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Optimization of parameters of a compact soft X-ray source for operation in the wavelength range 2−5 nm

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This paper presents a compact source of soft X-ray radiation for operation in the wavelength range of 2−5 nm with a pulse repetition frequency of more than 500 Hz. The source parameters were optimized to reduce the intensity of ablation of the discharge volume wall and obtain the maximum intensity of the spectral lines of multicharged ions C V — 4 nm and Ar IX — 4.87 nm. The obtained results can be used in the development of a microscope for the tasks of cell transmission microscopy with nanometer resolution.

Keywords: High-voltage pulse generator, Capillary plasma, „Water-window“microscopy.

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Introduction

Microscopy in the field of water transparency („water window“), i.e. in the wavelength range 2.3−4.4 nm, is a unique method for studying living and prepared cell cultures and various nano- and bio-objects. This method allows obtaining important information about the internal structure of cells (organelles) with a high lateral resolution reaching ~ 10 nm [1]. The area 4.5−5 nm is the carbon transparency region, the so-called „carbon window“ [2], in this range nitrogen- and oxygen-containing compounds have strong absorption. Using these two wavelength ranges, it becomes possible to perform detailed diagnostics (tomography) of the internal structure of biological objects in the transmission mode by obtaining contrast images and their subsequent stitching.

It is possible to receive radiation in the specified wavelength range on a synchrotron, however the work on installations of this level is not available to a wide range of researchers. In addition, the use of a synchrotron for solving routine biomedical tasks is obviously unreasonable. Therefore, compact desktop sources of soft X-ray radiation (SXR) are currently being actively developed for subsequent implementation on their basis of a microscope using diffraction and multilayer X-ray optics.

In this paper, the study and optimization of the parameters of a compact radiation source based on a nanosecond capillary discharge is carried out for its operation in the wavelength ranges 2.3−4.4 and 4.5−5 nm.

1. Experimental setup

The design of the discharge cavity of a soft X-ray source based on a capillary discharge is shown in Fig. 1.

The discharge was excited in the volume of a borosilicate capillary, the length of which was 20 mm, inner and outer diameters 1.5 and 8 mm, respectively. When choosing the capillary parameters, we were based on our own preliminary experiments and the results of the authors of [3]. It is known that the use of capillaries with a large internal diameter leads to a sharp drop in current density and, as a consequence, a significant decrease in the intensity of ion lines in the area of the „water window“. The use of capillaries with a diameter smaller than 1.5mm — leads to strong wall ablation. The diameters of the holes in the cathode and anode were 0.8 and 1.6 mm, respectively, the gas continuously flowed from the cathode side through a needle flow. The gas pressure at the capillary inlet was $\sim 1-4$ Tor, at the outlet $\sim 10^{-3}$ Torr, depending on the type of gas. Pumping of the entire system was carried out by a turbomolecular pump. Thus, a gas pressure gradient was formed in the capillary (Fig. 1, *a*), which provided conditions for the formation of cathode plasma — the effect of „hollow cathode“ [4], stability of the breakdown process itself and reduction of absorption radiation in the residual gas.

As an insulator between the cathode and the anode, we used a polyamide sleeve and vacuum rubber, the structure was compressed with plastic screws, which ensured the tightness of the system. We have abandoned the use of a liquid dielectric as an energy storage and insulator, as this

