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Electron density in dielectric barrier discharge argon plasma jet determination by using a microwave waveguide filter

© M.S. Usachonak,[1] Yu.S. Akishev,[2] A.V. Kazak,[1] A.V. Petryakov,[2] L.V. Simonchik,[1] V.V. Shkurko[3]

[1] B.I. Stepanov Institute of Physics of the National Academy of Sciences of Belarus,
Minsk, Belarus
[2] Troitsk Institute for Innovation and Fusion Research (TRINITI),
Troitsk, Moscow, Russia
[3] Graduate School of the National Academy of Sciences of Belarus,
Minsk, Belarus
e-mail: m.usachonak@dragon.bas-net.by

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Using a waveguide microwave filter, the electron density in an argon plasma jet of a dielectric barrier discharge was determined. The dynamic of the electron density behavior during the period of applied voltage with a frequency of about 100 kHz was determined. The 4 maxima of the average concentration, which are about $10^{13}\,cm^{-3}$, were observed.

Keywords: dielectric barrier discharge, waveguide microwave filter, transmission spectrum, electron density, plasma jet.

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Introduction

In the last decade plasma jets were one of the actual objects of study of non-equilibrium plasma at atmospheric pressure due to their biomedical applications [1–4] due to their ability to produce reactive oxygen species (ROS) and reactive nitrogen species (RNS) [5]. Atmospheric pressure jets can be classified according to the type of discharge used to generate them, including discharges of direct current, alternating current, radio frequency and microwave [6–10]. The high-frequency plasma jet is suitable for biomedical applications due to its relatively low cost and electrical safety, and the ability to treat non-conductive objects [1,4,5]. The reactive species are formed during the transport of species (electrons, ions, excited species, radicals) from the active region of the discharge to the sample being treated. These reactive species are closely related to electron impact reactions [11]. Therefore, on the one hand, it is necessary to know the electron density in order to understand and increase the generation of bioactive species. On the other hand, it is important to know the value of the electron density n_e when checking the developed plasma models, since n_e is usually the main input parameter [12].

There are several methods for measuring the electron density in the range $10^{12}-10^{13}\,cm^{-3}$ in plasma at atmospheric pressure with low gas temperature [13,14]. However, these methods either have insufficient sensitivity or do not allow tracking the dynamic processes in nonstationary discharges. One of the effective methods for diagnostic of the gas-discharge plasma is the scattering of laser radiation [15,16]. It is characterized by a good spatial resolution up to several tens of microns, but it has limited sensitivity, and its use is difficult in plasma diagnostics with an electron density of about $10^{13}\,cm^{-3}$ and below. Due to the low level of the useful signal, long-term accumulation is required, which does not allow the method of laser radiation scattering use in the study of non-stationary discharges.

The analysis of spectral data is also widely used to determine the electron density of plasma. Thus, the method of Stark line broadening H_β, based on optical emission spectroscopy, has good spatial and temporal resolution and is relatively easy to implement. This method was successfully used to study atmospheric pressure plasma in discharges of various types [17–19]. However, it is not easy to use this method to measure the electron density in cold plasma jets, since a very detailed analysis of the mechanisms of line broadening [19] is required at low electron density.

A large number of works are devoted to the probing of ionospheric and various types of gas-discharge plasma at low pressure by radio waves of various frequency ranges. However, to date, there are few studies using microwave methods to study atmospheric pressure plasma, in particular, plasma jets. Nevertheless, diagnostics of pulsed discharge between two electrodes in air was successfully performed in [20] with a high time resolution, and the results of determining the electron density in the barrier discharge jet and unipolar pulsed discharge in a helium flow are presented in [13,14], respectively. The disadvantage of the scattering method is that various external factors can strongly influence at plasma probing in free space. In [21] an attempt was made to determine the electron density in helium plasma jet at atmospheric pressure using a cylindrical microwave resonator with the TE112 mode.

