Research & Reviews: Journal of Pure and Applied Physics

e-ISSN: 2320-2459
p-ISSN: 2347-2316

High-Energy HF (DF) Lasers Based on Non-Chain Chemical Reaction

Victor Victorovich Apollonov* and Sergey Yu. Kazantsev

A.M. Prokhorov GPI RAS, Vavilov Str. 38, Moscow, 119991, Russia

Book Review

Received date: 29/01/2020

Accepted date: 22/02/2020

Published date: 03/03/2020

*For Correspondence

A.M. Prokhorov GPI RAS, Vavilov Str. 38, Moscow, 119991, Russia.

E-mail: vapollo@kapella.gpi.ru

Keywords: High-energy, Lasers, Deuterium, DF laser radiation electric discharge

ABSTRACT

The goal of the presented paper is to advertise the book which is going to be published by Cambridge International Science Publishing Ltd. at the end of 2019. The book content reflects the scientific activity of authors in the area of ecologically safe chemical lasers. Prof. Victor V. Apollonov is the leader of research and scientific advisor of the doctoral dissertation of his colleague and pupil-Sergey Yu. Kazantsev. The following will be focused on solving two fundamental problems:

- The formation of a scalable self-sustained volume discharge in strongly electronegative gases and the creation of wide-aperture high-energy non-chain HF (DF) chemical lasers
- Expanding the generation spectrum of non-chain HF (DF) chemical lasers and creating lasers with high energy per pulse, pulsed and average power in the spectral range >4.1 μm

INTRODUCTION

Chemical HF (DF) Lasers. The Principle of Operation and General Characteristics

With the discovery of chemical lasers (CL) high hopes were pinned on them. They were connected with the possibilities of their use in various fields, mainly in the field of military applications, in power engineering, in the creation of laser engines, etc., i.e. in those areas where laser beams with low divergence and extremely high energy characteristics are necessary [1-4]. A distinctive feature of the CL is the possibility of obtaining laser energy with an efficiency higher than 100%, i.e. the emitted energy may be several times higher than the energy expended to initiate a chemical reaction. A classic example of such a laser is a chain HF (DF) laser, in which lasing in the mid-IR range occurs on the vibrational-rotational transitions of the HF (DF) molecule, which is formed when fluorine gas interacts with hydrogen (deuterium). The characteristic reactions of the processes are as follows:

$H+F_2 \rightarrow HF(v)+F$; $F+H_2 \rightarrow HF(v)+D$;

$HF(v)+hv \rightarrow HF(v_1)+2\ h\nu\lambda = 2.5\div3.1\ \mu m$ (1.1)

$D+F_2 \rightarrow DF(v)+F$; $F+D_2 \rightarrow DF(v)+D$;

$DF(v)+hv \rightarrow DF(v_1)+2\ h\nu\lambda=3.5\div4.8\ \mu m$ (1.2)

The energy characteristics of the CL are record-breaking among the whole class of gas lasers. An additional factor that stimulated the development of HF (DF) laser research was that the DF laser radiation is little absorbed by atmospheric gases (falls into the atmospheric transparency window) (**Figure 1**). **Figure 1** shows the dependence of the atmospheric transmission on the radiation wavelength [5].

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p-ISSN: 2347-2316

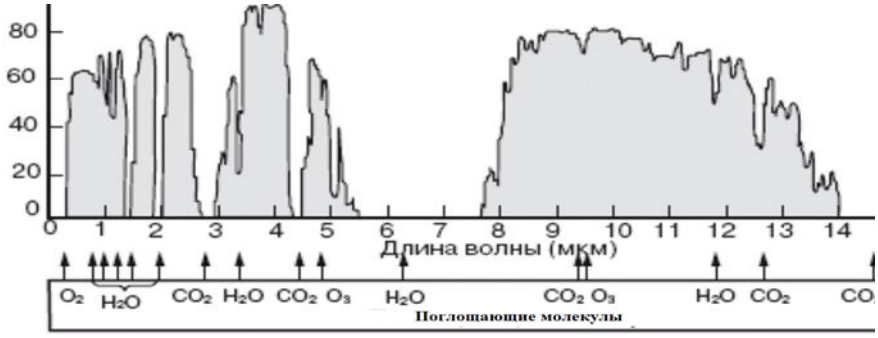

Figure 1. Transmission of the atmospheric surface layer (15°C, humidity 40%). Shaded areas are atmospheric transparency windows [5,6].

Due to the high interest in the DF laser from the military-industrial complex [6,7], by the beginning of this century, laser systems of the megawatt level had already been created, including in the mobile version [8]. The main drawback of the chain CL, greatly limiting their field of application, is the high toxicity and explosiveness of the starting components. It is because of this that it is possible to work with chain HF (DF) lasers only on specially equipped sites. Another significant drawback of the chain CL is the difficulty of implementing the IP mode, especially when a number of applications require work with a high pulse repetition rate (~100 Hz and more). At the same time, lasers on non-chain reactions lack the disadvantages listed above [3,9]. The efficiency and energy parameters of non-chain HF(DF) lasers are not as high as those of chain analogs, but the lasing spectrum also lies in the average IR range (λ=2.6 ÷ 3 μm for the HF laser and λ=3.6 ÷ 4.1 μm for the DF laser), while they are safer and more convenient to use. The first papers describing the generation of non-chain lasers based on vibrational-rotational transitions of HF and DF molecules appeared in 1967 [10,11], i.e. long before the CL were created on chain reactions. Over the next 50 years, interest in the non-chain HF (DF) CLs was unstable. Periods of a sharp increase in the number of publications devoted to HF (DF) laser studies (or their applications) alternated with periods of decline, when the activity of research groups in this area decreased markedly. The uneven activity of scientific groups working in the field of the non-chain HF (DF) CL is due to several reasons. First, the progress in the development of such lasers required for their decision a deeper understanding of the physical processes occurring in the gas-discharge plasma, the application of new approaches, the development of technologies or the development of a new elemental base. The second reason is associated with progress in the field of creating solid-state lasers, which gave hope for obtaining comparable energy characteristics, including in the spectral region of 2.7 μm-4.5 μm. This is the reason for the decrease in the level of funding for other areas of research. However, even today, in the spectral region of 2.6 μm-4.2 μm, the energy characteristics-pulse energy, pulsed and average power, of the non-chain HF (DF) CLs are still significantly superior to solid-state lasers.

