LASERS. ACTIVE MEDIA

PACS numbers: 42.55.Lt; 52.80.Hc DOI: 10.1070/QE2002v032n02ABEH002136

Development of a self-initiated volume discharge in nonchain HF lasers[†]

V.V.Apollonov, A.A.Belevtsev, S.Yu.Kazantsev, A.V.Saifulin, K.N.Firsov

Abstract. The dynamics of a self-initiated volume discharge (SIVD) is studied in SF₆-C₂H₆ mixtures, which are used as the working media in nonchain HF lasers, employing discharge gaps of different geometry. The results of investigation are consistent with the previously made assumption that there exist mechanisms which limit the current density of a volume discharge in SF₆ and SF₆-based mixtures. SIVD simulations were performed taking into account the SF₆ dissociation by electron impact, the dissociative electron-ion recombination, the electron detachment from negative ions by electron impact, and the ion-ion recombination. The simulation results are in qualitative agreement with experimental results. The electron-ion recombination and the electron detachment from negative ions were found to cancel each other, to within the accuracy of estimates of the rate constants. The dissociation of SF₆ (and other components of the mixture) is most likely responsible for the limitation of the current density in the diffusive discharge channel and for the increase in discharge volume with increasing input energy.

Keywords: nonchain HF laser, self-sustained volume discharge, self-initiated volume discharge, SF₆.

1. Introduction

We found earlier that, to obtain a self-sustained volume discharge in SF₆ and mixtures of SF₆ with hydrocarbons (deuterocarbons) that are used as the active media of nonchain HF (DF) lasers, there is no need to preionise the gas whatsoever when small-scale ($\sim 50 \, \mu m$) inhomogeneities are deposited on the cathode surface [1–4]. A specific feature of this volume discharge, which was called [5] a self-initiated volume discharge (SIVD), is the uniform distri-

[†]Paper presented at the V International Conference on Pulsed Lasers on Atomic and Molecular Transitions (Tomsk, Russia, 2001).

V.V.Apollonov, S.Yu.Kazantsev, A.V.Saifulin, K.N.Firsov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia; e-mail: vapollo@kapella.gpi.ru

A.A.Belevtsev Institute for High Energy Densities, Joint Institute for High Temperatures, Russian Academy of Sciences, ul. Izhorskaya 13/19, 127412 Moscow, Russia; e-mail: baa@hedric.msk.su

Received 22 October 2001 Kvantovaya Elektronika 32 (2) 95-100 (2002) Translated by E.N.Ragozin bution of the energy released in the plasma volume upon the discharge ignition in the gaps with a strong boundary amplification of the electric field [2-5].

These unique SIVD properties allow a design of simple, very compact and highly efficient lasers. At present, the output energy of a nonchain SIVD HF (DF) laser amounts to ~ 400 J for an electric efficiency above 4 % [2, 3]. Further improvement of the energy parameters and optical homogeneity of the active medium of nonchain lasers calls for a comprehensive understanding of the physics of the processes governing the SIVD development at all its stages.

A SIVD discharge has an essencially jet structure [2-5] formed by separate diffusion channels attached to cathode spots. However, for high energy inputs into the discharge plasma typical of nonchain lasers, the spot density at the cathode can be as high as $10-30 \text{ cm}^{-2}$ [3, 5], resulting in a complete overlap of the channels. For this reason, the appearance and optical homogeneity of the SIVD discharge are the same as those for a preionisation discharge in such gases as, for instance, CO_2 or N_2 . The special features of this discharge were revealed in the study of its dynamics.

The authors of papers [3, 5], who studied the SIVD development by a method similar to the interrupted-discharge method, by recording the plasma emission on a photographic film, found an effect unexpected for mediumand high-pressure self-sustained gas discharges: in the absence of preionisation (or the initiation of electrons over the entire cathode surface), a local breakdown of the gap was observed in the form of one or several diffusion channels produced in the region of boundary electric field amplification. Then, the SIVD spread over the gap perpendicular to the applied field by producing new diffusion channels at a nearly invariable voltage across the plasma, gradually filling the entire gap.