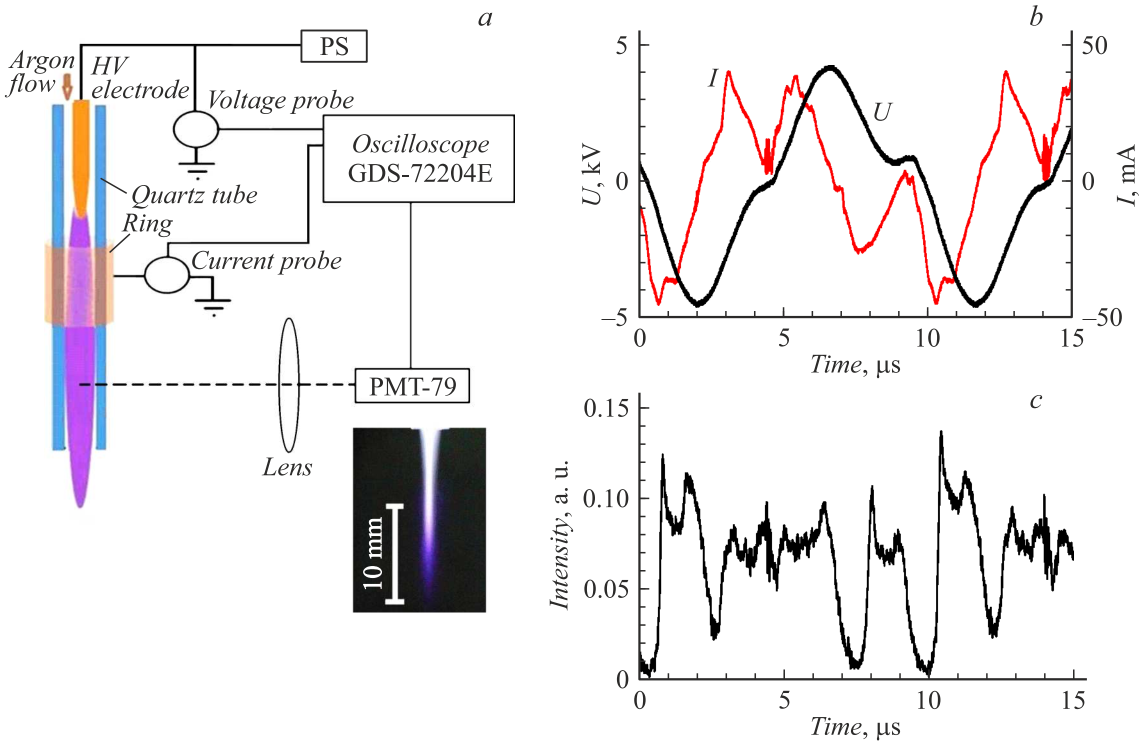

Figure 1. *a* — scheme of electrical connection of the DBD jet and its photograph; *b* — voltage and discharge current oscillograms; *c* — integral luminescence of jet.

The positive column of a non-self-sustained atmospheric pressure glow discharge in argon was used in [22,23] as a transmission control element of 10-cm waveguide filter with 6 rod diaphragms that form 5 coupled resonators. It was shown that the filter transmission at a frequency of 2840 MHz changes by about 10 dB in the range of discharge currents 0–50 mA. This change in transmission may be sufficient to determine the electron density in the argon plasma jet of a dielectric barrier discharge.

As is known from the theory and practice of cavity measurements [24], the plasma introduction into the cavity set to one of its eigenfrequencies leads to a shift in its resonant frequency. The magnitude of this shift is proportional to the relative change in the plasma concentration, if the electron collision frequency ν_e in the plasma is much less than the resonant frequency $\omega_0 = 2\pi f_0$. In the case of the atmospheric pressure plasma, the collision frequency ν_e can significantly exceed the resonant frequency ω_0. In this case, the resonant frequency shift and cavity transmission will depend on both the electron density n_e and the collision frequency ν_e.

In the general case, the plasma permittivity ε_p for microwave radiation propagating in the plasma with frequency f_0 is given by the expression

$$\varepsilon_{pe} = 1 - \frac{\omega_{pe}^2}{\omega_0^2(1 - i\,\frac{\nu_e}{\omega_0})} = 1 - \frac{e^2 n_e}{\varepsilon_0 m_e \omega_0^2(1 - i\,\frac{\nu_e}{\omega_0})}, \qquad (1)$$

where ω_{pe} — electron plasma frequency, e — electron charge, ε_0 — vacuum dielectric constant and m_e — electron mass. In this case, the plasma conductivity σ is determined as follows:

$$\sigma = \sigma_{re} + i\sigma_{im}, \quad \sigma_{re} = \frac{e^2 n_e}{m_e}\frac{\nu}{\omega^2 + \nu^2},$$

$$\sigma_{im} = -\frac{e^2 n_e}{m_e}\frac{\omega}{\omega^2 + \nu^2}. \qquad (2)$$

In the present work, first, to test the method, studies are performed with gas-discharge plasma source with more or less identified plasma parameters, and then the waveguide filter method is applied to the argon jet of dielectric barrier discharge (DBD).