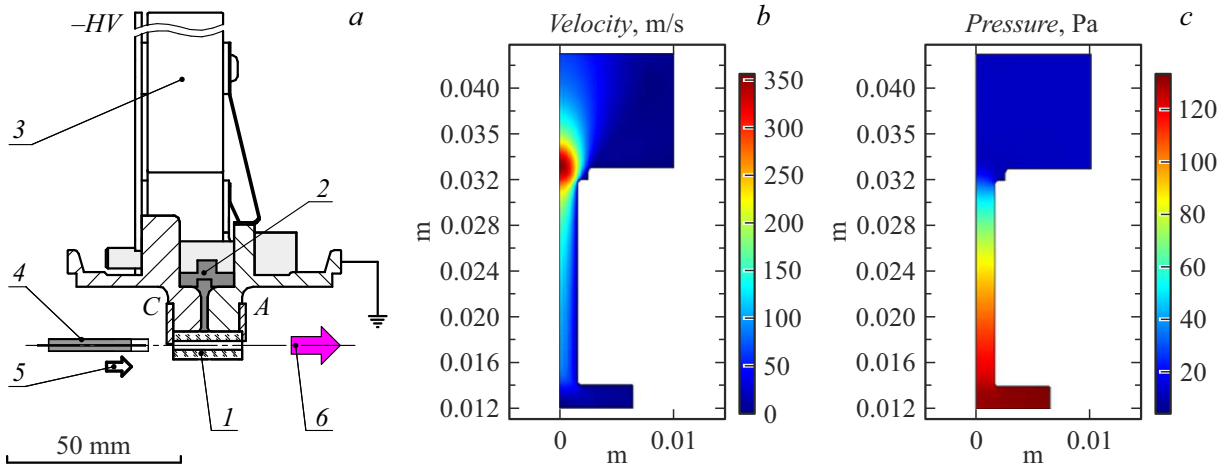

Figure 1. Capillary assembly design (a): *1* — borosilicate capillary, *2* —insulator, *3* — ceramic capacitor, *4* — plasma injector, *5* — gas flow direction, *6* — SXR pulse; C, A — cathode and anode, respectively; calculated distributions of velocity (b) and gas pressure in the capillary (c).

leads to degradation of the outer wall of the capillary and the ingress of water into the vacuum system, which was shown in the study [3]. Ceramic capacitors with a total capacity of 8 nF were installed as storage devices on the capillary junction.

It is known that the main problem of gas discharge sources is „contamination“ of the spectrum by spectral lines of the elements from which the capillary is made. Another problem — contamination of the optical system with ablation products — is partially solved, for example, by installing a thin-film filter for SXR in a vacuum system in the path of a beam or a particle capture system.

Nevertheless, there are several ways to minimize the intensity of the wall ablation process and, accordingly, the contamination of the spectrum: 1) by choosing the capillary material such that the elements from which the capillary is made do not have spectral lines in the desired radiation range (or are not excited in the plasma), 2) by a high rate of energy input into the plasma, which is provided by a low inductance of the discharge circuit and a corresponding increase in the rate of current growth. On the other hand, in order to reduce the thermal load on the capillary and the erosion of the cathode, we have developed a high-voltage generator that provides fast charging of the capacitor bank during ~ 100 ns, which is an order of magnitude less than that of gas-discharge compact sources known from the literature SXR [5].

In the study [6], a ceramic capillary Al_2O_3 was used and it was shown that even with an energy input of 1 J into the plasma in the region of 4–6 nm there are lines of ions Al VIII, IX, which are difficult to filter with metal filters. Therefore, a borosilicate glass capillary was used in this study, since this material is heat-resistant, and silicon ions in this range are difficult to excite. It is also significant that the quality of the inner surface of the glass is much higher than that of ceramics Al_2O_3.

1.1. High-voltage generator of the SXR source

To increase the rate of energy input into the plasma, a pulsed high-voltage generator based on a modern TPI-10k/50 thyratron was developed. A feature of the generator is the possibility of obtaining nanosecond voltage pulses with an amplitude up to 35 kV, therefore, the energy invested in the plasma was ~ 5 J. The electrical diagram is shown in Fig. 2.

The main idea of the high-voltage generator circuit is to charge the accumulator of the capillary discharge current generator ($C2$) in one oscillatory cycle when using an intermediate capacitive accumulator with the same energy, but at a lower initial voltage, approximately equal to twice the rectified voltage of the three-phase network. The central junction of this charge system is a pulse transformer ($T1$) made on a ferrite core made of N87 material, in the secondary and primary circuits of which $C1$ and $C2$ capacities are used.

The circuit is powered from a three-phase AC 380 V/50 Hz through a protective automatic. A three-phase bridge rectifier $VD1$ provides a charge of the filter capacity $C0$ on electrolytic capacitors to a voltage of $U_d = \sqrt{2}\,U_l \approx 530$ V (U_d — capacitor charging voltage, U_l — mains supply voltage). To limit the current surge through the rectifier at the moment of switching on, damping resistors $R1-R3$ are used, then shunted by the contactor.