According to Ref. [5], the appearance of new channels should have resulted in a reduction of the current through the channels produced at the beginning of the process (channel quenching). However, the experimental method used in this work did not allow us to gain the information on the time dependence of the plasma emission intensity and, hence, on the variation of the currents flowing through the channels, i.e., this method did not allow us to directly confirm the existence of this effect. We also showed in Ref. [5] that, for a small cathode area when only one discharge channel appeared in the form of a diffusion flame (an analogue of a solitary diffusion channel), the latter expanded with increasing energy introduced into the plasma.

Therefore, we found [5] that one of the main properties of a SIVD is the increase in its volume with increasing

energy introduced into the plasma. The volume is increased due to either the increase in the number of diffusive channels or (where new channels cannot be produced) due to the increase of the volume of already existing channels. As pointed out in Refs [5–7], these SIVD properties may be related to the existence of mechanisms responsible for limiting the current density of a diffusion discharge in SF₆ and SF₆-based mixtures, which prevent the total energy from flowing through only one channel.

In this paper, we studied the SIVD by the methods that allow us to observe the variation of its characteristics in space and time. The aim of our work is to study in more detail the SIVD development in discharge gaps with different geometry and to reveal the main parameters that govern this development.

2. Experimental setup

The SIVD dynamics was studied in discharge gaps with different geometry. The scheme of the setup for investigating the discharge in a gap with a linear cathode geometry is shown in Fig. 1a. The SIVD with a total duration of \sim 370 ns was ignited in the SF₆: C₂H₆ = 10:1 mixture at a pressure p = 33 Torr and an interelectrode distance of 4 cm. The electrodes were a 0.5-mm thick copper bar 16 cm in length (a knife-edge cathode) placed edgewise and a disk anode 6 cm in diameter rounded over the perimeter with a radius of curvature of 1 cm. The breakdown was forcedly initiated at the centre of the cathode with a weak-current spark limited with an $R = 900 \Omega$ resistor. This spark could not provide an initial electron density in the gas high enough for the development of a volume discharge, but allowed fixing the position of the primary gap breakdown. The SIVD glow was recorded with a single-frame streak camera tube with a frame exposure time of 20 ns, which was triggered with a variable delay T relative to the instant of gap breakdown.

To study the SIVD dynamics in the plane gap geometry, we used the circuit with a sectioned cathode shown in Fig. 1b. In this case, the interelectrode distance, the working mixture pressure, and the electrical circuit of the setup were the same as in the previous experiment, except that the cathode was not a copper bar, but a disk 6 cm in diameter rounded over the perimeter with a radius of curvature of 1 cm and subjected to sandblast processing [2-5]. Insulated conductors 1 mm in diameter were inserted into holes 2 mm in diameter drilled ~ 4 cm apart through the cathode in its plane region. One of the conductors (1) protruded above the cathode surface by ~ 1 mm to ensure the occurrence of the primary gap breakdown at precisely this point. By comparing the oscilloscope traces through the ignitor (1) and monitoring (2) electrodes, we could trace the SIVD spread over the gap.

The dynamics of an individual diffusion channel was investigated using the setup shown in Fig. 1c. The diffusion channel was simulated by a discharge with a rod (cathode) – plane gap geometry at a pressure p = 16.5 - 49.5 Torr of the SF₆: $C_2H_6 = 10:1$ mixture for an interelectrode distance of 4 cm. The cathode was the end of a polyethylene-insulated wire with a core diameter of 1.5 mm, and the anode was a disk 10 cm in diameter. The SIVD development was monitored with the streak cameras, as in the experiments performed using the setup of Fig. 1a.

In the circuits shown in Figs 1a and 1b, the capacitor C

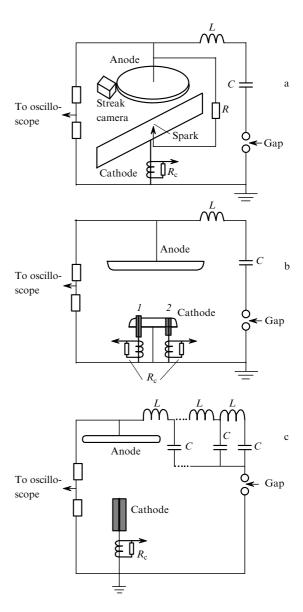


Figure 1. Schemes of experimental setups for studying the SIVD dynamics: ignitor (1) and monitoring (2) conductors.

discharged into the gap via inductance L upon switching spark gap. In the circuit of Fig. 1c, an artificial line discharged into the gap. The currents in the circuits of Fig. 1 were monitored with Rogowski coil R_c , the voltage across the gaps was measured with resistive dividers.