1. Experimental setup

The source of the plasma jet was the dielectric barrier discharge excited by a high-frequency voltage source in argon inside a quartz tube (Fig. 1). The outer diameter of the tube was 4 mm and the inner diameter — 2.5 mm. Argon was blown through the tube at flow rate of 10 l/min, the gas speed was about 30 m/s. The inner electrode was a metal rod 1 mm in diameter, located along the tube axis. The outer electrode in the form of a copper foil cylinder 20 mm wide was fixed at a distance of 2 mm from the end of the rod electrode. The distance from the outer ring electrode to the edge of the quartz tube was 2 cm. AC voltage with an amplitude of 4 kV and frequency of $f_0 \sim 100$ kHz was applied to the electrodes from PS power supply (Fig. 1, *a*). Typical oscillograms of the DBD current and voltage are shown in Fig. 1, *b*. It can be seen that the voltage at the electrodes remains close to sinusoidal. At the same time, the discharge current oscillogram differs from the initial sinusoidal form, which is recorded without a plasma jet, namely, there are 4 current maximums during the period. This may be due to the discharge current flow through two circuits: a) high-voltage electrode–plasma–barrier–capacitance–external grounded electrode and b) high-voltage electrode–jet plasma–capacitance of surrounding space–grounded objects. Such situation was discussed in [25]. The equivalent electrical circuit of the barrier discharge with plasma jet is shown in Fig. 2.

A photograph of plasma jet in argon is shown in Fig. 1, *a* (insert in bottom). The DBD integral luminescence in the visible region of the spectrum was registered using a photomultiplier PMT-79, the signal from which was recorded on a digital oscilloscope GDS-72204E. The oscillogram in Fig. 1, *c*, shows the change in time of the intensity of DBD plasma luminescence and, in general, reflects the dynamics of the change in the discharge current.

To measure the electron density in the plasma jet of the DBD, a waveguide filter of 10-cm wavelength range [23] was used, which was made from a segment of a rectangular waveguide with cross-section of 90 × 10 mm and a length of 500 mm. In the cross-sections of the waveguide, there were 6 rod diaphragms (Fig. 3.) at distances of about $\Lambda/2$ from each other ($\Lambda = \frac{\lambda_0}{\sqrt{1-(\lambda_0/2a)^2}}$ — wavelength in waveguide). Each diaphragm is a set of three cylindrical rods located perpendicular to the waveguide axis at distances $a/4$ from each other, where $a = 90$ mm — the size of the wide wall of the waveguide. These 6 diaphragms form 5 coupled filter resonators. The filter transmission spectrum was registered using a Keysight FieldFox N9918A portable vector spectrum analyzer. The time evolution of the filter transmission at individual frequencies was registered using G3-22 microwave generator, a detector section, and a GW Instek GDS-72204E digital oscilloscope.

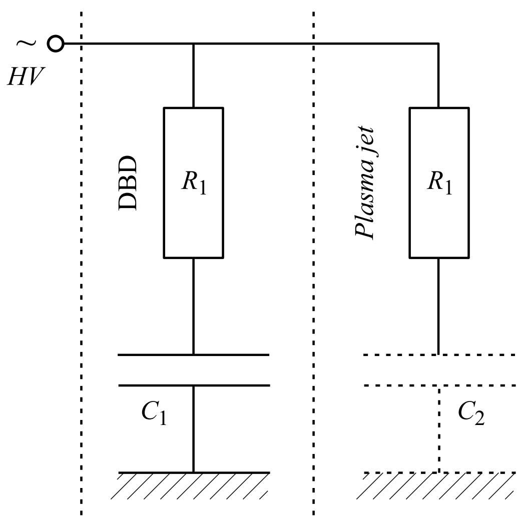

Figure 2. Equivalent electrical circuit of barrier discharge with plasma jet in free space [24]. R_1 and R_2 — effective resistances of barrier discharge plasma and plasma jet, C_1 and C_2— capacitance of the barrier discharge and plasma jet.

