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Detachment instability of self-sustained volume discharge in active media of non-chain HF(DF) lasers

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***Abstract*. The development of detachment instability in active media of electric-discharge non-chain HF(DF) lasers due to the electron-impact detachment of electrons from negative ions is considered. This instability is initiated in large volumes of SF_6-based gas mixtures, spatially separated from electrodes and heated by a pulsed CO_2 laser. The self-organisation of self-sustained volume discharge upon laser heating, which results in the formation of quasi-periodic plasma structures within the discharge gap, is experimentally investigated. The evolution of these structures, depending on the gas temperature and specific deposition of electric energy, is analysed. The possible relationship between the self-organisation and detachment instability is discussed. A mechanism of development of single plasma channels in the working media of HF(DF) lasers, based on electron-impact destruction of negative ions is proposed.**

Keywords: *non-chain HF laser, self-sustained volume discharge, SF_6, gas discharge instability, laser-induced heating.*

1. Introduction

The ionisation instability of self-sustained volume discharge (SVD) in SF_6 and its mixtures is of great interest in view of the development of non-chain chemical HF(DF) lasers [1].

Currently, several mechanisms of ionisation instability in electronegative gases are known. A general theoretical approach to their analysis was formulated in [2]. The development of instabilities in working media of CO_2 lasers as a result of detachment of electrons from negative ions by neutral and electronically excited gas components was considered in [3 – 5]. The mechanism of instability caused by electron-impact dissociation of a small amount of electronegative impurity ('burning away' of halogen additive) was analysed in detail for excimer lasers [6 – 9]. The instability in SF_6, according to [10], should be related to the step ionisation of SF_6 molecules.

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The ionisation instability caused by electron-impact detachment of electrons from negative ions in strongly electronegative polyatomic gases at intermediate pressures and for times of few tens of nanoseconds may develop according to a qualitatively different scenario. This issue was considered by us for the first time in [11] for SVD in SF_6 and its mixtures. In this context, the following circumstance is of fundamental importance.

At intermediate pressures of SF_6-based gas mixtures at room temperatures the best agreement between the calculated time dependences and experimental SVD current and voltage oscillograms, including those recorded in the quasi-stationary SVD phase (at $E/N \approx (E/N)_{cr}$), is observed (see below) when setting $\beta_{ei} \approx k_d$. Here, E is the electric field strength; N is the gas concentration; β_{ei} and k_d are the rate constants of electron – ion recombination and electron-impact detachment of electrons from negative ions, respectively; and $(E/N)_{cr}$ is the critical reduced electric field strength. In this case, the increase in the electron concentration due to the electron-impact destruction of negative ions is almost completely compensated for by their loss as a result of dissociative electron – ion recombination. The above-mentioned nonlinear mechanism of electron multiplication manifests itself only at a significant unbalance between the constants k_d and β_{ei}, which may occur either upon strong gas heating or at $E/N \gg (E/N)_{cr}$. Indeed, the constant $\beta_{ei} \sim T_g^{-1} T_e^{-\chi}$ ($\chi > 0$, T_g is the gas temperature, $T_e = 2\langle\varepsilon\rangle/3$, $\langle\varepsilon\rangle$ is the mean electron energy [12]) decreases under these conditions, whereas the constant k_d can only increase.

The conditions under which $E/N \gg (E/N)_{cr}$ are implemented, for example, near the tip of incomplete channel, developing from the cathode side. However, in this case ionisation processes develop at distances of the same order of magnitude as the channel radius; hence, they are fairly difficult to study experimentally. At the same time, one can implement a significant unbalance between the constants k_d and β_{ei} and initiate the mechanism of detachment instability under consideration even in large gas volumes. An efficient way to do this is to heat SF_6-based gas mixtures by a pulsed CO_2 laser [13]. Here, we used this approach to study the SVD instability caused by electron-impact detachment of electrons from negative ions in SF_6 and SF_6-based gas mixtures, including working media of HF(DF) lasers.