The principle of the non-chain HF (DF) CL is based on the fact that in a laser working medium, which is a stable mixture of fluorine and hydrogen-containing (deuterium-containing) gases, a non-chain reaction is initiated by the input of energy from an external source. Molecules of the starting materials dissociate to form chemically active centres (fluorine atoms), the reactivity of which is many times greater than the reactivity of the starting materials. As a result of the production of fluorine atoms in the working volume, it becomes possible for a chemical reaction to form HF (DF) molecules in a vibrationally excited state [3,9]. In a non-chain laser, the active centres form only as a result of the external energy source, and after the cessation of energy supply, the chemical reaction quickly fades [12]. In a simplified form, these processes can be written as follows:

1) $W+AF \rightarrow A+F$-dissociation of the fluorine-containing component AF due to the supply from an external energy source W

2) $F+RH\ (RD) \rightarrow HF^{*}\ (DF^{*})$+R-interaction of F atoms with a hydrogen or deuterium-containing component RH (RD), as a result of which a vibrationally excited molecule HF (n) (or DF (n) is formed), where n is the vibrational quantum number (n>0)

3) $hv+HF\ (n) \rightarrow HF\ (n-1)+2$ hv-radiation of a quantum of light

As a working medium of a non-chain laser, a mixture of SF_6 with hydrogen (deuterium) is mainly used. SF_6 is a non-toxic, chemically stable compound that is used in firefighting as a gas flame arrester, and in high-voltage technology as a gas electrical insulation [13].

Reactions in the SF_6:H_2 (D_2) gas mixture become possible only after SF_2 molecules dissociate with the formation of fluorine atoms due to an external energy source.

A schematic diagram of the IP of a non-chain HF (DF) CLs is shown in **Figure 2**. The flow of non-reacting gases is pumped through laser zone 2, where a non-chain chemical reaction is initiated from an external energy source. The reaction in this mixture is possible only while the fluorine atoms are being generated. After the laser zone, the mixture is pumped through filter 3, on which HF (DF) molecules and SF_6 dissociation products are deposited, and the mixture again enters the laser

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zone. The degree of dissociation in the laser zone, as a rule, does not exceed 5%; therefore, the gas consumption for one cycle is insignificant (in real lasers, usually the volume of the laser chamber significantly exceeds the volume of the laser zone) [9]. During long-term operation with a high pulse repetition rate in a non-chain HF (DF) CL, a continuous supply of a fresh mixture is carried out to compensate for the reacted components.

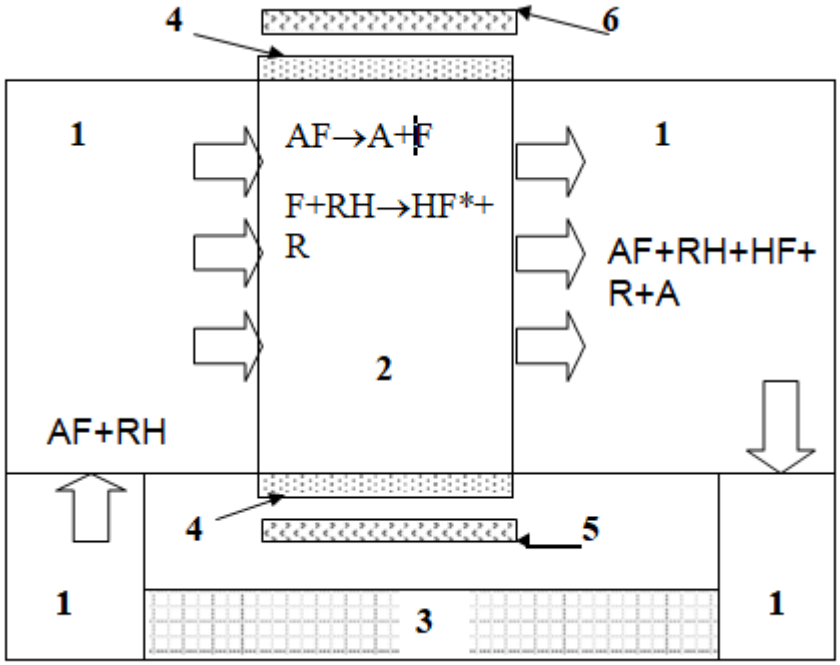

Figure 2. I-P scheme of non-chain HF (DF) laser: 1: chamber; 2: laser zone, where using an external energy source cause the dissociation of the fluorine-containing component; 3: filter for binding HF (DF); 4:windows for outputting radiation to the resonator; 5,6: resonator mirrors.

The non-chain HF (DF) CLs are characterized by large values of the gain, while lasing occurs according to a cascade mechanism, i.e. the lower laser level of the previous transition is the upper for the next [14]. For example, transitions in the vibrational bands n=3 → n=2, n=2 → n=1, n=1 → n=0 are observed in an electric-discharge HF laser based on SF_6:H_2 mixtures [3]. In this regard, the laser has a rather complex spectrum of output radiation, which is a large number of closely spaced lines corresponding to different rotational numbers of vibrational transitions [2,10]. **Figure 3** shows a typical lasing spectrum of a chemical DF laser [15]. (The spectrum of a non-chain laser is usually poorer [3], but depending on the particular implementation in the spectral region up to 4.1 μm, it is similar to the lasing spectrum shown in **Figure 3.** When using a selective cavity mirror or introducing a selective absorber inside the cavity, it is possible to control the spectral composition of non-chain CLs quite efficiently, highlighting certain vibrational-rotational lines [16].