3. Experimental results

Fig. 2 shows the streak-camera images of a SIVD in the gap with a knife-edge cathode obtained at different instants of time. The corresponding oscilloscope traces of discharge current and voltage are given in Fig. 3. One can see from Fig. 2 that the gap is broken down at the centre (at the region of the auxiliary spark discharge). At this instant of time, the SIVD has the form of one diffusion channel, with a cathode spot already formed. Then, new channels appear near the first channel, which have a significantly lower brightness, and are developing at a voltage close to the static breakdown voltage for SF₆ (see Fig. 3).

The number of new channels increases with time, they

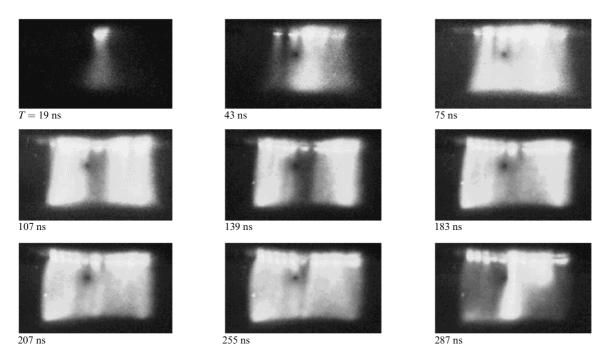


Figure 2. Streak camera images obtained at different instants of time T.

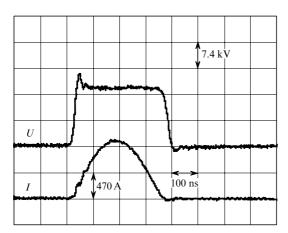


Figure 3. Oscilloscope traces of the voltage and current of a SIVD.

'disperse' from the middle to the periphery of the gap, and their brightness gradually achieves that of the first channel, the channels closest to the first channel being the brightest. All the channels become equally bright with time, while the brightness of the first channel decreases significantly. Upon a further SIVD development, the brightness of the channels at the gap periphery (i.e., the channels removed from the primary breakdown region) gradually increases. However, for T > 210 ns the glow becomes uniform again over the cathode length, the glow in the first channel also being restored. This effect will be referred to as a current return (to the channel). Then, we observed the development of discharge instability against the background of the total diffusion glow.

Therefore, the results outlined above confirm directly the fact that the SIVD spreads over the gap at a voltage close to the static breakdown voltage in the form of a sequence of diffusion channels. In this case, the first channel, which was produced upon the gap breakdown, fades away as new

channels emerge. Note also that the current return (see Fig. 2), which is manifested in the equalisation of the brightness of all the channels and in the increase in the brightness of the first channel following its initial decrease, is observed only when the energy introduced into the plasma is high enough. This effect vanishes when the energy is lowered or the linear size of the cathode is increased. For high energy inputs (150–200 J L⁻¹) typical of HF (DF) lasers, the SIVD spreading over the gap is so fast that the time resolution of our streak camera is insufficient.

A similar pattern of SIVD development is also observed with the plane discharge gap geometry. Fig. 4 shows the oscilloscope traces from the ignitor and monitoring conductors (see Fig. 1b) obtained in the experiment with the sectioned cathode. One can see from Fig. 4 that the current begins to flow through the monitoring conductor with a significant delay relative to the current through the ignitor.

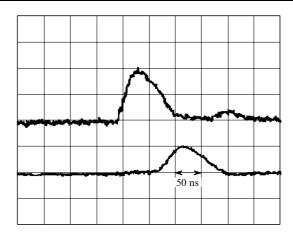


Figure 4. Oscilloscope traces of the current through the ignitor (the upper trace) and monitoring (the lower trace) conductors.