2. Experimental setup

The experimental setup (see schematic in Fig.1) and measuring technique did not differ much from those used

by us previously [11]. We investigated SVD in SF_6 and its mixtures with C_2H_6, H_2, and Ne at a partial SF_6 pressure of 15 Torr and a total mixture pressure up to 50 Torr. A discharge was ignited in the needle (cathode)–cylinder (anode) geometry at an interelectrode distance of 43 mm. The needle was imitated by a segment of polyethylene-insulated copper wire. The cathode was placed in a segment of a dielectric tube with an inner diameter of 15 mm, protruding above the needle tip by 12 mm. The specific electric energy deposition W_{el} in the plasma discharge was varied from 0.02 to 0.2 J cm^{-3}.

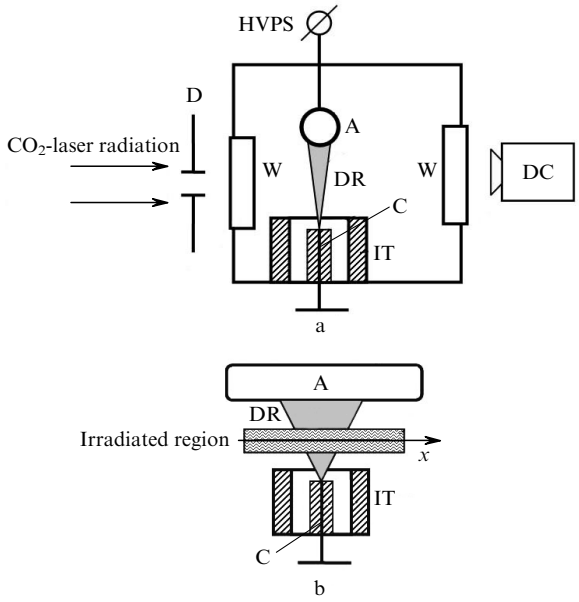

Figure 1. Experimental setup, viewed (a) form aside and (b) from the side of the camera: (A) anode, (C) cathode, (D) slit diaphragm, (HVPS) high-voltage power supply, (W) BaF_2 windows, (IT) insulating tube, (DC) digital camera, and (DR) discharge region.

The gas was preliminarily heated only in a narrow region of the discharge gap due to the gap irradiation by a pulsed CO_2 laser through a 10-mm-wide slit diaphragm, oriented perpendicularly to the applied electric field (Fig. 1). As will be shown below, this irradiation scheme makes it possible to observe SVD constriction directly in the heated-gas volume (similarly to the glow discharge constriction [12, 14]) rather than in the form of a channel, stemming from the cathode spot and finally closing the discharge gap [15]. The gas temperature T_g, which is set in the SVD burning region, was determined from the amount of laser energy absorbed by SF_6 molecules [13] and from the propagation velocity of the shock wave formed at the boundaries between the cold and heated gas regions [16]. The temperature T_g was varied in the range of 800–2100 K (the specific energy W_a of the laser radiation absorbed by SF_6 in the SVD region was 0.05–0.23 J cm^{-3}). A voltage pulse was applied to the discharge gap with a delay of 4 μs with respect to the μs laser pulse (the delay was counted with respect to the leading edges of the pulses [17]) at a level of 0.1, which ensured establishment of a thermal equilibrium between the translational and internal degrees of freedom of the irradiated-gas components by the instant of discharge ignition in the pressure range under study [18].

During the experiments we monitored the voltage across the discharge gap and the SVD current using a resistive voltage divider and low-resistivity shunt, respectively, and recorded SVD by a digital camera, synchronised with the laser pulse. To identify the main processes determining the SVD current–voltage ($I-U$) characteristics, we also recorded voltage and current oscillograms for a confined discharge, which is known to exclude the influence of the factors caused by the increase in the discharge volume with an increase in the amount of energy deposited into the plasma on the discharge current and voltage [19]. To this end, SVD was ignited in a quartz tube 8.5 mm in diameter at an interelectrode distance of 43 mm (in the needle–plane geometry). The experimental oscillograms were compared with the calculation results (the calculation technique was described in detail in [20]).