The charge of the intermediate storage $C1$ occurs according to the principle of resonant recharge of the filter capacity $C0$ to a much smaller capacity $C1$ when the control signal ICP transistor $VT1$ is opened by an external command for a period of time $T/2 \sim 0.5\pi((L0 + L1)C1/(1 + C1/C0))$, where $L0$ — inductance of the charging choke and $L1$ — inductance of the primary winding of the transformer $T1$. The charge current passes through the circuit: choke $L0$, intermediate storage

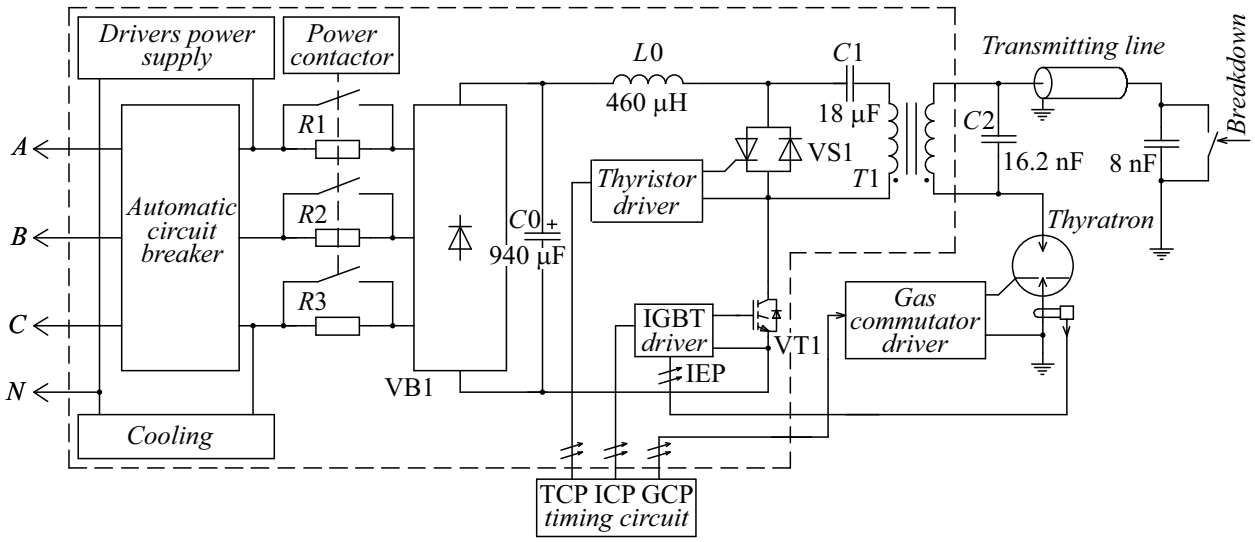

Figure 2. Electrical diagram of the high-voltage generator of the SXR source.

$C1$, primary transformer winding $T1$, $VT1$. After the time $T/2$ expires, the first half-cycle of the charging current ends, the capacity $C1$ will be charged to the voltage ~ 1000 V.

Then, at time $T/2$, the thyristor $VS1$ is started by an external signal TCP. The time of energy transfer from the capacity $C1$ to the storage capacity of the generator $C2$ is determined by the coupling coefficient of the windings of the pulse transformer $T1$, which is ≈ 0.995. This leads to a good energy transfer in this junction. The charging time of the capacity $C2$ to the maximum voltage ~ 32 kV was $\sim 5.7\,\mu$s.

Next, the thyratron should be turned on with a delay of T_z relative to the start of the thyristor, taking into account the delay of signals in the channel drivers. After the discharge of the storage capacity $C2$ to a complex (active-reactive) load (discharge interval), a time interval is maintained during which the current in the thyristor circuit is damped and the filter capacity $C0$ is recharged by the rectifier to the nominal operating voltage U_d (the time interval is not less than the discharge interval $C0$, i.e. $T/2$). A sequence of three driver starting signals is formed in the time-consuming module, which is started by an external signal source with the required frequency.

In order to avoid accidents caused by failures of charging processes, a thyratron trigger sensor has been introduced, allowing the operation of the first stage when a current exceeding the setpoint is detected in the gas switch circuit (IEP signal).

2. Experimental results

In our installation, a mixture of argon and helium (2 : 1) and carbon dioxide were taken as working gases, since argon ions emit single spectral lines in the region of the „carbon window“, and carbon ions in the „water window“. In the study [3] it was shown that the addition of helium to argon increases the life of the ceramic capillary, so we used this data for our conditions. No data was found in the literature about discharge under similar conditions for CO_2, so in our research we took a pure CO_2.

To diagnose radiation in our experiments, a silicon photodiode FDUK-1 (CJSC „Technoexan“) with an active region size of 1 mm^2 was used, which made it possible to register the time profile of the radiation pulse with a resolution no worse than ~ 1 ns, dark current of the photodiode did not exceed 100 nA. The sensitivity of the photodiode in the wavelength range 2–10 nm was 0.26 A/W. Two free-hanging thin-film filters were installed in front of the photodiode — the first Al, the second was a Ti–C composition; the filter thicknesses were ~ 200 nm [7]. To deflect the electron beam, a permanent magnet was installed behind the anode of the capillary junction. The filters attenuated the visible radiation by 10^6 times, and their combination made it possible to select the desired radiation range from 2.5 to 5 nm.

