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Investigation of the emission properties of gas-jet targets in the „water transparency window“ of 2.3−4.4 nm under pulsed laser excitation

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The article considers the results of studying the emission radiation of Ar, Kr, CO_2, CHF_3, CF_4 and N_2 gas jets excited by pulsed laser radiation. The shape of the spectral dependence in the spectral range 2.3−4.4 nm and the emission intensity of a number of lines in absolute units were studied. To excite the targets, a Nd:YAG laser, $\lambda = 1064$ nm, $\tau = 5.2$ ns, $E_{imp} = 0.8$ J was used. A supersonic conical nozzle with $d_{cr} = 450\,\mu$m, $2\alpha = 11^\circ$, $L = 5$ mm was used to form a pulsed gas jet. The pressure at the nozzle inlet was 25 bar, which corresponds to the developed condensation of molecular gases in gas jets. The spectra were deciphered and the ions emitting under the given experimental conditions were determined.

Keywords: gas jets, shock waves, extreme ultraviolet radiation, emission spectra, laser plasma, X-ray spectrometer monochromator.

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Introduction

Currently, a number of applications related to soft X-ray radiation (SXR) are actively developing, in particular, X-ray microscopy [1,2]. So, in the department of multilayer X-ray optics of the IFM RAS, an X-ray microscope is being created to conduct research in the „window of water transparency“ 2.3−4.4 nm [3]. For the successful development of these applications, there is a need for high-intensity laboratory sources of soft X-ray radiation. Currently, the most convenient in terms of the set of properties are laser-plasma radiation sources (LPRS) [4].

The physics of the interaction of laser radiation and target matter has previously been studied in a large number of papers [5–12], including of a fundamental nature [13,14]. The main attention in these papers was paid to the laser systems used, with slightly less attention to the targets used. Various types of LPRS targets were studied: solid-state, liquid-jet, gas-jet. Such sources have various advantages and disadvantages, but the most developed at the moment are gas-jet target formation systems.

It is known that for effective radiation in the „water transparency window “ 2.3−4.4 nm in the LPRS, it is necessary to form a dense high-temperature plasma, which requires the following conditions: high-power laser radiation, strong absorption of radiation by the gas target substance, high density of emitting ions. Various laser systems have been studied, differing in pulse energy and duration, radiation wavelength, etc. [1,15–18]. Lasers with nanosecond pulse duration have received the greatest practical application.

For gas-jet targets, high absorption of laser radiation and a large concentration of emission centers can be achieved due to the high density of matter in the laser plasma formation zone. In turn, obtaining a high density can be achieved in two ways — using gases at elevated pressures, or using chemical compounds in which the emitting chemical elements are „packed„more tightly. Here it is worth noting an important aspect of the use of such target formation systems — when easily condensing gases flow out of profiled nozzles, their partial condensation with the formation of clusters is observed, which significantly changes the integral density of the gas target and laser absorption. It can be stated that such LPRS targets are insufficiently studied with an undoubted interest in them.

The objectives of the research were to study the emission spectra of gas jet targets in the „water transparency window“ 2.3−4.4 nm and to measure the absolute values of radiation in this spectral range.

1. Research plant

To obtain the materials, an installation was used, the scheme of which is shown in Fig. 1.

The plant operated as follows. The gas under study enters the high-speed valve 8 and then into the conical supersonic nozzle *7*. The gas jet is pumped out by cryocondensation and cryoadsorption pumps. Laser radiation *1* hits the dividing plate *3*, from where a small part of the radiation is fed to the first radiation power detector *2*. The main part of the radiation, passing through the prism *4* and the

optical input *5*, falls on the lens *6*. In the focus of a short-focus lens, laser radiation causes a breakdown and formation of plasma in a gas jet, the polychromatic SXR radiation of which, passing through an electropneumatic vacuum shutter *9* and a free-hanging X-ray filter *10*, enters the lattice spectrometer-monochromator RSM-500. Further, monochromatic SXR radiation is detected by a pulse detector. Pumping of the RSM-500 is carried out by a turbomolecular pump *12*. When carrying out the study, we used spherical mirrors and lattices with a gold coating. These mirrors and grilles were supplied complete with the RSM-500 device. The radius of curvature of the mirror is 4 m, lattice — 3 m, number of strokes 600 lines/mm. The spectral resolution of the device measured by the width of the characteristic lines FeL_{α}, as well as by the half-width of the zero order was 0.04 nm. For the gratings and mirrors used, the studied wavelength range was 1.7–12 nm.