3. Experimental results

Figures 2a and 2b show photographs of SVD in an SF_6–Ne–C_2H_6 mixture, obtained at $W_{el} = 0.2$ J cm^{-3}, discharge current pulse duration $\tau_{dis} = 150$ ns, and different T_g values. The distributions of discharge glow intensity along the x axis (which is parallel to the boundaries of the heated region and passes through its middle; see Fig. 1), corresponding to the aforementioned photographs, are shown in Figs 2c and 2d. It can be seen that SVD in the heated region becomes filamentary, showing a quasi-periodic plasma (current) structure. Its spatial period decreases with an increase in T_g. It is noteworthy that the large plasma (current) channels in Fig. 2b consist of thinner filaments. Despite the SVD filamentary structure, the plasma channels in the heated region are diffuse at not very large W_{el} and τ_{dis} values. An increase in W_{el} or τ_{dis} leads to dominance of one of the channels (lying generally

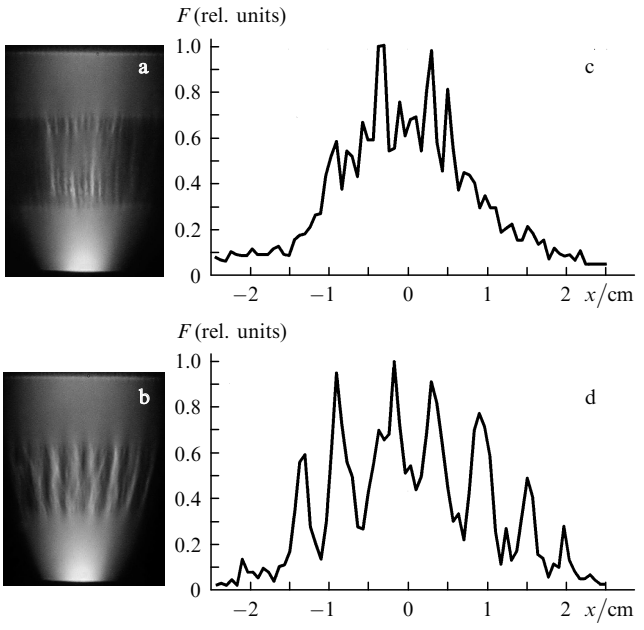

Figure 2. (a, b) SVD photographs at $T_g =$ (a) 1550 and (b) 800 K and (c, d) distributions of SVD radiation intensity F along the coordinate x in the heated region at $T_g =$ (c) 1550 and (d) 800 K; SF_6:Ne:C_2H_6 = 5:5:1 mixture, $p = 33$ Torr; $W_{el} = 0.2$ J cm^{-3}, and $\tau_{dis} = 150$ ns.

near the central electric field line) and its constriction. It is noteworthy that both in the case of cold gas [19] and upon heating the SVD constriction threshold with respect to W_{el} and τ_{dis} in pure SF_6 is lower than in $SF_6 - C_2H_6$ mixtures. This is illustrated in Fig. 3 by a photograph of SVD in pure SF_6, recorded at $\tau_{dis} = 160$ ns and $W_{el} = 0.12$ J cm^{-3}. One can see that an instability develops directly in the irradiated region of the discharge gap. Apparently, a pertinent analogy for this situation is the constriction of low-pressure glow discharge [12, 14]. With a further increase in W_{el} or τ_{dis} the spark channel bridges the entire gap. For constant W_{el} and τ_{dis} values the probability of SVD constriction also increases with an increase in T_g in the irradiated part of the gap.