By the time of onset of the current through the monitoring conductor, the amplitude of the current through the ignitor lowers by more than a factor of two, i.e., in this experiment, too, we observed quenching of the initially produced channel upon the emergence of subsequent channels. One can also see from the oscilloscope trace of the current through the ignitor (Fig. 4) that a current returns to the first channel after its almost complete quenching in the case of a plane gap as well, provided the energy input is high enough.

In the rod-plane gap, the spreading of the flame, which simulated a single diffusion channel, was observed to occur in two stages. During the first 20-30 ns following the gap breakdown, the discharge volume rapidly increased, out of proportion to the energy inputted. Then, the discharge volume increased approximately linearly with the energy. For a constant total energy W introduced into the plasma, the largest volume V occupied by the SIVD by the instant of energy input termination increased with lowering the mixture pressure p. Fig. 5 shows the dependence of V on the parameter W/p obtained for different p. One can see that this dependence is satisfactorily, to within the error of experimental determination of V, described by a linear function. This supposedly reflects the fact that, beginning with some volume $V = V_0$ determined by the stage of initial fast channel spreading, the discharge volume increases at the approximately constant parameter $\Delta W/(p\Delta V) \approx \text{const} \approx$ 2.8 mJ cm⁻³ Torr⁻¹, where $\Delta V = V - V_0$ is the volume increase and ΔW is the energy inputted into the volume ΔV .

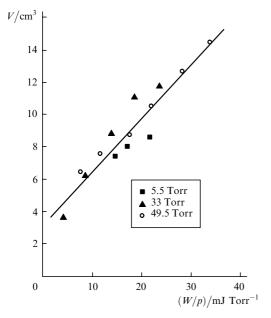


Figure 5. Dependences of the volume V occupied by discharge on the parameter W/p for the SF₆: $C_2H_6 = 10:1$ mixture at a pressure p.

4. Discussion of results

The results presented above confirm assumptions that there exist mechanisms of current density limitation in the diffusion channel of a discharge in SF_6 and SF_6 -based mixtures [5–7] (and also in several other strongly electronegative gases [6, 7]). It would be reasonable to attempt to relate these mechanisms to the specific feature of SF_6 , namely, with its strong electronegativity.

Strongly electronegative gases have high reduced electric-field strengths E/N (E is the field strength and N is the concentration of neutral molecules) at which an electrical breakdown occurs and the energy is inputted into the plasma. They also have large electronegativity parameters χ_a defined as the ratio of concentrations of negative ions and electrons. For this reason, the following features of the charge kinetics inherent in these gases should be taken into account in the study of a volume self-sustained discharge in strongly electronegative gases such as SF_6 and SF_6 -based laser mixtures.

First, the electron-impact dissociation of SF_6 molecules and other components of the mixture plays a significant role in this discharge. Indeed, for high E/N close to the critical value $(E/N)_{cr} = 360$ Td, the average energy of plasma electrons in the quasi-stationary stage of a self-sustained discharge approaches the SF_6 dissociation threshold and more than 80 % of the energy inputted into the discharge is spent to the dissociation [8, 9] resulting in the production of an F atom. In the submicrosecond range typical for the discharge under study, decomposition products have no time to escape from the discharge channel, resulting in a local increase in the concentration N of particles, a decrease in E/N, and a decrease in the electric conductivity of the diffusion channel due to the increase in electron losses through attachment.

Second, since the parameter χ_a is significantly greater than unity, high negative ion concentrations are attained in the discharge, with the effect that the electron detachment from negative ions can make a significant contribution to the balance of charged particles. Usually the disintegration of negative ions in their collisions with neutral molecules and/or due to associative ionisation is taken into account. However, for medium gas pressures (p = 10 - 100 Torr) and a submicrosecond discharge duration, these processes do not make any noticeable contribution to electron multiplication [10]. However, there are firm grounds to believe that the electron-impact detachment of electrons from negative ions can be an efficient channel of the delivery of secondary electrons to the discharge plasma [11].

Third, a significant influence on the parameters of an SF₆-discharge plasma, including its conductivity, can be exerted by dissociative electron—ion recombination because high concentrations of positive ions are achieved in the discharge, and the energy is mainly inputted into the plasma when $E/N \approx (E/N)_{\rm cr}$, i.e., for $\alpha \approx \eta$ (where α is the ionisation rate coefficient and η is the electron attachment rate coefficient). Ion—ion recombination must also be taken into account, because it may significantly limit the concentration of ions in the plasma.

