = \varepsilon_0(E_{c,a}/p)/[(J_t/p^2)p]$ of ions through this layer. For example, we obtain $\tau_{c,a} \sim 10^{-8}$ s for $p = 30$ Torr and $J_t \sim 1$ A cm⁻². Values of $\tau_{c,a}$ of the same order of magnitude are also obtained under other conditions. Because we are interested only in the microsecond region in this work, the quasi-stationary approximation used above is fully justified.

For the characteristic values of E_c and E_a in the investigated gases under the conditions described above, the relation $l_c \sim l$ is satisfied in the electrode regions, where l and l_c are the mean free path and the energy relaxation length of electrons, respectively. It also follows from relations (5), (6), (11) and (12) that $d_{c,a} \gg l_i$, where l_i is the ionisation length in the cathode or anode region. Taking into account that $l/l_i < 1$, we obtain $l_e/d_{c,a} \ll 1$. Therefore, local dependences of transport coefficients on the field strength can be used in the above description.

5. Conclusions

We have measured the ion-ion recombination coefficients in SF₆ and in SF₆:C₂H₆ = 10:1 mixtures in the pressure range 15–90 Torr for reduced electric field strengths $E/N = 100 - 250$ Td. The error in the estimates of fields in the plasma does not exceed 10% for an overall measuring error below 20%. The extrapolation of the results of measurements to the region of higher values of E/N provides an estimate 10^{-8} cm³ s⁻¹ for the coefficient of recombination β of the SF₆⁻ and SF₅⁻ ions with the SF₅⁺ in SF₆ for $E/N \sim (E/N)_{cr}$. The values of β for SF₆ at $p = 90$ Torr and for $E/N < 160$ Td are close to those obtained in Ref. [5] by a different technique. For pressures $p = 60$ Torr typical of HF lasers, the recombination coefficient in a decaying plasma is $\beta = 4.3 \times 10^{-8}$ cm³ s⁻¹ in the SF₆:C₂H₆ = 10:1 mixtures. Calculations [4] show that

a decrease in the ion concentration by an order of magnitude occurs over a time ~ 200 ns comparable with the duration ~ 300 ns of the entire discharge. This leads to the assumption that ion-ion recombination in SF₆-C₂H₆ mixtures may considerably limit the ion concentration at the stage of self-sustained discharge, and should be taken into account in the calculations of the HF-laser characteristics. The simulation of the SVD in SF₆ using the value $\beta = 10^{-8}$ cm³ s⁻¹ for the recombination coefficient shows that in this case, the ion-ion recombination also may considerably affect the ion density balance at all stages of the discharge.

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