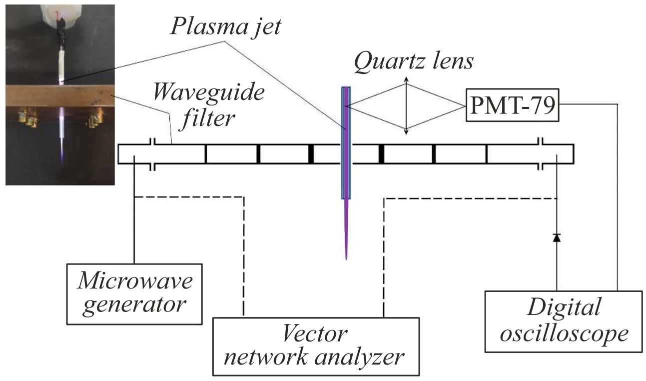

Figure 3. Block diagram of filter transmission measurement.

2. Results and discussion

At the first stage, a positive discharge column in a standard neon lamp GSh-5, known as a noise generator in microwave technology, was used as a plasma inhomogeneity with known parameters placed into the middle resonator of the filter. The outer diameter of the discharge tube GSh-5 is 3.8 mm, the inner diameter is — 3.0 mm, the interelectrode distance is 18 cm, the lamp has incandescent cathode. The lamp is filled with neon at a pressure of about 70 Torr.

When the discharge in the lamp GSh-5 is switched on, the transmission spectrum of the filter changes (Fig. 4, *a*). It can be seen that the discharge current increasing leads to decrease of the microwave signal level passing through the filter, and its transmission spectrum shifts towards higher frequencies. Note that at discharge current of 30 mA the transmitted signal decreases by more than one order of magnitude. In order to determine frequency, at which the greatest change in the transmitted signal occurs, the spectra from Fig. 4, *a* were normalized on the transmission spectrum of the filter with lamp without switching on the discharge. The normalized transmission spectra are shown in Fig. 4, *b*. It can be seen that the most significant change in transmission occurs in the range 2.63–2.64 GHz.

The electron density and collision frequency in the GSh-5 lamp were determined from the reduced electric field strength using the BOLSIG+ program [26]. To do this, the voltage and the corresponding discharge current in the lamp were measured, and then the magnitude of the reduced electric field and the current density were calculated. The electron density dependence on the discharge current in the GSh-5 lamp is shown in Fig. 5. Consider that the electron

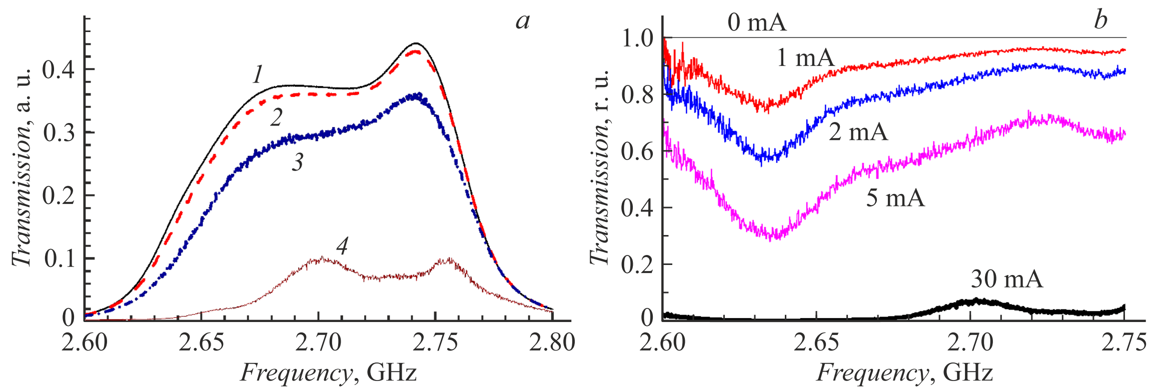

Figure 4. Transmission spectrum of microwave filter at different values of the discharge current in the GSh-5 lamp. *1* — 0, *2* — 1, *3* — 5, *4* — 30 mA.