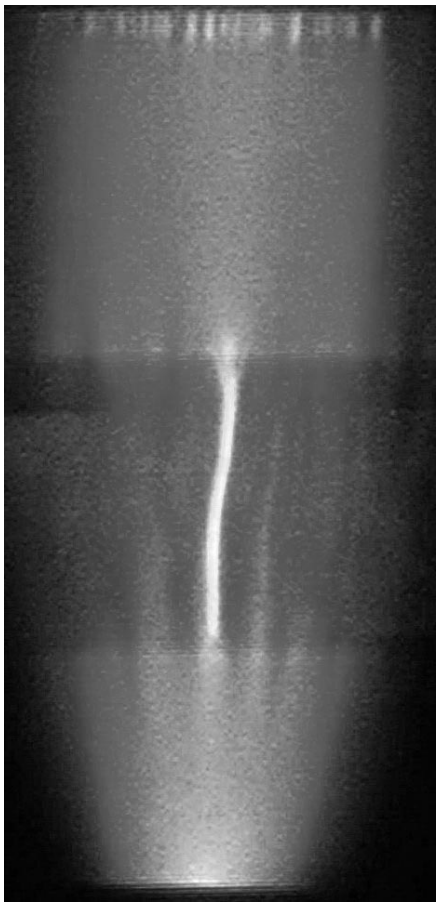

Figure 3. Photograph of SVD in SF_6 at $W_{el} = 0.12$ J cm^{-3}, $T_g = 1150$ K, $\tau_{dis} = 160$ ns, and $p = 15$ Torr.

Figure 4a shows a voltage oscillogram for a confined SVD in SF_6 at a pressure $p = 15$ Torr and $W_{el} = 0.12$ J cm^{-3}. Figure 4b presents the time dependences of voltage, calculated taking into account the following processes: electron-impact ionisation and attachment of electrons (IA), ion–ion recombination (IIR, rate constant $\beta_{ii} = 2 \times 10^{-8}$ cm^3 s^{-1} [21]), dissociative electron–ion recombination (EIR, rate constant β_{ei} varied in the range of $(0.5 - 3) \times 10^{-7}$ cm^3 s^{-1} in the calculation), electron-impact detachment of electrons from negative ions (ED, $k_d = 3 \times 10^{-7}$ cm^3 s^{-1} [22]), and electron-impact dissociation of SF_6 (EID, the dissociation energy per fluorine atom, according to different researchers, was 4–6 eV (see [23, 24] and references therein). The time dependence of current in Fig. 4b is given for the case where all these processes are taken into account. The attachment of electrons to vibrationally excited SF_6 molecules was disregarded, because in our previous experiments we found the contribution of this process to the total balance of charged particles in SVD plasma in SF_6 to be small [18]. A comparison of the calculated time dependences with the experimental SVD voltage oscillogram shows their best agreement at $k_d \approx \beta_{ei}$.

Thus, we can note three results that are most important for further analysis of the mechanisms of SVD instability development in SF_6-based gas mixtures, including working media of HF lasers: (i) SVD in the heated region has a filamentary structure with the formation of quasi-periodic plasma channels, whose spatial period depends on T_g; (ii) the development of SVD instability begins with the discharge constriction directly in the irradiated region of the discharge gap; and (iii) the increase in the concentration of electrons due to their electron-impact detachment from negative ions in the gas at room temperature is compensated for by their loss during electron–ion recombination ($k_d \approx \beta_{ei}$).