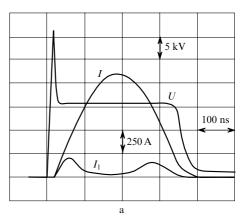
Therefore, it follows from the above discussion that the main mechanisms of SIVD current density limitation are the dissociation of SF_6 molecules by electron impact resulting in a decrease in the ionisation rate and an increase in the electron attachment rate due to the local reduction of the parameter E/N in the diffusion channel, and also electron—ion recombination, which is responsible for the growth of electron losses with increasing current density in the channel. It was shown qualitatively [5] that the current density can be also limited due to the attachment of electrons to vibrationally excited SF_6 molecules. However, the absence of reliable data on the rate constants for these reactions in the literature complicates a quantitative estimate of the role of this process in the balance of charged particles in the plasma.

We performed SIVD simulations to verify whether the above assumptions are consistent with the available experimental data. The channel structure of the discharge was modelled with a set of resistive elements connected in parallel, whose resistance was determined by the formula $R_c = U/[Se(n_i^-v_i^- + n_i^+v_i^+ + n_ev_e)],$ where U is the voltage across the discharge gap; S is the cathode area; e is the electron charge; n_i^- , n_i^+ , n_e , v_i^- , v_i^+ , and v_e are concentrations and drift velocities of negative and positive ions and electrons, respectively. Similarly to Ref. [12], the concentrations of particles were determined by solving continuity equations for particles of each sort in combination with the Kirchhoff equations for the discharge circuit. The nonuniformity of initial conditions for the development of channels along the cathode length was simulated by prescribing different initial electron concentrations in each of the channels. In addition to the electron-impact ionisation of SF₆ and electron attachment to SF₆ molecules, the following processes were included in the calculation.

- (1) SF_6 dissociation by electron impact. The number of dissociated molecules was defined as $N_d = W/q_F$, where W is the energy inputted into the discharge and $q_F = 4.5$ eV is the energy spent to produce an F atom [13].
- (2) Electron detachment from negative ions by electron impact. It was assumed that SF_6^- negative ions dominate in the plasma because the charge exchange reactions have no time to occur during the discharge period, while the cross sections for the electron-impact production of other negative ions are too small [9, 10]. The rate constant k_d for the disruption of negative ions by electron impact was estimated as 10^{-7} cm³ s⁻¹ assuming that it should not be smaller than the rate constant for elastic electron scattering by SF_6 molecules [11].
- (3) Dissociative electron-ion recombination. The rate constant for this process $\beta_{\rm ei} = 10^{-7}~{\rm cm}^3~{\rm s}^{-1}$ was estimated assuming that the SF₆ discharge plasma is dominated by SF₅⁺ positive ions [9] and that $\beta_{\rm ei} \sim T_{\rm e}^{-1/2}$, where $T_{\rm e}$ is the electron temperature.
- (4) Ion-ion recombination. In this case, the rate constant $\beta_{ii} = 2 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ for the E/N values close to the critical value was taken from Ref. [14].

We simulated an SIVD with nine channels, which corresponds approximately to the conditions of the experiment performed using the setup of Fig. 1a. Fig. 6 shows the oscilloscope traces of the voltage U, the total discharge current I, and the current I_1 through a single channel (for which we prescribed the maximum initial electron concentration) calculated for the energy density inputted into the discharge plasma equal to 80 and 40 J L⁻¹. One can see from Fig. 6a that the current through a single channel has two maxima when the energy input is high. This is in qualitative agreement with the oscilloscope trace of current through the ignitor (see Fig. 4) in the plane gap geometry and in agreement with the experimentally observed redistribution of the channel glow intensity upon the SIVD development in the knife edge-plane gap (see Fig. 2). The current return vanishes when the energy input is lowered (Fig. 6b).