Table 1. The energy effect of the chemical reaction of the interaction of fluorine atoms with various hydrogen donors RH (deuterium RD) and the fraction of this energy (η), which is used to excite vibrational levels of the HF (DF) molecule.

F+RH(RD)=HF(DF)+R	Q eV	Q kcal/ mol	Chemical efficiency η x
H2	1.43	32.9	0.71
D2	1.4	21.7	0.7
HI	1.95	44.8	0.95
HBr	2.11	48.5	0.24

e-ISSN: 2320-2459
p-ISSN: 2347-2316

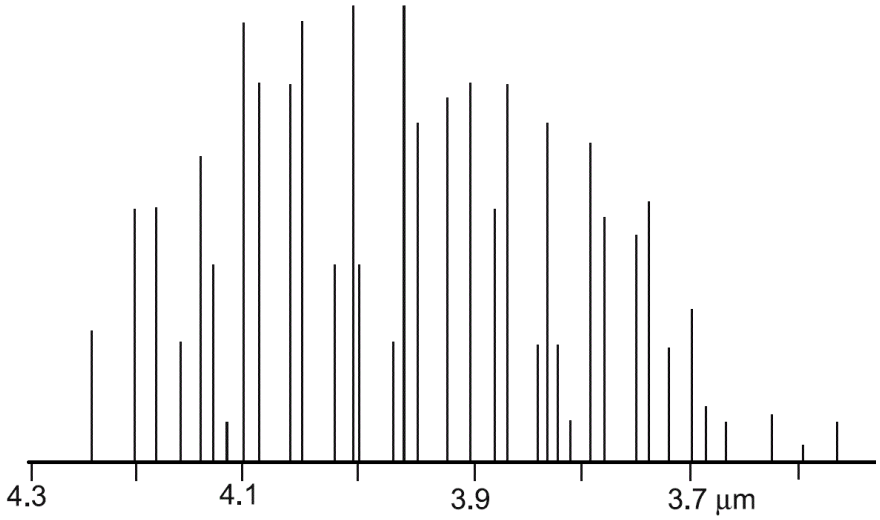

Figure 3. Relative intensity of lasing lines of a pulsed DF laser [15].

The amount of energy concentrated in the vibrational degrees of freedom of the HF (DF) molecules is largely determined by what substance was used as a hydrogen (deuterium) donor [3,17,18]. **Table 1** presents data on the amount of energy released during the chemical exothermic reaction (Q), as well as its fraction (η_x), which is used for vibrational excitation of the HF (DF) molecule for various hydrogen donors (deuterium). The value of η_x is at the same time the limiting value of the chemical efficiency of an HF (DF) CL in mixtures containing the RH (RD) molecules. It should be noted that Q and η_x do not depend on the specific form of the fluorine-containing compound AF since the AF compounds do not directly interact with RH, and a false assumption may arise that the lasing spectrum of the HF (DF) laser also does not depend on which fluorine-containing compound the substance is used. However, this is not the case. The fact is that AF dissociation products are formed not only in the ground state, but also in the excited state, and thus carry some excess energy, the magnitude of which, with the chosen initiation method, depends on AF [3]. The choice of AF also has a direct effect on the overall efficiency of CL, since the main energy consumption in non-chain HF (DF) CLs is the dissociation of AF fluoride molecules to form F atoms. Most studies have been carried out on lasers in which fluoride dissociation occurred due to electron impact, in the literature this type of lasers is also called electrochemical. It should be noted that the difference in the energy consumption for the dissociation of AF molecules leads to different absolute values of the gain of the active medium of non-chain HF (DF) CLs, which also affects the lasing spectrum [18,19].

In [20], it was shown that the problem of creating an effective non-chain HF (DF) CL reduces, firstly, to the choice of suitable fluorine and hydrogen-containing gases, in which, with minimal initiation energy, it is possible to obtain the maximum output energy of the laser $W_{out,}$ and second, to the choice of an effective method of initiating a non-chain chemical reaction.

The method based on the dissociation of fluorine-containing molecules by electron impact, carried out in an SSVD, has the greatest advantages (low energy costs for the formation of fluorine atoms, the possibility of exciting large volumes of the medium, etc.) over all other methods of initiating non-chain chemical reactions [20]. In contrast to photolysis, this method is also more versatile, since it allows the use of a much wider range of fluorine-containing compounds, and in contrast to the use of an electron beam, it does not require sophisticated protection from X-rays and has higher characteristics in terms of structural reliability [21].

At present, among various gas lasers, it is electric discharge systems that have found the greatest application. Electric-discharge non-chain HF (DF) CLs are used in medicine [22-25], for optical pumping of crystals [26], in laser chemistry [27-29], in military science [7]. for recording holograms [30] and in environmental monitoring [5,31,32]. In this regard, we consider electric discharge HF (DF) CLs in more detail.

HF (DF) Lasers with the Initiation of Non-Chain Chemical Reaction by Electric Discharge

The first lasing on the HF in an electric discharge was obtained by Deutsch in 1967 [10]. After that, the main effort of researchers was focused on finding the most appropriate fluorine and hydrogen-containing compounds and improving the methods for obtaining the discharge [17,33-35]. In the first works, schemes with a longitudinal electric discharge were used (the highest characteristics of the HF laser with this method of excitation were obtained by Zapolskiy AF [36] but the possibility of increasing the working volume and laser energy was limited by the need to supply extremely high voltages to the electrodes. Therefore, almost immediately after the appearance of the first publications on the creation of a CO_2 laser with pumped transverse SSVD, the methods developed for this purpose began to be used to initiate a chemical reaction in

e-ISSN: 2320-2459
p-ISSN: 2347-2316

non-chain HF (DF) CLs [37-42]. It is this type of electric discharge that allows more energy to be introduced into the gas and to excite large volumes of the active medium [43,44]. Independently of one another by many experimental groups, it was found that the best parameters of a non-chain HF laser are realized when SSVD is ignited in the SF_6:H_2 or SF_6: C_2H_6 working mixture.