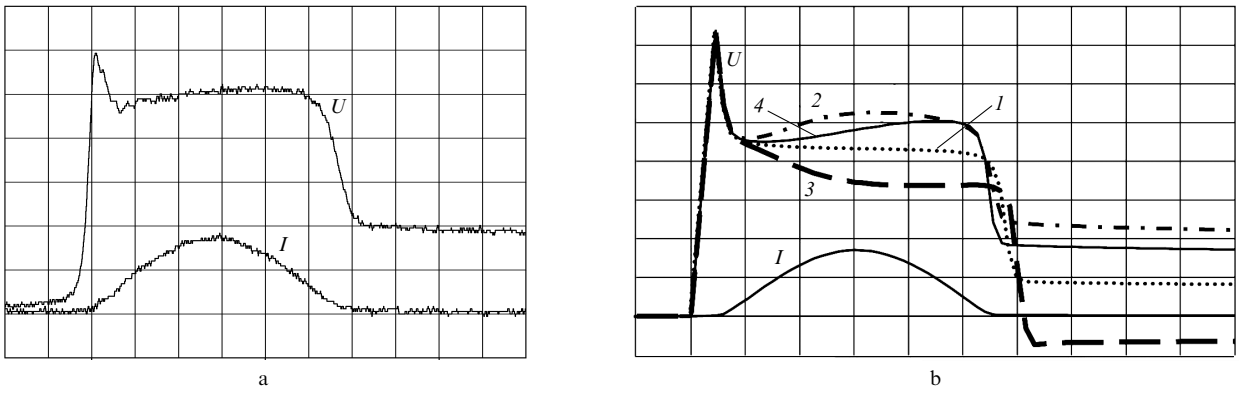

Figure 4. (a) Experimental oscillograms and (b) calculated time dependences (disregarding the cathode drop) of the SVD voltage U and current I in SF_6 at $p = 15$ Torr and interelectrode distance of 43 mm (confined discharge). The calculation was performed taking into account the following processes: (*1*) IA, (*2*) IA and EIR, (*3*) IA and ED, and (*4*) IA and EID; $k_d = \beta_{ei}$. The scale-division values are 2 kV (U), 80 A (I), and the time scale is 50 ns (sweep).

4. Results and discussion

4.1 Nonlinear mechanism of ionisation development in active media of HF(DF) lasers

SF_6^- and SF_5^- ions, which are formed as a result of electron attachment to SF_6 molecules [25], are dominant in SVD plasma. The nonlinear mechanism of electron generation in SF_6 due to the electron-impact destruction of negative ions was considered for the first time in [22], where the electron-impact detachment rate of electrons from negative SF_6^- ions was estimated to be $k_d(SF_6^-) = 3 \times 10^{-7}$ cm^3 s^{-1}. This estimation was obtained on the assumption that the detachment cross section is no less than the elastic electron scattering cross section from the SF_6 molecule. The latter parameter is known to exceed 10^{-15} cm^2 [25]. It was also taken into account that at $E/N \sim (E/N)_{cr}$ the mean electron energy $\langle\varepsilon\rangle \sim 8-10$ eV is much higher than the electron binding energy in negative SF_6^- ions (0.65–1 eV [25]), due to which the electron-impact destruction of SF_6^- ions can be considered as thresholdless.

Concerning SF_5^- ions, which have a binding energy $E_b \sim 2.8$ eV [25], the corresponding value $k_d(SF_5^-)$, with allowance for the Boltzmann factor $\exp(-E_b/T^*)$ ($T^* < 2\langle\varepsilon\rangle/3$), is smaller than $k_d(SF_6^-)$ but by no more than 40 %. Hence, when quantitatively describing the electron-impact detachment of electrons from SF_6^- and SF_5^- ions, one can use the approximation of negative ions of the same type and introduce a single detachment rate constant k_d.

A fact of fundamental importance is that no other processes of electron detachment can compete with the electron-impact destruction of negative ions in the above-mentioned ranges of pressures and times. Indeed, at $E/N \approx (E/N)_{cr}$ almost all heavy polyatomic negative ions, except for their negligible fraction in the energy spectrum tail have energies insufficient for electron detachment in collisions with neutral molecules [25], and the relatively low gas density practically excludes the effect of electronically excited components on the negative ion destruction.

The expressions for the electron concentration $n_e(t)$ were obtained in [11] based on the exact analytical solutions of the nonlinear integro-differential equation. The character of increase in $n_e(t)$ depends decisively on the parameter ξ [11]:

$$\xi = \frac{a^2/\lambda - 2}{\lambda}, \quad a = \frac{\alpha}{\eta} - 1, \quad \lambda = \frac{n_e(0)(k_d - \beta_{ei})}{\eta u_e}, \qquad (1)$$

where $n_e(0)$ is the electron concentration established by the onset of a quasi-stationary phase; α and η are the impact ionisation and attachment rates, respectively; and u_e is the electron drift velocity.