Calculations also show that the electron detachment by electron impact and electron—ion recombination virtually compensate each other within the accuracy of estimates of the rate constants $k_{\rm d}$ and $\beta_{\rm ei}$. The dissociation of SF₆ molecules and other components of the HF(DF)-laser mixture should therefore be considered as the principal



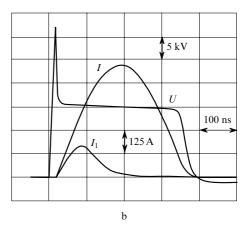


Figure 6. Calculated oscilloscope traces of the voltage U, the total discharge current I, and the current I_1 through the first channel for energy density 80 (a) and 40 J L⁻¹ (b).

mechanism responsible for current density limitation in the SIVD channel. However, it is reasonable to refine the magnitudes of k_d and β_{ei} . Note that our model (like any zero-dimensional model which neglects the time variation of the SIVD volume) pretends only to a qualitative illustration of the current redistribution in the channels when account is taken of possible mechanisms of current density limitation.

Note also that the electron detachment from negative ions by electron impact should be taken into account in the analysis of the processes determining the instability development of the volume discharge in SF_6 and other strongly electronegative gases.

Once again we draw attention to the result that follows from the investigation of the dynamics of a single diffusion channel: an SIVD develops at an approximately constant value of the energy parameter W/(Vp).

5. Conclusions

The results of investigations of the SIVD dynamics in discharge gaps of different geometry are consistent with the assumption that the development of this discharge is governed by the mechanisms of limitation of the current density in a diffusion channel. We assume that the most significant process responsible for the current limitation and determining the SIVD properties is the electron–impact dissociation of SF_6 and other components of the mixtures of nonchain HF (DF) lasers.

References

- Apollonov V.V., Kazantsev S.Yu., Oreshkin V.F., Firsov K.N. Pis'ma Zh. Tekh. Fiz., 22, 60 (1996).
- Apollonov V.V., Kazantsev S.Yu., Oreshkin V.F., Saifulin A.V., Firsov K.N. Izv. Akad. Nauk, Ser. Fiz., 64, 1439 (2000).
- Apollonov V.V., Firsov K.N., Kazantsev S.Yu., Oreshkin V.F., Saifulin A.V. Proc. SPIE Int. Soc. Opt. Eng., 3886, 370 (1999).
- Apollonov V.V., Kazantsev S.Yu., Oreshkin V.F., Firsov K.N. II Mezhdunar. konf. po fizike plazmy i plazmennym tekhnologiyam (II International Conference on the Plasma Physics and Plasma Technologies) (Minsk, 1997) p. 154.
- Apollonov V.V., Belevtsev A.A., Kazantsev S.Yu., Saifulin A.V., Firsov K.N. Kvantovaya Elektron., 30, 207 (2000) [Quantum Electron., 30, 207 (2000)].
- Apollonov V.V., Belevtsev A.A., Firsov K.N., Kazantsev S.Yu., Saifulin A.V. Proc. XIII Intern. Conf. on Gas Discharge and their Applications (Glasgow, UK, 2000) Vol. 1, p. 409.
- Apollonov V.V., Belevtsev A.A., Firsov K.N., Kazantsev S.Yu., Saifulin A.V. Proc. XXV Intern. Conf. on Phenomena in Ionized Gases (Nagoya, Japan, 2001) Vol. 1, p. 255.
- Slovetskii D.I., Deryugin A.A., in *Khimiya plazmy* (Plasma Chemistry) (Moscow: Energoizdat, 1987) p. 240.
- Nakano N., Shimura N., Petrovic Z.L., Makabe T. *Phys. Rev. E*, 49, 4455 (1994).
- Hilmert H., Schmidt W.F. J. Phys. D: Appl. Phys., 24, 915 (1991).
- 11. Belevtsev A.A., Biberman L.M. Izv. Akad. Nauk SSSR, Ser. Energ. Transp., 3, 74 (1976).
- Apollonov V.V., Kazantsev S.Yu., Saifulin A.V., Firsov K.N. *Kvantovaya Elektron.*, 30, 483 (2000) [*Quantum Electron.*, 30, 483 (2000)].
- 13. Gordon E.B., Matyushenko V.I., Repin P.B., Sizov V.D. *Khim. Fiz.*, **8**, 1212 (1989).
- Apollonov V.V., Belevtsev A.A., Kazantsev S.Yu., Saifulin A.V., Firsov K.N. Kvantovaya Elektron., 31, 629 (2001) [Quantum Electron., 31, 629 (2001)].