It should be clarified why the SSVD in gas mixtures containing fluorine atoms as SF_6 as a donor turned out to be the most effective way to initiate a chemical reaction in the non-chain HF (DF) CLs. When an electric current flows in a gas, the main parameter that determines such parameters as the average electron energy, ionization constants, adherence, etc., is the parameter E/N, where E is the magnitude of the electric field strength and N is the number of gas particles per unit volume [45]. **Figure 4** shows the dependences of the ionization (α) and sticking (η) coefficients on E/N in SF_6 [13]. The point of intersection of these curves determines the reduced critical field $(E/N)_{cr}$, which remains approximately constant during the SSVD discharge pulse [46-48]. The value of $(E/N)_{cr}$ sets the average electron energy in the discharge and the rate constant for various plasma-chemical processes [45]. It is noteworthy that due to the strong electronegativity of SF_6, electron multiplication occurs at a very high value of the parameter $(E/N)_{cr}$ and the average electron energy in an independent discharge is almost 10 eV, with more than 80% of the energy going to dissociate SF6 with the formation of fluorine atoms [49]. The value of the parameter $(E/N)_{cr}$=360 Ta, which is set in the discharge, is close to the optimum for SF_6 dissociation (1 Ta=10-17 V cm^2). As a result, the energy cost of the formation of fluorine atoms in SF_6 is the lowest among stable and non-toxic fluorides-Ef ≈ 4 eV [50,51]. **Table 2** shows the values of the energy cost of a fluorine atom during electron beam dissociation of various molecules. Note that in some respects a non-chain HF laser with electric discharge initiation turned out to be much simpler than other lasers excited by an SSVD. For example, for CO_2 laser, the value of (E/N) which is established in the SSVD, is higher than the optimum, and vice versa in nitrogen, and we have to resort to various tricks to increase the efficiency of these lasers [52]. In working mixtures of the non-chain HF (DF) laser, nothing of the kind needs to be done it is only necessary to obtain a uniform discharge in the laser working mixture. As the technique of producing SSVDs was improved, many different schemes of electric-discharge non-chain HF lasers were proposed [2,20]. In fact, all these systems reproduced the electrical circuits and design of discharge chambers, which were previously developed for CO_2 and excimer lasers, without taking into account the specifics of the gas mixtures used in HF laser working environments. Therefore, the radiation energy of the most efficient non-chain electric-discharge HF laser until 1996 (beginning of this work) did not exceed 11 J [53]. and from 1970 to 1996 the radiation energy of CO_2 lasers pumped by an electric discharge increased by more than three orders of magnitude, and by the beginning of this century, it reached~5 kJ [54]. Excimer lasers were also developed rapidly [55].

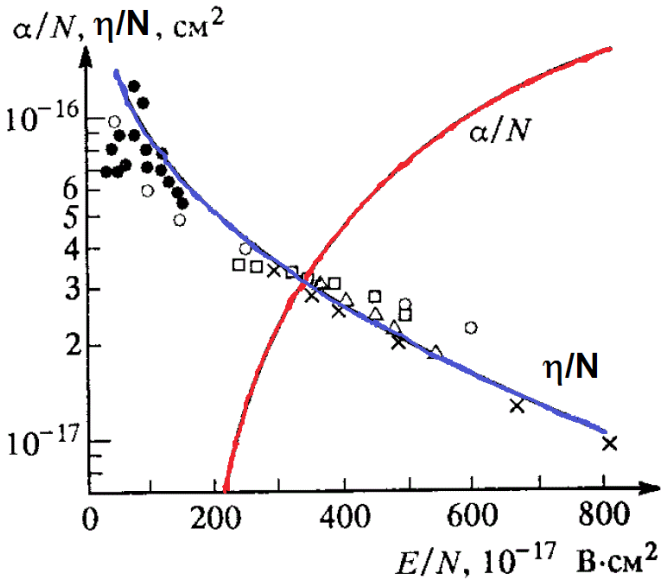

Figure 4. Dependence of the ionization factor and the sticking factor of electrons in SF_6 on E/N parameter (at room temperature).

The Problem of Increasing the Energy Characteristics of Non-Chain HF Lasers

Low progress in increasing the output characteristics of non-chain electric-discharge HF lasers was associated by most researchers with the difficulty of obtaining (in large discharge volumes) SSVD in SF -based gas mixtures. Indeed, the traditional approach to the ignition of SSVD in dense gases, which was formed by the 90s of the last century and became classical already [43-48,55], involves the creation of volumetric gas pre-ionization (UV, soft X-rays) with subsequent application to the discharge gap with a homogeneous electric field of high-voltage pulse [43]. In non-chain HF (DF) laser working mixtures, the ability to meet these requirements is limited by the strong electronegativity of SF_6. In addition, the simplest

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p-ISSN: 2347-2316

sources of UV radiation, in this case, are not effective due to the strong absorption of UV radiation in SF_6 [56]. **Figure 1** shows who dependence of the SF_6 photoabsorption cross-section on the radiation quantum energy [56]. It is easy to see that with an SF6 pressure of p=60 mm Hg the characteristic photon path length with an energy of about ~10 eV (the ionization potential of 'heavy' hydrocarbons) is only, which is clearly not enough to initiate SSVD in wide-aperture lasers. The creation of soft X-ray sources with an emitting area in excess of 100 cm^2 is an independent problem [55,57,58]. In addition, SF_6 is a highly electronegative gas, which causes large losses of free electrons in the process of sticking and their rapid disappearance in the discharge gap. It is enough to note that already at $E/p \approx 0.9$ (E/p) (E is the electric field strength in the discharge gap, p is the pressure of SF_6) and at a pressure of SF_6 p=60 mm Hg, the electron lifetime is $\tau=1/(\eta x v) \approx 1$ ns. (Here η is the sticking co-efficient, v is the electron drift velocity).

