Applications of the gas discharge sustained by the powerful radiation of THz gyrotrons

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Abstract. The terahertz frequency range currently remains the least studied from the point of view of gas-discharge physics. Recent progress in its mastering is associated, first of all, with the creation of powerful sources of terahertz radiation - gyrotrons. Nevertheless, the discharge sustained by the powerful radiation of the terahertz frequency range is of interest not only from the fundamental point of view (because of its lack of knowledge), but also from the applied one. This paper presents results of investigations aimed at studying the point-like THz discharge as a source of vacuum ultraviolet radiation and at research of CW THz plasma torch existing at atmospheric pressure and therefore having promising applications in various fields: plasma medicine, plasma chemistry, material processing etc. It is shown that it is possible to create a point-like plasma in an inhomogeneous gas flow with the density above the cut-off one (4·10¹⁵ cm⁻³ for 0.67 THz). It has been demonstrated that such plasma is an effective source of VUV and EUV radiation. Investigations of the atmospheric plasma torch, sustained by CW sub-terahertz radiation (1 kW @ 0.26 THz), demonstrated the possibility of the creation dense non-equilibrium plasma (2·10¹⁵ cm⁻³) with electron temperature in the range from 1 to 2 eV.

1. Introduction

The mastering of the terahertz frequency band from the point of view of the gas discharge physics has recently become possible due to the creation of powerful generators of electromagnetic radiation in the specified band — gyrotrons [1–4] and free electron lasers [5]. The interest shown for the terahertz discharge is also associated with potential applications. In this paper, two such applications are considered: discharge in an inhomogeneous gas flow as a point-like source of extreme ultraviolet radiation for projection lithography and a plasma torch of atmospheric pressure for plasma chemical applications.

2. Point-like source of VUV and EUV

In [6], it was proposed to use a discharge in an inhomogeneous gas flow as a point source of vacuum and extreme ultraviolet. The idea is that high-pressure gas (1-5 atm) is fed into a continuously evacuated vacuum discharge chamber through a nozzle of small diameter (about 100 μm). The rate of pumping and gas inlet is regulated in such a way as to maintain the background pressure in the discharge chamber at a level of 10⁻² Torr and below to ensure the necessary degree of gas flow heterogeneity and background gas transparency for VUV and EUV radiation (see figure 1). Inhomogeneous gas inlet provides localization of the breakdown conditions for the radiation of the
terahertz frequency band only in the region of high gas pressure (dozens and hundreds of Torr) near the gas inlet opening. In this case, the discharge, arising under the action of THz radiation, directed into the discharge chamber through the input dielectric window, in the high-pressure region practically does not propagate towards heating radiation, which ensures its localization. In [7], using the 0.67 THz gyrotron, it was experimentally demonstrated that it is possible to create and maintain a point-like discharge with characteristic sizes of less than 1 mm in a non-uniform gas flow.

![Figure 1](image1)

**Figure 1.** Scheme of the point-like discharge in an inhomogeneous gas flow. High-pressure gas (1-5 atm) is fed into a continuously evacuated vacuum discharge chamber through a nozzle of small diameter. THz radiation comes from the left.

For efficient generation of multiply charged ions in such a discharge, the line radiation of which is the source of VUV and EUV, it is necessary to maintain a sufficiently high plasma density and electron temperature (dozens of eV). Measurement of the electron density in a point-like discharge in argon from the Stark broadening of the hydrogen Balmer series demonstrated the ability to maintain a plasma with a density not less than cut-off for a given heating frequency ($4 \times 10^{15}$ cm$^{-3}$ for 0.67 THz) [8]. It is worth noting that under these conditions it is possible to effectively absorb heating electromagnetic radiation at plasma resonance, and commensurability of plasma size and wavelength of heating radiation ensures good coupling between radiation and plasma. Indirectly, the presence of a substantial electron temperature (dozens of eV) in a point-like THz discharge was demonstrated in [9], where the presence of radiation from the discharge in the EUV band was shown, as well as the fact that observed ratio between different bands, including the visible one (figure 2), is possible only at a plasma density of $10^{16}$ cm$^{-3}$ and electron temperature of 50 eV. Unfortunately, in these experiments, the wavelength of sub-terahertz radiation (1.2 mm) was noticeably larger than the size of the discharge (hundreds of μm), so most of the heating radiation did not hit the discharge, and it was not possible to achieve higher power values in the EUV.