At the entrance to the RSM-500 spectrometer, film free-hanging filters based on Ti/Be (total thickness 150 nm) and Al (thickness 150 nm) [19] were installed. These filters transmit radiation in the spectral range „of the water transparency window“ and at the same time effectively absorb the long-wave noise component of the signal. Also, free-hanging filters simultaneously protect against particles of various nature formed during the operation of the SXR radiation source, which effectively reduces background noise. The research setup is described in more detail in [20].

To study the absolute radiation intensities, a spectrometer based on a multilayer X-ray mirror (MXRM), calibrated in absolute units, was used. The spectrometer is a $\varphi-2\varphi$ goniometer, in which MXRM is used as a dispersing element. The operation of the device is carried out as follows: The SXR radiation of the laser plasma passes through the input free-hanging film filter and enters the MXRM. In accordance with the Wolf–Bragg condition, radiation with a certain wavelength is reflected from the mirror. The reflected radiation passes through the second free-hanging film filter at the detector and is recorded. Spectrum scanning is performed by rotation (by an angle of φ) The MXRM is relative to the incident beam, while the detector rotates relative to the incident beam by a doubled angle (2φ). The rotation of the mirror and detector is carried out using a stepper motor, the condition $\varphi - 2\varphi$ is provided by a gear train.

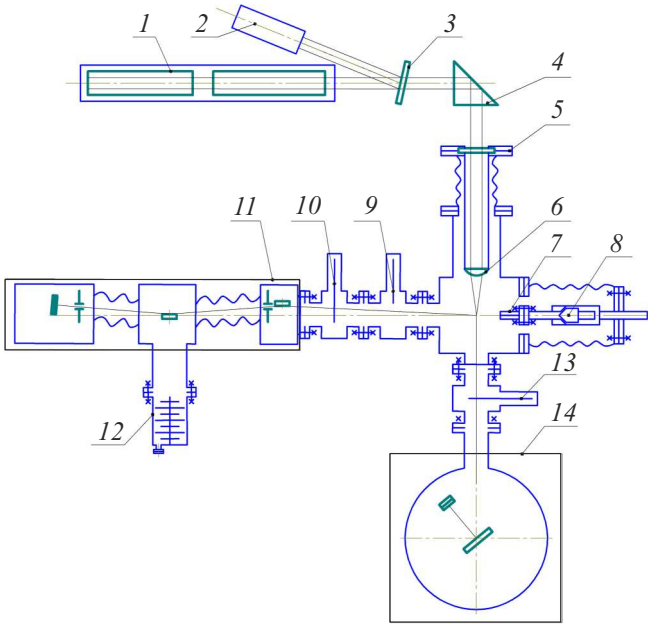

Figure 1. Scheme of the research facility: *1* — laser, *2* — laser power sensor, *3* — dividing plate, *4* — prism, *5* — optical input, *6* — lens, *7* — nozzle, *8* — high-speed valve, *9* — vacuum gate, *10* — film free-hanging filter, *11* — spectrometer-monochromator RSM-500, *12* — turbomolecular pump, *13* — film free-hanging filter, *14* — spectrometer for the study of absolute radiation intensities.

Free-hanging Ti/Be film filters with layer thicknesses of 3,nm/2 nm, the number of periods of 30 were installed in the mirror spectrometer, which are a spectral filter that effectively transmit SXR radiation for wavelengths of 10–3.1,nm. MXRM based on Cr/Sc were used (structure period 3.65 nm, thickness Cr — 1.3 nm, number of structure periods — 140). A detailed description of the MXRM characteristics based on the Cr/Sc structure is given in [21]. When using such a combination of MXRM and film filters, it is possible to study the wavelength range 3.1–7 nm. The spectral resolution of the device is determined mainly by the half-width reflection curve of the MXRM. Experimentally, the resolution was determined by the half-width of narrow free-standing emission lines of carbon ions and was about 0.06 nm. The spectrometer and the principles of its operation are described in more detail in [22].

To excite the gas jet, NL300 Series Nd:YAG Laser was used with the following parameters: wavelength 1064 nm, laser pulse energy 0.8 J, pulse duration 5.2 ns, frequency up to 10 Hz. Laser radiation is focused on a gas target using a lens with a focal length of 45 mm. The calculated focal spot diameter is 66 μm. When laser radiation is focused on a gas beam, an optical breakdown occurs, accompanied by the formation of a plasma cloud of repeatedly ionized ions.