The presence of such a powerful channel for the death of free electrons places high demands on the power of the pre-ionization source and almost nullifies the possibility of fulfilling the condition of creating initial electrons with a concentration ne ~108 cm^3 in the entire discharge volume. Additional difficulties with increasing the aperture and volume of the active medium of non-chain lasers arise due to the need for special profiling of the electrodes to ensure the uniformity of the electric field in the DF since this leads to an increase in the size of the laser and the inductance of the discharge circuit with extremely limited duration of stable combustion of OCP in gas mixtures containing SF_6 [39,59].

Table 2. The cost of the formation of fluoride atoms when dissociated by electron impact of various fluoride-containing compounds.

F+RH(RD)=HF(DF)+R	Q, eV	Q, kcal/ mol	Chemical efficiency ηx,
H2	1.43	32.9	0.71
D2	1.4	21.7	0.7
HI	1.95	44.8	0.95
HBr	2.11	48.5	0.24
CH4	1.5	34.5	0.6
C2H6	1.7	39	0.62
C4 H10	1.66	38	0.56
CH3Cl	1.61	37	0.68
CHCl3	1.83	42	0.37
CH3 Br	1.56	35.9	0.67
CH 2Cl2	1.66	38	0.51
C2 H3 F3	1.46	33.5	0.67
C6H12	1.86	42.9	0.53

The parameters of a non-chain HF laser initiated by an SSVD with the energy of ~400 J were estimated on the basis of literature data [20]. The choice of the composition of the working environment was quite obvious SF_6 should be used as a donor of fluorine atoms. The reasons for this choice are, firstly, in the convenience of operation (SF_6 is a non-toxic and non-flammable gas) and, secondly, in the low cost of formation of fluorine atoms, $q_f \approx 4$ eV [50] **(Table 2)**. Indeed, as established by Jacobson TV [35]. The maximum efficiency of an electric-discharge non-chain HF laser is achieved when using SF_6 atoms as a donor of fluorine atoms. The choice of the second necessary component of the medium, namely, the donor of hydrogen atoms RH, is not so unambiguous since this component determines not only the chemical efficiency but also to a large extent the SSVD characteristics [3]. Since this issue will be discussed in detail in the next section, here we only note that most often H_2 or hydrocarbons (C_2H_6 and C_3H_8) are used as RH. Regarding the specific characteristics of non-chain HF (DF) lasers initiated by the SSVD, one important note needs to be made: in the literature, one can find report on the specific energy output W_{sp}~15 ÷ 20 J/l [40], however, such high parameters were achieved only in installations with very small active volume (V), which actually did not exceed a few cubic centimeters.

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p-ISSN: 2347-2316

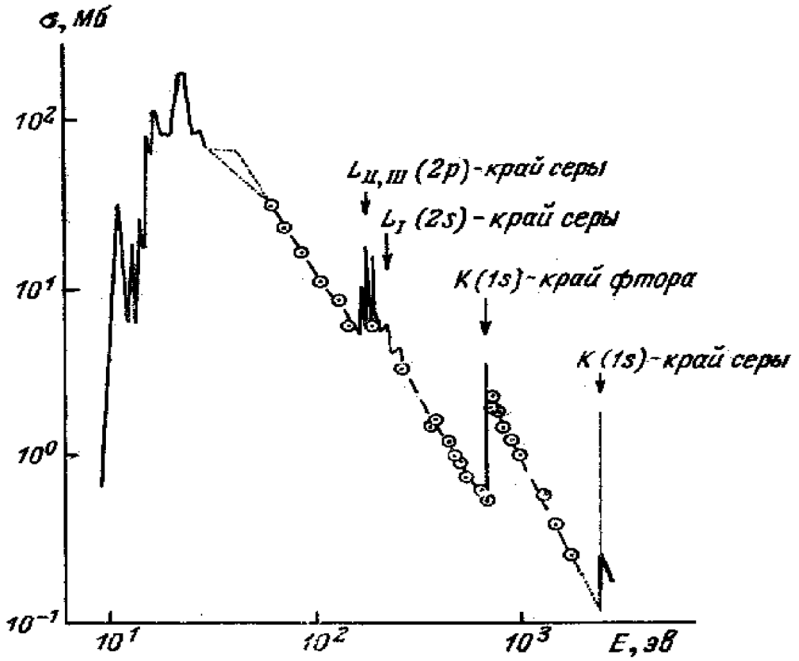

Figure 5. The dependence of the cross-section of the photoabsorption of SF_6 on the energy of a soft X-ray quantum [56].

Numerous attempts by researchers to achieve the same W_{sp} values on units with V~1 l were unsuccessful. Apparently, there is a common scaling problem for electric-discharge gas lasers. Therefore, when estimating the laser parameters, it is necessary to take into account not the limiting values of the specific characteristics of the laser, obtained in experiments on installations with a small active volume, but some average values that will be approximately preserved with increasing active volume. Analysis of the literature [32-42] shows that the average values of the specific energy output W_{sp} and the total electrical efficiency in SF_6:H_2 mixture lasers (hydrocarbons) are $W_{sp} \approx 5$ J/l and $\eta_T \approx 3\%$, respectively. The exceptions are quite compact systems [40,60] with a working volume not exceeding 60 cm^3. Since such systems do not scale, the estimates should focus on more realistic average results. As a result, we obtain the following estimated parameters: a discharge volume of V ≈ 80 l, an aperture of ~28 cm, and the energy input to the plasma of the SSVD $W_{sp} \approx 100$ J/l. The problem of creating CO_2 lasers with a similar aperture and the same level of energy input was solved in the late 80s of the last century [54,61]. However, attempts to increase the discharge volume and, accordingly, the radiation energy of non-chain HF (DF) CLs were far from being so successful as they were associated with low SSVD stability in the working mixtures of such lasers [62]. It is obvious that without solving the problem of the formation of a stable volume discharge in large discharge gaps, it is not possible to increase the energy of a non-chain laser. In this connection, let us pay attention to works in which the conditions for obtaining SSVD in working mixtures of HF (DF) CL differed from those usually considered.