If $\xi > 0$,

$$n_e(t) = \frac{2n_e(0)b^2\lambda A}{(1-A)^2}, \quad A = \frac{a/\lambda - b}{a/\lambda + b}\exp(b\lambda\eta u_e t), \quad b = \sqrt{\xi}. \qquad (2)$$

In this case, nonlinear electron multiplication occurs against the background of significant contribution of linear processes of impact ionisation and attachment. Therefore, it is no wonder that solution (2) remains valid at arbitrarily small values of $n_e(0)$ and/or difference $k_d - \beta_{ei}$. In the limit $\lambda \to 0$ it goes, as one should expect, to the well-known 'classical' expression for electronegative gases [15]:

$$n_e(t) \approx n_e(0)\exp[(\alpha - \eta)u_e t)]. \qquad (3)$$

At $\xi < 0$ the electron-impact detachment of electrons from negative ions plays a key role, and the expression for the electron concentration,

$$n_e(t) = n_e(0)\left\{\cos^2(b_1\lambda\eta u_e t)\left[1 - \frac{a}{b_1\lambda}\tan\left(\frac{b_1\lambda\eta u_e t}{2}\right)\right]\right\}^{-1}, \qquad (4)$$

$$b_1 = \sqrt{-\xi}$$

is not transformed into (3) at any values of $n_e(0)$ and difference $k_d - \beta_{ei}$.

It follows from relations (1), (2), and (4) that, when the detachment and recombination constants are unbalanced ($k_d - \beta_{ei} > 0$), in the stage of increasing discharge current ($\alpha > \eta$), both solutions, (2) and (4), have a pronounced 'explosive' character; i.e., some finite time after the ionisation onset the electron concentration formally becomes arbitrarily high. One can easily find the corresponding characteristic 'explosion' times τ_{exp}^i from (2) and (4). If $\xi > 0$,

$$\tau_{exp}^i = \ln\left(\frac{a/\lambda + b}{a/\lambda - b}\right)\frac{1}{b\lambda\eta u_e}. \qquad (5)$$

At $\xi < 0$

$$\tau_{exp}^i = 2\arctan\left(\frac{\lambda b_1}{a}\right)\frac{1}{b_1\lambda\eta u_e}. \qquad (6)$$

In fact, this result means only that after some time ($\tau_d^i \sim \tau_{exp}^i$) the attachment of electrons is partially compensated for by their detachment from negative ions, as a result of which the electron multiplication rate sharply increases. By analogy with the terminology used in the theory of excimer lasers, this process could be referred to as the electronegativity 'burning away', because in this case we deal with not destruction of electronegative molecules but with the loss of gas electronegativity as a result of electron detachment.

When deriving relations (2) and (4), we did not take into account the small change in the time of the difference $\alpha - \eta$; however, the 'explosive' character of these solutions, which in a sense tend (in the limit) to the exact dependence $n_e(t)$, indicates that the dependence $n_e(t)$ has the same specific feature.

In the stage where the discharge current decreases, in the quasi-stationary SVD phase $\alpha < \eta$ and, correspondingly, $a < 0$. It follows from relations (2) and (4) that in this case the electron concentration always tends to zero with time. In other words, the volume multiplication of electrons as a result of their electron-impact detachment from negative ions cannot compete with the attachment-caused electron loss if the electron capture efficiency exceeds that of impact ionisation. Thus, in the quasi-stationary SVD phase the ionisation instability in the plasma volume, which is related to the 'explosive' character of electron multiplication, may develop only when the discharge current increases.

4.2 Self-organisation of SVD plasma upon laser heating of SF_6-based mixtures

The nature of self-organisation of SVD plasma in heated SF_6-based gas mixtures remains not quite clear. However,

there are some indications that this phenomenon can be due to a great extent to the development of ionisation instability in SVD plasma, caused by the above-considered electron-impact detachment of electrons from negative ions. Here are some qualitative considerations on this subject.

At high gas temperatures $k_d - \beta_{ei} > 0$; when the discharge current increases ($\alpha > \eta$), this relation leads to an 'explosive' increase in $n_e(t)$. The characteristic time τ_c of current change in the quasi-stationary SVD phase is controlled by the LC circuit ($\tau_c \sim \sqrt{LC}$); in our experiments it was about several hundreds of nanoseconds, whereas $\tau_d^i \sim 20 - 30$ ns. Since $\tau_c \gg \tau_d^i$, in the quasi-stationary SVD phase (at $E/N \approx (E/N)_{cr}$) the current should inevitably be redistributed in the discharge gap volume with the formation of current channels in the form of individual filaments (thin at large T_g values) with a higher electron concentration. Indeed, a standard linear analysis shows that under the conditions considered here specifically spatially inhomogeneous perturbations oriented perpendicularly to the current are characterised by the largest increment. Quasi-periodic structures can be formed in the nonlinear stage of perturbation development, which are experimentally observed.