The registration of capillary plasma spectra was carried out using a GIS grazing incidence spectrometer with a diffraction grating 1200 mm^{-1} [8], a GreatEyes GE 1024 1024 BI UV1 CCD camera was used as a detector, the spectral resolution of the spectrometer $\lambda/\Delta\lambda$ was ~ 200.

The current pulse was recorded by a Rogowski coil installed above the capillary at the insulator separating the vacuum flanges, a high-voltage divider was used to measure the voltage pulse. All signals were recorded by a four-channel Tektronix DPO-7104C oscilloscope at a bandwidth of 1 GHz and averaged over 20 shots. Note that all signals were transmitted via radio frequency cables of the Sukoform series (manufacturer Huber+Suhner), which had an external metal screen, which made it possible to avoid interference from a high-voltage generator. On the other hand, the generator itself was placed in a specially

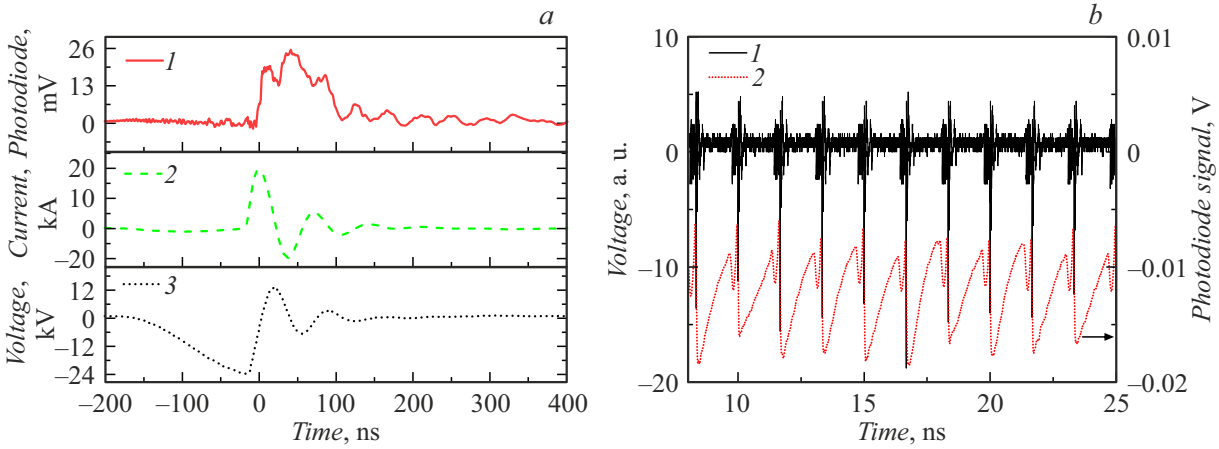

Figure 3. *a* — waveforms of the radiation pulse through two filters (Ti–C, Al) (*1*), current (*2*) and voltage (*3*) at a breakdown in Ar : He (the pressure at the cathode inlet was 4.5 Torr); *b* — oscillogram of the source operation at a frequency of 600 Hz: *1* — voltage pulse, *2* — the signal from the photodiode through metal filters.

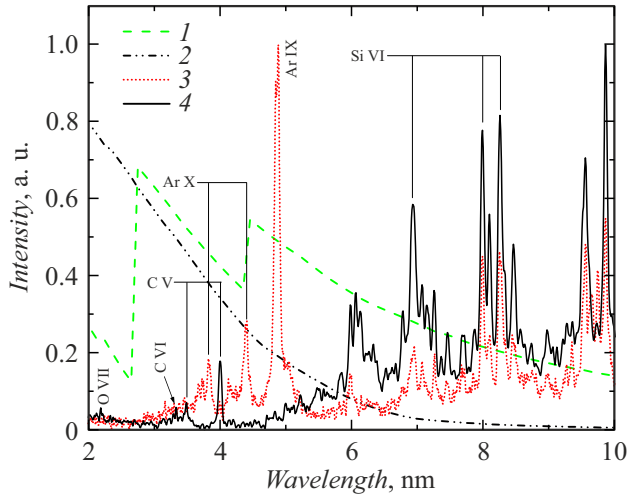

Figure 4. Transmission spectra of thin-film metal filters: *1* — Ti–C, *2* — Al, radiation spectra of capillary plasma at breakdown b: *3* — Ar : He (pressure on at the cathode inlet 1.5 Torr), *4* — CO_2 gas (pressure at the cathode inlet 0.7 Torr).

designed and manufactured metal housing. As evidenced by the experimental data presented below, these factors made it possible to eliminate high-frequency interference, all signals in this work are presented without any processing, the level of interference from the signal from the photodiode did not exceed 1 mV. Fig. 3 shows characteristic oscillograms of voltage, current and pulse of radiation through two filters (Ti–C and Al) at a breakdown in Ar : He, as well as an oscillogram of the source operation at a frequency of 600 Hz.