Search for Methods for SSVD Formation in the Working Mixtures of HF (DF) Laser

An interesting result was obtained by Baranov VY, Wlodarczyk GA, Maluta DD [27,37,63], the authors of which found that the electrode system used by them makes it possible to ignite a uniform SSVD at a working mixture pressure of up to 100 mmHg without preionization. It is possible to note that it can be used in the workplace. without preionization. At the same time, at a pressure of 30 mmHg-50 mmHg, the effect of UV preionization on the generation level was weak [27], but pre ionization made it possible to stabilize the spread of laser energy from pulse to pulse (changes in the output energy from pulse to pulse during UV preionization were 1%, without preionization ~20%). Taking into account the fact that the creation of initial electrons in working mixtures of an HF (DF) laser is a complex problem, and the rejection of a preionization source can significantly simplify the laser design, let us consider the electrode system [27,63] .

In [27,63] the authors used a single crystal of germanium with a specific resistivity of ρ=25 Ohm.cm and a mesh brass cathode through which UV preionization was carried out. To ensure a uniform field in the RP, the anode had a Chang profile. The laser operated on an SF_6-C_3H_8 mixture (technical propane-butane mixture with a C_3H_8 content of ≈ 60%; $C_4H_{10} \approx 40\%$) and had high specific energy characteristics. Unfortunately, the authors Baranov VY, Maluta DD [27,63] did not carry out any analysis of the reasons that make it possible to obtain SSVD at sufficiently high energy inputs W_{in}~100 J/l without preionization. As the main reason for the effect, the authors considered the use of an anode of single-crystal germanium. A positive role in increasing the uniformity and stability of SSVD when using electrodes made of semiconducting materials was also reported in other publications [27].

e-ISSN: 2320-2459
p-ISSN: 2347-2316

However, an analysis of the literature on electric-discharge HF (DF) CLs allows one to doubt that the main reason that makes it possible to obtain SSVD without preionization is the use of a semiconducting anode with volume resistance. So, earlier it was possible to obtain SSVD without preionization in He: SF_6: C_3H_8=300-900:15:1 mixture at a total pressure of up to 600 mm Hg, reported in [29]. Installations [37,63] have close output energy characteristics Wout~400 mJ, power Pout~15 MW, however, with the same energy input W_{sp}~100 J/l, the specific characteristics [63] are about 1.5 times better. The setup execution scheme [37] is similar to the scheme of Baranov VY, Maluta DD [27,63], only in [37], the discharge was ignited between the Al cathode made along with the Rogowski profile and the mesh anode through which the discharge gap was illuminated by UV. It was also noted by Wlodarczyk GA [37] that sandblasting the cathode surface improves the structure of the discharge, while it is especially noted that the presence of C_3H_8 in the working mixture was a prerequisite for ignition of the SSVD, without which it was not possible to obtain an SSVD without a spark even in the presence of preionization. An improvement in the quality of the discharge when using a cathode with a rough surface in [37] was associated with an increase in the photoemissive capacity of the cathode (due to an increase in the effective area). The reasons for the influence of C_3H_8 on the improvement of the discharge were indicated: low, ~11 eV ionization potential, and a large photoionization cross-section. However, no specific mechanisms for this influence were proposed, so we will consider this issue in more detail.

It is known [64] that the addition of certain substances with a low ionization potential to the CO_2 working media of lasers makes it possible to increase the stability of SSVDs by reducing the electron energy due to a change in the electron energy distribution function and, as a consequence, suppressing the development of instabilities due to stepwise ionization of nitrogen. It could be assumed that the effect of C_3H_8 (and other hydrocarbons) on the stability of SSVD in working mixtures of an HF laser has a similar nature, i.e. a change in the electron energy distribution function, but no special studies of this issue were carried out until 1996. As regards the relatively high value of the photoionization cross-section of C_2H_6 (C_3H_8, etc.), it should be noted that with strong UV illumination and small distances from the illumination source to the active volume, this fact allows increasing the initial electron concentration. Apparently, this explains the improvement in the SSVD structure noted in [40] and the increase in the lasing energy of the non-chain HF laser when H_2 is replaced by C_2H_6. In general, it is quite obvious that the use of compounds with a low ionization potential and large photoionization cross-sections can reduce the requirements for the preionization source in small-sized installations, but it does not allow solving the problem of scaling the discharge volume in principle. In this regard, it is necessary to pay closer attention to the devices that were used to ignite the SSVD [27,37,63]. In these systems, the sources of UV illumination had a separate power supply, and when the authors Baranov VY, Wlodarczyk GA, Maluta DD [27,37,63] say that the discharge was obtained without preionization, this only means that the power supply circuit of the backlight was not turned on. However, no special experiments with shielding of the spark gaps of the UV illumination source were performed in these studies. At the same time, it is well known that when the switches of the main discharge generator are turned on, spark gaps of the UV illumination source in such schemes can breakthrough due to the induced potential difference on them. An indirect confirmation of the above is a relatively small scatter in the output energy. If the backlights of the main gap were completely absent, then in the case of a small cathode area [27,37,63] and low over voltages on the discharge gap, there should have been a significant spread in the delay time of the breakdown of the gap, and the spread in the values of the output energy would be much higher. However, in [27,37,63] there is generally no mention of the instability of the breakdown of the main gap under conditions when the source of preionization was not turned on.