According to [26], one can show that such channels in the form of separate current filaments are stable. The difference $k_d - \beta_{ei}$ also increases with an increase in the gas temperature, causing a more rapid increase in $n_e(t)$. One might suggest that specifically for this reason an increase in T_g is accompanied by an increase in the number of conducting channels and, correspondingly, a decrease in the spatial period of the current structure. Anyhow, the experimental data in Fig. 2 do not contradict this suggestion. An additional argument in favour of the above considerations is that quasi-periodic current structures did not arise at very small T_g values implemented in the experiment. Indeed, in this case $\tau_d^i \gg \tau_c$.

When the discharge current decreases the above-considered 'explosive' mechanism does not work, because $E/N < (E/N)_{cr}$ and $\alpha < \eta$ (see Section 4.1). In this case, an instability may develop on the falling portion of the quasi-static $I - U$ discharge characteristic (the $I - U$ characteristic relaxation time $\tau_{UI} \ll \tau_c$), which is controlled by the effective ionisation coefficient $\alpha_{eff}(n_e) = \alpha - \eta + (k_d - \beta_{ei})N_n(n_e)/u_e$, where $N_n(n_e)$ is the concentration of negative ions. In this case, the excess of the electron attachment rate above the impact ionisation rate is compensated for by the electron-impact detachment of electrons from negative ions. On this portion of $I - U$ characteristic SVD is unstable with respect to spatially homogeneous fluctuations of plasma parameters. However, inhomogeneous fluctuations, leading to the formation of spatial structures in the form of separate current filaments with elevated electron concentration may also increase in SVD plasma. The corresponding scenarios of self-organisation and problem of stability of the structures formed in this plasma have been considered in detail (see, for example, [26, 27]). It is likely that the instability developing on the falling portion of the $I - U$ characteristic may lead to SVD constriction at long discharge pulses (Fig. 3).

4.3 Mechanism of evolution of conducting channels in SF_6 and its mixtures

The nonlinear mechanism of electron multiplication due to the unbalance between k_d and β_{ei} may lead to the development of ionisation instability in SF_6-based gas mixtures even at room temperatures if, as was noted above, $E/N \gg (E/N)_{cr}$. Specifically this situation is implemented near the tip of a single conducting channel, developing from the side of the cathode.

The reduced electric field strength at the tip of this channel significantly exceeds the $(E/N)_{cr}$ value [15]. This leads to a significant increase in T_e and, accordingly, to a dramatic decrease in β_{ei} (see Introduction). A situation arises, where, as in the case of high gas temperature, the difference $k_d - \beta_{ei} > 0$. As a result, the above-considered mechanism of 'explosive' increase in the electron concentration is initiated and a new, plasma-filled portion of the channel is formed, which promotes the channel propagation into the discharge gap bulk. In this case, one does not need to apply the mechanism of step ionisation of SF_6 molecules [10] to explain the development of conducting channels in SF_6 and its mixtures.

5. Conclusions

We investigated a radically new mechanism of detachment instability, which develops in active media of HF(DF) lasers due to the electron-impact detachment of electrons from negative SF_6^- and SF_5^- ions. The analysis of the main mechanisms of the formation and destruction of negative ions in SF_6 and its mixtures showed that the instability is caused by the unbalance between the rate constants of electron-impact detachment of electrons from negative ions and dissociative electron–ion recombination. Analytical expressions were obtained for the time dependence of the electron concentration. It was shown that an increase in the discharge current resulted in 'explosive' instability development; the characteristic 'explosion' time was estimated. The development of detachment instability in large volumes was initiated by heating SF_6-based gas mixtures with a pulsed CO_2 laser for experimental study. The possible relationship of this process with the spatial self-organisation (formation of current filaments) in the SVD plasma in previously irradiated SF_6 and its mixtures was discussed. The mechanism of development of a single incomplete channel due to the electron-impact detachment of electrons from negative ions was considered.

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