As can be seen from Fig. 3, the current amplitude reaches 19 kA with the duration of the leading edge ~ 10 ns, the rate of rise of the front was $\sim 2 \cdot 10^{12}$ A/s. With such short current pulses, effective ionization of the working gas occurs, and heating of the capillary walls becomes minimal. At the same time, the magnitude of the current amplitude makes it possible to reach a plasma temperature sufficient to excite transitions whose spectral lines lie in the region of 2–5 nm. The appearance of a pulse of radiation from a photodiode through two filters was observed after the maximum of the current pulse, and its duration was about 100 ns. Thus, the generation of SXR occurs at the stage of recombination of capillary plasma. The oscillograms had similar time forms at the breakdown in CO_2 gas.

For a detailed analysis of radiation and capillary wall ablation processes, we investigated the dependence of the intensity of the plasma emission spectrum and its composition on the pressure at the entrance to the capillary for both gases. Fig. 4 shows the maximum normalized emission spectra of capillary plasma when discharged in Ar : He and CO_2 gas for pressures at the cathode inlet at which ions of the highest multiplicity appeared in the plasma, the most intense lines were identified using the NIST database [9]. Fig. 4 also shows the transmission spectra of thin-film metal filters used in measurements using a photodiode.

As can be seen from Fig. 4, the spectrum contains highly excited ions in the wavelength range 2–10 nm. In the case of the argon–helium mixture, the most intense line is the Ne-like Ar IX $2p6 - 2p5.3s$ at a wavelength of 4.87 nm, lying in the region of the „carbon window“, lines of Ar X $2p5 - 2p4.(3P).3d$, $2p5 - 2p4.(3P).3s$ ions also appeared in the spectrum, lying in the area of the „water window“. These lines were observed in the entire range of operating gas pressures at the cathode inlet from 0.7 to 4.5 Torr.

When discharged in CO_2, a sufficiently intense line of He-like ion C V $1s2 - 1s2p$ (with wavelength 4.02 nm) was observed lying in the region of the „water window“, at a given gas pressure, an H-like ion was also observed C VI $1s - 2p$ (wavelength 3.37 nm) and He-like oxygen ion O VII $1s^2 - 1s2p$ (wavelength 2.16 nm) lying outside the