Obviously, neither the spontaneous breakdown of some spark gaps in the preionization scheme nor the corona from the surface of the electrodes can provide the necessary, according to traditional concepts, initial electron concentration at the level of n~106 cm^3. It is noteworthy that the analysis of other works also points to the weak role of preionization in the formation of SSVD in the non-chain lasers. So, for example, approximately the same output energy and laser efficiency were obtained by Pummer H, Apollonov VV [41,64]. Pummer H [41], where a series of metal pins connected to a common bus via resistances (resistive isolation) served as the cathode of the discharge gap, there was no preionization in the traditional representation (coronation from pins, but this is a very weak source), and Burtsev VA [53] carried out by a high-current a creeping discharge, in the emission spectrum of which not only UV radiation was present, but also soft X-ray radiation. Moreover, Pummer H [41] the electrical efficiency (3.8%) turned out to be even higher than Burtsev VA [53] (2%). In this regard, it is necessary to especially note the results obtained by the authors Wensel RG, Arnold GP [65,66]. Arnold GP [66], the high energy characteristics of an HF laser were achieved using a setup in which a barrier discharge distributed over the cathode surface was used to obtain SSVD. Moreover, Apollonov VV [67], the voltage on the discharge gap having a rather high edge amplification of the electric field was applied with a short front, which, in principle, did not allow uniform filling of the discharge gap by electrons from the plasma created at the cathode using a barrier discharge. We also note that the radiation intensity of the barrier discharge in SF_6-based gas mixtures is insufficient to create an initial electron concentration in the discharge gap even at the level n~106 cm^3, which is necessary according to traditional ideas [52,55].

Looking ahead, it should be noted that when conducting prospecting studies of the possibility of increasing the energy of a non-chain electric discharge HF laser at the end of the 90s of the last century, our research team found that in mixtures of SF6 with hydrocarbons it is possible to obtain a volume discharge without preionization. Having realized the possible prospects for using this effect in lasers, our group began systematic studies of SSVD in gas mixtures based on SF_6.

e-ISSN: 2320-2459
p-ISSN: 2347-2316

Subsequently, the shape of the discharge, which is realized in mixtures based on SF_6, was called a Self-Initiating Volume Discharge (SIVD) [68,69] and the main task of the authors in subsequent years was to study the physics of this peculiar form of volume discharge.

P–P non-chain HF (DF) lasers with a high pulse repetition rate

The physical aspects of the implementation of the P-P mode in non-chain electric-discharge HF (DF) CLs are considered in detail [70], therefore, we present only the main results. **Figure 5a** shows a diagram of a laser setup in which the authors of Harris MR [71] demonstrated the possibility of operation of a non-chain closed-circuit HF (DF) electric discharge laser with a repetition rate of up to 3 kHz. To obtain the SSVD in the discharge gap of 5 mm × 5 mm × 150 mm, preionization with UV radiation from a number of spark sources located along the surface of the electrodes was used. The duration of the discharge pulse in this laser did not exceed 100 ns. The laser worked on mixtures of SF_6:H_2=10:1.

The results of the authors' studies [71] were the following observations:

- The characteristics of the laser (output energy, the maximum possible pulse repetition rate at which the energy in the pulse does not decrease) strongly depended on the power of the preionization source. It was also noted that with an increase in the spark gap (due to the burning of the pre ionizer electrodes), the maximum frequency of the laser operation increases
- The speed of pumping working gas through the discharge gap should provide more than 5-fold gas change in the working volume (in the article, the speed was stated that the flow rate provided an 8-fold gas change)
- The consumption of components in the mixture under the IP mode of laser operation was as follows: $2H_2$ molecules were consumed per SF molecule
- Sorbents based on zeolite 5A, which were used for the adsorption of HF (DF), also absorb significant amounts of SF
- The adsorber is the largest element of the laser, through which it is necessary to purge the gas flow, and it should ensure complete absorption of the products of plasma-chemical reactions

The main limiting factor limiting the laser operation time was the low survivability of the pre ionizer. The tungsten needle electrodes used for spark UV preionization quickly burnt out. A pre ionizer, whose electrodes are made of silicon, served for a much longer time; the discharge from these semiconductor electrodes burned in the quasi-volume form A corona discharge semiconductor preionizer provided the best laser parameters.

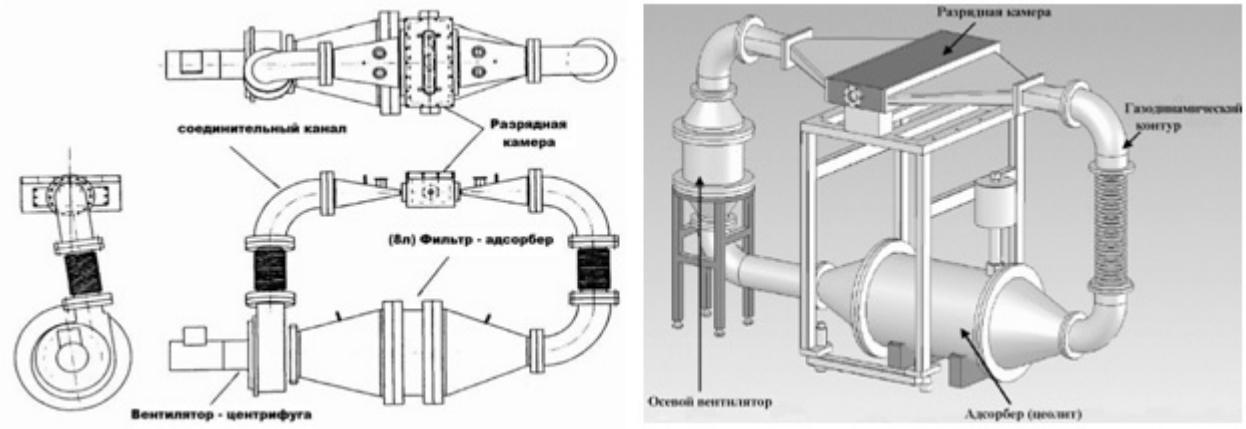

Figure 5. Scheme of a non-chain electrochemical HF (DF) closed-loop laser: a) according to Harris MR [71]; b) according to Zhou S [72].

Much later, Zhou S [72], a similar scheme was reproduced (**Figure 5b**), but they were able to significantly improve the stability of the I-P discharge when ethane was added to SF_6. Hydrocarbons not only increased the stability of the discharge, leveling the strong dependence of the laser characteristics on pre ionization but also allowed to increase the laser operating time (i.e., the number of shots without reducing energy in a closed-loop mode). True, it should be noted that the pulse repetition rate in Zhou S [72] was almost an order of magnitude lower than in Harris MR [71], since in Harris MR [71] there was a more powerful centrifugal fan, and Chinese researchers used a block of axial fans.