For the system of forming a pulsed gas jet, a pulse valve was used, at the inlet of which a large gas pressure was created (up to 25 bar), and a conical nozzle was fixed at the outlet. As a valve for gas, we used a Bosch nozzle 0 280 158 017. A hole with a diameter of 1 mm was cut in the end of the nozzle by the electroerosion method. A clip was soldered to the surface of the nozzle, to which the nozzle was attached. A conical supersonic nozzle with a critical section of 450 μm, a length of 5 mm, a solution angle of 11° was used. In conducted experiments, the duration of the gas pulse was 0.5 ms.

Gas jets formed in the process of outflow from conical nozzles into vacuum generally have a complex spatial structure determined by the gas parameters at the nozzle inlet and the geometrical parameters of the nozzles. Particularly difficult are the problems of describing atomic-cluster jets formed by outflow of condensing gas from convergent-divergent nozzles into vacuum. The gas-dynamic calculation

of the structure of such an atomic cluster target is very laborious and is a separate task that can be solved outside the scope of this study.

2. Procedure for calculating the number of photons emitted in the spectral band

Finding the radiation intensities in absolute units was carried out according to the following technique.

The recorded voltage from the detector generated during one laser pulse for our experiment can be defined as

$$V = \int_0^\infty \frac{\gamma \cdot \beta^2}{4\pi \cdot \alpha} \cdot E(\lambda) \cdot \delta(\lambda) \cdot T^2(\lambda) \cdot R(\lambda) d\lambda, \qquad (1)$$

where V [V] — the signal registered by the detector; α [C/V] — the sensitivity of the amplifier; γ [sr] — the solid angle at which the SXR radiation was observed from the detector; E [J/nm] — spectral density of the energy emitted by the plasma, δ [C/J] — sensitivity of the photodiode; $T(\lambda)$ — transmittance of a free-hanging film filter, $R(\lambda)$ — MXRM reflection coefficient.

For the directly used installation, filters and mirrors in accordance with [21], the energy concentrated in the emission line E_{line} and the number of photons N_{line} can be determined as follows:

$$E_{line} = \frac{4\pi \cdot \alpha V}{\gamma \delta T^2 R}, \qquad (2)$$

$$N_{line} = E_{line} \cdot \frac{\lambda_{line}}{hc}. \qquad (3)$$

For the spectral range studied by us, the sensitivity of the amplifier was $\alpha = 10^{-11}$ C/V; the solid angle at which the laser radiation was observed from the detector, $\gamma = 5.45 \cdot 10^{-5}$ sr; the sensitivity of the photodiode $\delta = 0.25$ C/J. T — transmittance of the used Ti/Be film filter at the studied wavelength, R — reflection coefficient Cr/Sc MXRM at the studied wavelength. The spectral resolution of the specular spectrometer is 0.06 nm, therefore, all measurements of absolute intensities are carried out in a spectral band with a width of at least 0.06 nm. If one emission line is located in the studied spectral band with a width of 0.06 nm and its surroundings, then its absolute intensity can be measured. If one emission line is located in the studied spectral band with a width of 0.06 nm and its surroundings, then its absolute intensity can be measured. The spectral resolution of the specular spectrometer is almost completely determined by the properties of the MXRM used. The effect of the size of the input aperture was evaluated and found to be insignificant.

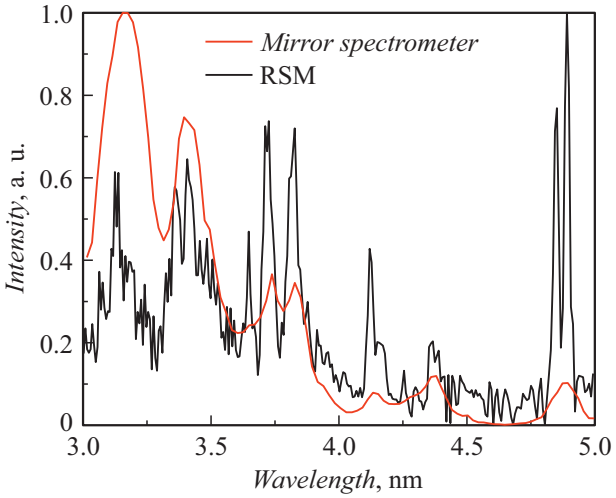

Figure 2. Emission spectra of argon under pulsed laser excitation.

3. Experimental

For all the gas-jet targets studied as part of the study, the studies were carried out in the following order: initially, the emission spectra on the RSM-500 and the mirror spectrometer were studied, then the absolute values of the radiation intensity were calculated.