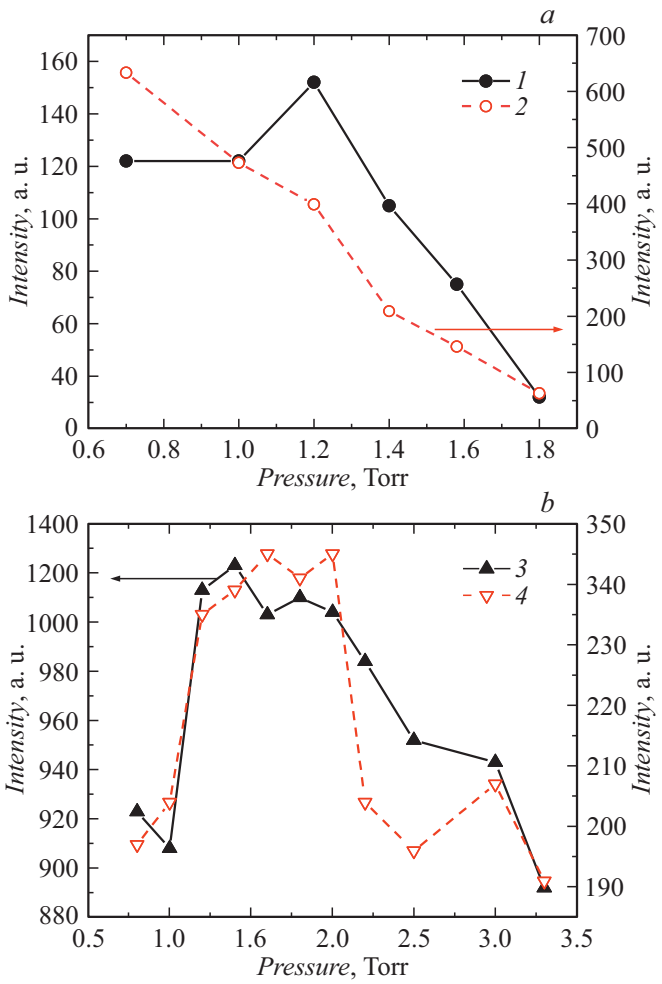

Figure 5. Dependences of the luminescence intensity of the lines of multicharged ions on the working pressure at breakdown in CO_2 gas (*a*): *1* — C V 4.02 nm, *2* — Si VI 6.92 nm; at breakdown in the mixture Ar : He (*b*): *3* — Ar IX, *4* — Si VI.

area of the „water window“. Note that at gas pressures greater than 1 Torr, the ion lines C VI, O VII were not observed in the spectrum, and the intensity of the line C V 4.02 nm decreased.

Silicon ion lines Si VI transitions $2s^2 2p^5$–$2s^2 2p^4(^1D)4d$, $2s^2 2p^5 - 2s^2 2p^4(^3P)3d$ were identified in spectra, lying in the region of 7 – 9 nm, the presence of which indicates ablation of the capillary wall. In the case of the argon–helium mixture, their intensity over the entire pressure range was always lower than the intensity of the working gas lines (Fig. 5), in the case of CO_2 gas, their intensity decreased significantly at a pressure of more than 1.2 Torr. Fig. 5 shows the integral dependences of the intensity (area under the contour of the spectral line) of the glow of ions Ar IX, C V, Si VI on the operating pressure.

As can be seen from Fig. 5, with increasing pressure, the intensity of all components of the emission spectrum decreases, which is explained by a decrease in the degree of plasma ionization and, possibly, the absorption of radiation in the residual gas with increasing pressure. At the same time, at a certain pressure, the radiation maxima of the Ar IX lines at 1.5 Tor and C V at 1.2 Torr are observed. The intensity of the silicon ion Si VI in the case of a mixture of Ar–He drops sharply with an increase in pressure of more than 2 Torr, in the case of CO_2, its intensity drops almost linearly with an increase in pressure. Such a difference in the behavior of the luminescence of silicon ions from pressure, apparently, is associated with the role of helium in the gas mixture, the presence of which increases the capillary resource by reducing the intensity of wall ablation. Thus, by regulating the gas pressure during its breakdown by short voltage pulses, it becomes possible to minimize the ablation of the wall while maintaining a sufficiently high intensity of the glow of multicharged ions.

Separately, we note the question of the capillary resource. In the present work, after $\sim 50\,000$ pulses in the frequency-burst mode, there was no decrease in the intensity of spectral lines in the range of 2 – 10 nm, their intensity remained constant. There was also no significant increase in the internal diameter of the capillary itself. All this can be explained by the use of short nanosecond voltage pulses to initiate plasma, which obviously reduces the erosion of the capillary wall. However, further studies of the lifetime (resource) and the effect of the capillary material on plasma parameters require additional experiments.

Conclusion

In this paper, the results of experiments conducted on a compact gas-discharge SXR source were presented. It is shown that fast charging of the storage tank on the capillary leads to a significant increase in the growth rate of the discharge current ($\sim 2 \cdot 10^{12}$ A/s), which makes it possible to achieve a high degree of ionization even with the breakdown of molecular gases. The low inductance of the discharge circuit makes it possible to achieve a high current amplitude — in this work, more than 18 kA. These factors lead to an efficient energy transfer of electrical energy into the plasma, and short pulses of voltage and current reduce the intensity of the capillary wall ablation process.

In this paper, the optimization of operating pressures for a mixture of Ar : He and CO_2 gas has been carried out. The possibility of achieving radiation generation modes without significant evaporation of the capillary material wall is shown. In the spectra of capillary plasma, lines of multicharged ions lying in the region of the „carbon window“ and „water window“are registered: Ar IX — 4.78 nm, C V — 4.02 nm. The lines can be highlighted using free-hanging thin-film filters.

The source allows generating SXR pulses with a frequency of up to 600 Hz, higher frequencies will be achieved when the cooling system of the discharge junction is integrated. The obtained results can be further used in the creation of a microscope for the study of cell cultures and other nano-biological objects in the lumen mode.

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Conflict of interest

The authors declare that they have no conflict of interest.

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