![Figure 2](image2)

**Figure 2.** Photo of the point-like discharge in xenon in visible band. Background gas pressure $4 \times 10^{-3}$ Torr. Sub-terahertz radiation (250 kW @ 0.25 THz) comes from the top.

All of the above leads us to the fact that to create an effective EUV source, it is necessary to switch to shorter wavelengths, which will provide better focusing and a higher plasma density. In particular, experiments on the Novosibirsk FEL (130 μm) [10] demonstrated the possibility of maintaining plasma with a density of $10^{17}$ cm$^{-3}$, which is optimal from the point of view of the conversion of heating radiation into radiation with a wavelength of about 10 nm [11].

At present, powerful gyrotrons with a radiation frequency of about 1 THz have been created and are being improved at IAP RAS and experiments are planned to create a point-like discharge in xenon.

### 3. CW plasma torch

At present, interest in the development of reliable sources of non-equilibrium atmospheric pressure plasma is growing in a wide range of plasma-chemical applications [12]. This is due to the need of...
modern industry in the implementation of the processes of decomposition of highly stable molecules and carrying out reactions with a high activation threshold.

To maintain disequilibrium in the plasma of atmospheric pressure, a high energy input to the discharge is needed, which can be provided by modern microwave and THz sources of radiation — gyrotrons. The discharge maintained in quasi-optical wave beams is ideal for obtaining and practical use of reactive plasma with a high degree of purity due to the absence of electrodes and the localization of the discharge far from the chamber walls.

Studies of the plasma of the sub-atmospheric and atmospheric pressure ranges, maintained by continuous microwave radiation from a magnetron with a frequency of 2.45 GHz are widely known [13]. The interest of researchers here is associated with the wide availability of such sources and the relatively simple electrodeless design of the plasma torch. Plasma, sustained by the radiation of a magnetron, is widely used in plasma chemistry for carrying out chemical reactions in the pressure range from $10^{-3}$ to 760 Torr [14]. However, due to the relatively long wavelength of radiation, the power density currently available in these sources is insufficient to maintain a non-equilibrium discharge at atmospheric pressure. Such atmospheric pressure plasmatrons are generators of equilibrium plasma. The transition to a higher frequency of radiation allows, on the one hand, to increase the power density input into the discharge by improving the focusing of the radiation, on the other hand, to increase the plasma density, which accelerates various reactions involving electrons.

In this paper, we studied the non-equilibrium discharge of atmospheric pressure, maintained by focused radiation of a gyrotron with a frequency of 0.26 THz and a power of up to 1 kW. In particular, the electron density was measured by the Stark broadening of the hydrogen Balmer series lines ($H_\alpha$ and $H_\beta$) and the electron temperature was measured by study of plasma emission spectra. The research of such a discharge is of interest for a number of important applied problems in plasma chemistry.

Figure 3. Scheme of the experimental setup. 1 — gyrotron, 2 — defocusing mirror, 3 — focusing mirror, 4 — discharge chamber, 5 — THz beam waist, 6 — gas inlet tube.

Figure 3 shows the experimental setup. The gyrotron radiation (1) by means of two quasi-optical mirrors (2) - (3) was directed into the discharge chamber (4). The power density at the focus of the second mirror (5) reached 20 kW/cm$^2$, which corresponds to an intensity of the rms electric field of 2.7 kV/cm. The end of the metal tube (6), through which the plasma-forming gas was supplied, was led to the beam waist. The inner diameter of the tube was 4 mm. Discharge, initiated by a high-voltage spark at the end of the gas tube, and was maintained by focused THz radiation. The discharge is an elongated torch, about 1 cm long and with a diameter equal to the diameter of the gas tube. In a series of experiments, argon was used as a plasma-forming gas, and the discharge was ignited in an external air atmosphere at atmospheric pressure. The flow of argon could be changed in the range from 5 to 30 l/min.