3.1. Argon research

The view of the argon emission spectrum measured using the RSM-500 and a mirror spectrometer is shown in Fig. 2.

The decoding of the lines and their relative and absolute intensities are given in Table 1. The resolution of the specular spectrometer is noticeably inferior to the resolution of the RSM-500, which leads to a significant broadening of the lines. As you can see, the argon emission lines measured on the RSM-500 are located close enough to each other, therefore, according to the results of measurement on a mirror spectrometer, only absolute radiation intensities for bands with a width of 0.06 nm with a center falling on the wavelengths given in Table 1 can be given. Thus, the table shows the emission intensity in bands with a width of 0.06 nm in absolute units per full solid angle per laser pulse.

Table 1 shows that the radiation intensities are very high and comparable to those obtained in the works of [8,16,17]. Thus, when using argon-based targets, it is possible to implement a sufficiently intense SXR source emitting in a wide band. After additional measurements, it was found that the maximum intensity of SXR radiation in the range „of the water transparency window “ for argon is observed at a gas pressure at the nozzle inlet significantly less than 25 bar. Thus, when using argon-based targets to obtain even more intense SXR radiation, it is necessary to switch to more powerful laser systems.

Table 1. Decoding of the lines, their intensity according to RSM-500 and absolute values when using argon at a nozzle inlet pressure of 25 bar

Wavelength, nm	Ion	Transition	Intensity by RSM-500, a.u.	Energy in band, J	Intensity bands, ph/pulse
4.89	Ar IX	$2s^2 2p^6 - 2s^2 2p^5 3s$	540	$9 \cdot 10^{-4}$	$2.2 \cdot 10^{13}$
4.3	Ar X	$2s^2 2p^5 - 2s^2 2p^4 3s$	80	$5.54 \cdot 10^{-4}$	$1.2 \cdot 10^{13}$
4.15	Ar IX	$2s^2 2p^6 - 2s^2 2p^5 3d$	210	$3.17 \cdot 10^{-4}$	$6.6 \cdot 10^{12}$
3.83	Ar X	$2s^2 2p^5 - 2s^2 2p^4 3d$	380	$7.8 \cdot 10^{-4}$	$1.5 \cdot 10^{13}$
3.74	Ar X	$2s^2 2p^5 - 2s^2 2p^4 3d$	390	$7 \cdot 10^{-4}$	$1.3 \cdot 10^{13}$
3.4	Ar XI	$2s^2 2p^4 - 2s^2 2p^3 3d$	340	$8.5 \cdot 10^{-4}$	$1.5 \cdot 10^{13}$

Table 2. Decoding of lines, their intensity according to RSM-500 and absolute values when using krypton at the nozzle inlet pressure of 25 bar

Wavelength, nm	Ion	Transition	Intensity by RSM-500, a.u.	Energy in band, J	Intensity bands, ph/pulse
4.07	Kr XIII	$3d - 5f$	140	–	–
3.75	Kr XIV	$3d - 5f$	170	$2.1 \cdot 10^{-4}$	$3.9 \cdot 10^{12}$
3.39	Kr XV	$3d - 5f$	100	–	–

3.2. Krypton Studies

The view of the krypton emission spectrum measured using the RSM-500 and a mirror spectrometer is shown in Fig. 3.

The decoding of the lines and their relative and absolute intensities are given in Table 2. The resolution of the specular spectrometer is significantly inferior to the resolution of the RSM-500, which leads to a significant broadening of the lines. As you can see, when measuring krypton on the RSM-500, emission bands are visible, against which a number of lines appear. The decryption of these lines was carried out in accordance with [23,24]. Thus, based on the results of measurements on a mirror spectrometer, only absolute radiation intensities for bands with a width of 0.06 nm with a center falling on the wavelengths given in Table 2 can be given. The table below shows the emission intensity in bands with a width of 0.06nm in absolute units per full solid angle per laser pulse.

Table 2 shows that the radiation intensities are very low and comparable to those obtained in the work [8]. After additional measurements, it was found that the maximum intensity of SXR radiation in the range „of the water transparency window “ for krypton is observed at a gas pressure at the nozzle inlet significantly less than 25 bar. Thus, when using krypton-based targets to obtain intense SXR radiation, it is necessary to switch to more powerful laser systems.