Many researchers working with non-chain P-P HF (DF) CLs (this is especially evident in low-aperture installations operating with a high pulse repetition rate) faced a dilemma: on the one hand, to stabilize laser characteristics at high frequencies, it is necessary to increase the pre ionizer power, and on the other hand, an increase in the power of the spark pre ionizer leads to rapid degradation of the working mixture and the burning of the pre ionizer electrodes. In this connection, the results of Serafetinidest AA, Hatch CBA [73,74] are noteworthy, in which the authors used a quasi-volume discharge for preionization, which develops from semiconductor electrodes made of SiC. It should also be noted that a detailed analysis of the design of the discharge chamber and the location of the UV pre ionizer Harris MR [71] allows us to note that the

e-ISSN: 2320-2459
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improvement in laser characteristics always correlated with an increase in cathode illumination. When the length of the arc channel from the pre ionizers was increased or a quasi-volume discharge was used, such geometry of the pre ionizer source provided a significantly larger flux of UV radiation to the cathode. In general, we have to admit that many authors point to the degradation of the pre ionizer electrodes as the main factor that reduces the working time of a closed-loop laser. Therefore, the solution to the problem of increasing the life of the pre ionizer in a closed-loop laser is very important. Obviously, the possibility of completely abandoning the preionization system could greatly simplify the design and increase the life of the non-chain HF (DF) CLs initiated by the SSVD.

When operating a non-chain HF (DF) CL in the P-P mode with a high pulse repetition rate, cooling of the working mixture is also an important factor. The release of additional thermal energy from pre ionization devices and from a gas stream that slows down when it encounters obstacles in its path is a negative factor. Many authors have shown that increasing the gas temperature in the working zone leads to a decrease in the output energy and laser efficiency, and, conversely, cooling the mixture leads to an increase in the output energy and the laser spectrum becomes richer [3,38].

It should be noted that, in contrast to pulsed lasers, when implementing the P-P mode, the stability of a volume discharge has much higher requirements. A certain margin of stability is required, since in the discharge gap under conditions of powerful gas flow and periodic disturbances from previous discharges, in homogeneities in the density and temperature of the gas can occur. The best characteristics for solving several problems of a P-P laser at once were shown by the approach proposed by Velicanov SD, Andramanov AV [75,76]. In these works, it was shown that the use of a blade system of electrodes with inductive stabilization allows for high gas flow rates through RP and high discharge stability. **Figure 6** shows a compact HF (DF) closed-loop laser circuit [77]. Using the approach described above, lasers were created with a pulse repetition rate of more than 3 kHz, as well as a laser with an average power of more than 400 W [78].

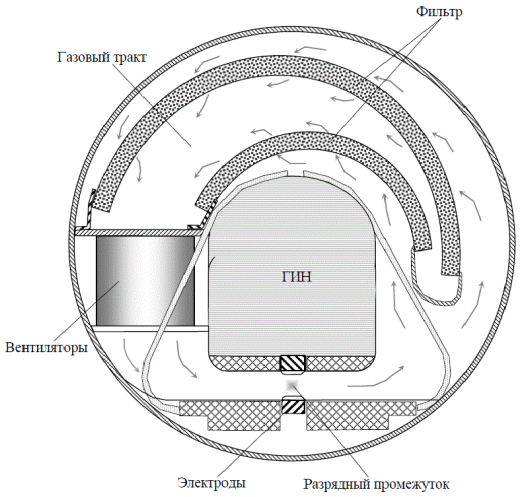

Figure 6. Schematic of a compact P-P non-chain HF (DF) closed-loop laser [78](PVG -Pulsed Voltage Generator).

The high stability of the discharge is also ensured by the use of semiconductor electrodes [27,74] or anisotropic-resistive cathodes, which provide high stabilization of the discharge [79,80]. However, high heat losses in the material of the anisotropic-resistive cathode during long-term operation can lead to damage to the cathode, therefore, it is desirable to study in more detail the possibilities to increase the stability and uniformity of the SSVD through the use of semiconductor and anisotropic-resistive cathodes.

To conclude this section, we note that success in the implementation of high-energy CL implies the following task-the conversion of the radiation frequency of the HF (DF) laser to obtain lasing in a wider spectral range. The problem of finding effective frequency converters of high energy CLs has been dealt with a long time [3]. Andreev YM [81], the second harmonic of DF CL radiation was generated in a ZnGeP crystal; to obtain wavelengths with λ<2.5 μm, there is also the possibility of generating overtone lasing [3]. A parametric light generator based on a CdSe crystal pumped by an HF laser was created by Weis TA [82]. Recently, there has been great interest in the search for powerful sources of radiation from the terahertz region of the spectrum [83]. To obtain lasing in this long-wavelengthh region of the IR spectrum, lasers based on HF rotational transitions were studied [84-86]. However, the most urgent for those application areas in which the HF (DF) CL was used is the search for the possibility of creating laser systems for generating powerful, tunable in the spectral range of 4 μm-6 μm coherent radiation [87-91]. Thus, to increase the energy characteristics of non-chain HF (DF) CLs initiated by SSVD, it is necessary to search for new principles for the formation of a scalable volume discharge. Another important problem is the search for methods for producing high energy, pulsed, and medium power in the spectral region with >4.1 μm, where the generation efficiency of electric-discharge non-chain HF (DF) CLs is low.

e-ISSN: 2320-2459
p-ISSN: 2347-2316

CONCLUSION

Therefore, we focused on solving two problems:

• The formation of a scalable SSVD in strongly electronegative gases and the creation of wide-aperture high-energy non-chain HF (DF) CLs

• Expanding the generation spectrum of a non-chain HF(DF) CL and creating lasers with high energy per pulse, pulsed and average power in the spectral range >4.1 μm

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