Plasma density was measured by the Stark broadening of the hydrogen atom lines. Figures 4 and 5 show examples of the $H_\alpha$ and $H_\beta$ line profiles recorded using an MS5204i monochromator with a instrumental full width at half maximum of 0.17 nm. The measured line widths at half maximum for hydrogen in this case were 0.2 nm and 0.44 nm, respectively. Taking into account the instrumental width, it turns out that the real half-width of the lines was 0.12 and 0.4 nm, which gives the same
plasma concentration with good accuracy [15] $2 \cdot 10^{15}$ cm$^{-3}$. It should be noted that this value exceeds the cut-off value for 0.26 THz ($7 \cdot 10^{14}$ cm$^{-3}$) and practically does not depend on the power supplied to the discharge.

![Figure 4](image1.png)  **Figure 4.** An example of a measured fragment of the spectrum with H$\alpha$. The half-width is 0.2 nm.

![Figure 5](image2.png)  **Figure 5.** An example of a measured fragment of the spectrum with H$\beta$. The half-width is 0.44 nm.

The emission spectra of pure argon plasma were recorded in the range 300-1000 nm using an ASEQ LR1 emission spectrometer with a resolution of 0.3 nm. Having the relative intensities of the argon atomic emission lines and taking into account their transition constants, we can determine the electron temperature by comparing these intensities [16]. Based on the obtained spectra (for example, see figure 6), the dependence of the electron temperature on the power was determined. The values of the electron temperature were in the range from 1 to 2 eV.

![Figure 6](image3.png)  **Figure 6.** Emission spectra of pure argon plasma torch surrounded by the air.

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**References**
[1] Glyavin M Yu, Luchinin A G, Nusinovich G S, Rodgers J, Kashyn D G, Romero-Talamas C A and Pu R 2012 *Appl. Phys. Lett.* **101** 153503
[2] Bratman V L, Kalyonov Yu K and Manuilov V N 2009 *Phys. Rev. Lett.* **102** 245101
[3] Glyavin M Yu et al 2015 *Rev. Sci. Instrum.* **86** 054705
[4] Denisov G G et al 2018 *Rev. Sci. Instrum.* **89** 084702
[5] Bolotin V P et al 2004 Status of the Novosibirsk free electron laser and first experiment with high power terahertz radiation Proc. 1 Int. Conf. on Submillimeter Sci. and Tech. (Ahmedabad, India) pp 1–8

[6] Glyavin M Yu, Golubev S V, Izotov I V, Litvak A G, Luchinin A G, Razin S V, Sidorov A V, Skalyga V A and Vodopyanov A V 2014 Appl. Phys. Lett. 105 174101

[7] Glyavin M Yu, Golubev S V, Zorin V G, Izotov I V, Litvak A G, Luchinin A G, Morozkin M V, Razin S V, Sidorov A V and Skalyga V A 2014 Radiophys. and Quantum Electron. 56 561–5

[8] Sidorov A V, Razin S V, Golubev S V, Safronova M I, Fokin A P, Luchinin A G, Vodopyanov A V and Glyavin M Yu 2016 Phys. Plasmas 23 043511

[9] Shalashov A G, Vodopyanov A V, Abramov I S, Sidorov A V, Gospodchikov E D, Razin S V, Chkhalo N I, Salashchenko N N, Glyavin M Yu and Golubev S V 2018 Appl. Phys. Lett. 113 153502

[10] Kubarev V V, Getmanov Ya V and Shevchenko O A 2017 AIP Advances 7 095123

[11] Abramov I S, Gospodchikov E D and Shalashov A G 2018 Phys. Rev. Applied 10 034065

[12] Méndez I, Gordillo-Vázquez F J, Herrera V J and Tanarro I 2006 J. Phys. Chem. A 110 6060–6

[13] Choi S Y, Minami W, Kim L H and Kim H J 2007 Solid State Phenomena 124–126 1621–4

[14] Latrasse L, Radoiu M, Lo J and Guillot P 2017 Journal of Microwave Power and Electromagnetic Energy 51 43–58

[15] Ochkin V N 2009 Spectroscopy of Low Temperature Plasma (New York: Wiley) p 362

[16] Zhu X M and Pu Y K 2010 J. Phys. D: Appl. Phys. 43 403001