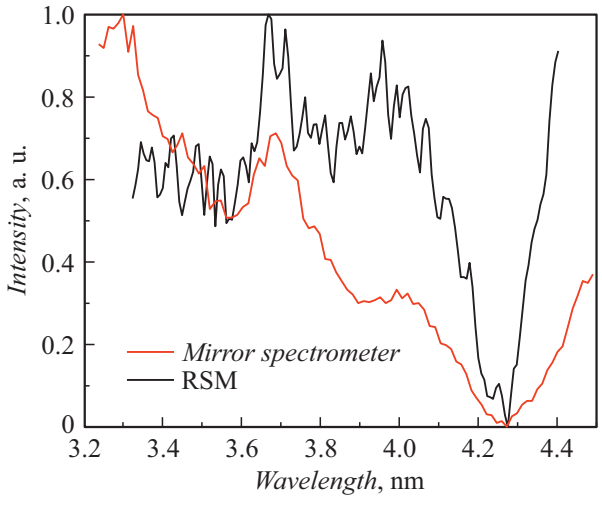

Figure 3. Emission spectra of krypton under pulsed laser excitation. The gas pressure at the nozzle inlet is 25 bar.

3.3. Nitrogen studies

A view of the nitrogen emission spectrum measured using the RSM-500 and a mirror spectrometer is shown in Fig. 4.

The interpretation of the observed line, its relative and absolute intensities are given in Table 3. The resolution of the specular spectrometer is significantly inferior to the resolution of the RSM-500, which leads to a significant broadening of the line. At the same time, the observed line is located in isolation, which allows absolute measurements of the radiation intensity. The nitrogen line corresponds to a wavelength of 2.88 nm, for which a strong absorption is observed in the MXRM used during operation of the mirror spectrometer, which leads to a sharp drop in the reflection coefficient. This also explains the low signal/background ratio, about 1 to 1.5. Nevertheless, we were able to estimate the absolute intensity of the 2.88 nm line. Thus, Table 3 shows the emission intensity for the 2.88nm line in absolute

Table 3. Decoding of the lines, their intensity according to RSM-500 and absolute values when using nitrogen at a nozzle inlet pressure of 25 bar

Wavelength, nm	Ion	Transition	Intensity by RSM-500, a.u.	Energy in line, J	Intensity line, ph/pulse
2.88	N VI	$1s^2-1s2p$	300	$1.2 \cdot 10^{-3}$	$1.7 \cdot 10^{13}$

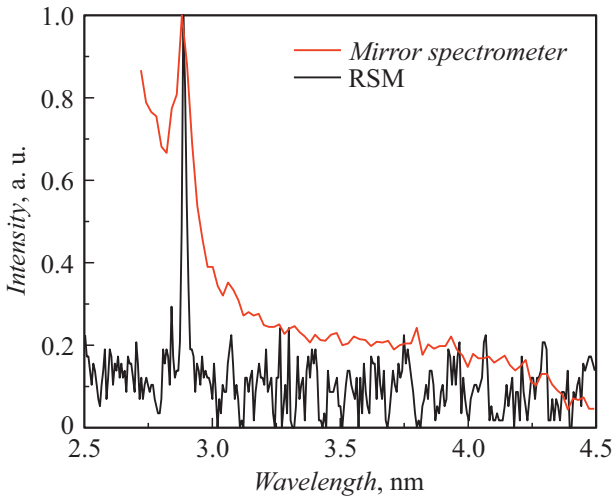

Figure 4. Emission spectra of nitrogen under pulsed laser excitation. The gas pressure at the nozzle inlet is 25 bar.

units per full solid angle per laser pulse. The presented value is estimated.

Table 3 shows that the radiation intensities are very low and comparable to those obtained in the work [{]2,15,25,26}. Thus, when using nitrogen-based targets, a very intense source of SXR radiation can be realized. The maximum intensity when using nitrogen is observed at the gas pressure at the nozzle inlet in the region of 25 bar.

3.4. Research CO_2, CHF_3 and CF_4

The type of carbon dioxide emission spectrum measured using the RSM-500 and a mirror spectrometer is shown in Fig. 5.

The decoding of the lines and their relative and absolute intensities are given in Table 4. The resolution of the specular spectrometer is significantly inferior to the resolution of the RSM-500, which leads to a significant broadening of the lines. At the same time, the lines are located far enough apart from each other, which allows for absolute measurements of the radiation intensity of these lines. Thus, the table shows the emission intensity for the lines in absolute units per full solid angle per laser pulse.

Table 4 shows that the radiation intensities are very low and comparable to those obtained in the studies [{]1,8,18}. Thus, when using targets based on CO_2, it is possible to implement a sufficiently intense source of SXR radiation. After additional measurements, it was found that the maximum intensity when using carbon dioxide is observed at a gas pressure at the nozzle inlet significantly greater than 25 bar.

Along with studies of carbon dioxide, studies of CHF_3 and CF_4 were conducted. The emission spectra of these gases are similar to the spectra for CO_2. The results of absolute measurements are given in Table 5.

It can be seen from the above values that when using targets based on CHF_3 and CF_4, the radiation intensity is reduced compared to CO_2, but still remains very significant. After additional measurements, it was found that the maximum intensity when using CHF_3 and CF_4 is observed at the gas pressure at the nozzle inlet in the region of 25 bar.

Conclusions

In this work, the emission spectra of gas jets Ar, Kr, CO_2, CHF_3, CF_4 and N_2 were studied when excited by pulsed laser radiation in the range „water transparency windows“ 2.3–4.4 nm. Emission spectra were obtained, lines were decoded, and absolute emission values from bright spectral lines were determined.

The results of the study make it possible to make a choice between different target gases when designing various equipment using laser-plasma radiation sources in its composition.

A separate comparison was made between different carbon-containing gases. It is established that the most

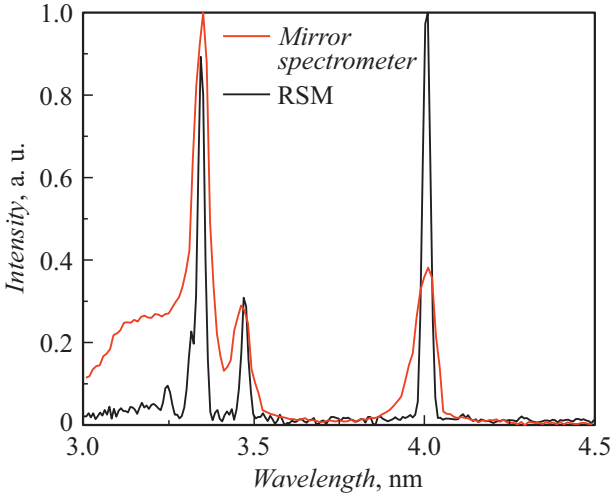

Figure 5. Emission spectra of CO_2 under pulsed laser excitation. The gas pressure at the nozzle inlet is 25 bar.

Table 4. Decoding of lines, their intensity according to RSM-500 and absolute values when using CO_2 at a nozzle inlet pressure of 25 bar

Length waves, nm	Ion	Transition by RSM-500, a.u.	Intensity lines, J	Energy lines, ph/pulse	Intensity
4.026	C V	$1s^2-1s2p$	2050	$4.9 \cdot 10^{-4}$	$9.9 \cdot 10^{12}$
3.49	C V	$1s^2-2s3p$	630	$1.54 \cdot 10^{-4}$	$2.7 \cdot 10^{12}$
3.37	C VI	$1s-2p$	1830	$4 \cdot 10^{-4}$	$6.9 \cdot 10^{12}$
3.343	C V	$1s^2-1s4p$	460	–	–
3.27	C V	$1s^2-1s5p$	200	–	–
2.84	C VI	$1s-3p$	260	–	–

Table 5. Absolute emission values for targets CO_2, CHF_3 and CF_4 at a pressure of 25 bar at the nozzle inlet

Length waves, nm	Ion	Transition	CO_2		CHF_3		CF_4	
			J	ph/pulse	J	ph/pulse	J	ph/pulse
4.026	C V	$1s^2-1s2p$	$4.9 \cdot 10^{-4}$	$9.9 \cdot 10^{12}$	$2.5 \cdot 10^{-4}$	$5 \cdot 10^{12}$	$1.9 \cdot 10^{-4}$	$3.85 \cdot 10^{12}$
3.49	C V	$1s^2-2s3p$	$1.54 \cdot 10^{-4}$	$2.7 \cdot 10^{12}$	$9.7 \cdot 10^{-5}$	$1.7 \cdot 10^{12}$	$7.5 \cdot 10^{-5}$	$1.3 \cdot 10^{12}$
3.37	C VI	$1s-2p$	$4 \cdot 10^{-4}$	$6.9 \cdot 10^{12}$	$2 \cdot 10^{-4}$	$3.3 \cdot 10^{12}$	$1.5 \cdot 10^{-4}$	$2.5 \cdot 10^{12}$

intense lines are formed when using gas-jet targets from CO_2.

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Conflict of interest

The authors declare that they have no conflict of interest.

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