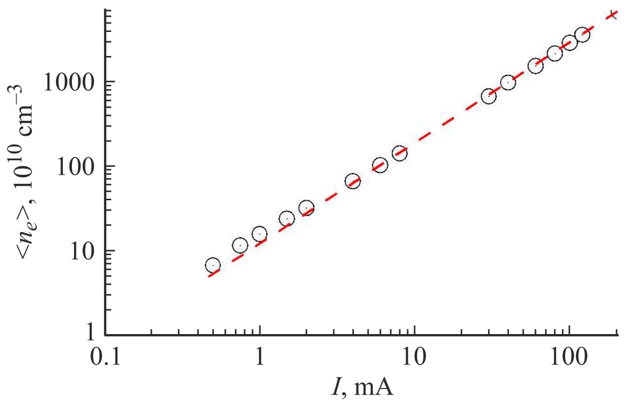

Figure 5. The electron density dependence on discharge current in the GSh-5 lamp.

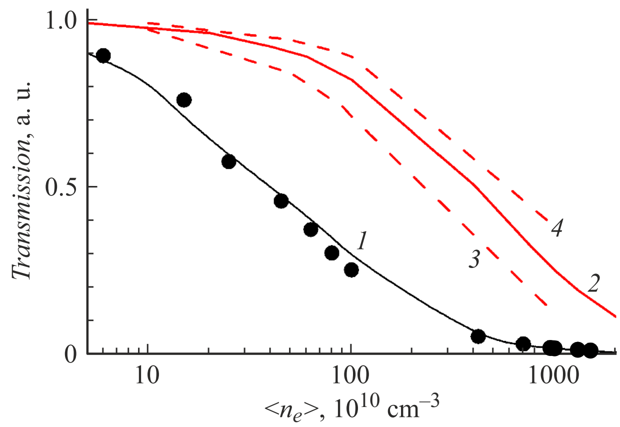

Figure 6. The dependence of the transmission spectrum change at frequency of 2.635 GHz on electron density. *1* — GSh-5, *2–4* — argon plasma jet with $\nu = 3 \cdot 10^{11}$, $1.5 \cdot 10^{11}$ and $4.5 \cdot 10^{11}\,s^{-1}$ respectively.

density $\langle n_e \rangle$ obtained in this way is averaged over the entire positive discharge column in GSh-5.

The calculated value of the collision frequency for the discharge current 5 mA is $4 \cdot 10^{10}\,s^{-1}$. For comparison according to [24] the effective collision frequency in neon, calculated from the data on collision cross-sections in [27] is $(5-7) \cdot 10^{10}\,s^{-1}$.

A certain dependence of the average concentration $\langle n_e \rangle$ on the discharge current (Fig. 5) and the corresponding values of the filter transmission at these current values make it possible to find the dependence of the transmission at frequency of 2.635 GHz on the electron density (circles in Fig. 6). The obtained data were compared with the results of calculating the dependence of the filter transmission on the electron density, performed in the Ansys HFSS program similarly [28,29] (curve *1*, Fig. 6), in which the model of the above waveguide filter was implemented, and the material „plasma“ was specified through the values of the real parts of the permittivity and conductivity (1) and (2). The transmission is normalized on the transmission of the waveguide filter without discharge in the GSh-5 tube. In this case, the electron density varied from 10^9 to $10^{14}\,cm^{-3}$, the collision frequency was assumed to be constant and equal to $4 \cdot 10^{10}\,s^{-1}$. As can be seen, the obtained results are in good agreement with the experimental data, which made it possible to use the developed model to estimate the microwave filter transmission versus the electron density in the DBD plasma jet in argon.

The luminous region of the plasma jet has a length of about 15 mm (Fig. 1, *a*). This exceeds the size of the narrow wall of the waveguide (10 mm). However, to isolate the jet from touching the grounded waveguide, the tube-DBD barrier was elongated by 6 cm. At the same time, a jet was observed at the end of the elongated tube, although it was somewhat shorter than that shown in Fig. 1, *a*. A quartz tube with plasma jet was placed into holes in the wide walls of the waveguide at the center of the middle resonator (Fig. 3, insert on the left). Argon at atmospheric pressure is used as the working gas in the investigated DBD plasma jet. The collision frequency estimate made using BOLSIG+ gives $\nu \sim 3 \cdot 10^{11}\,s^{-1}$. Using this value of the collision frequency ν, we calculated the transmission of the

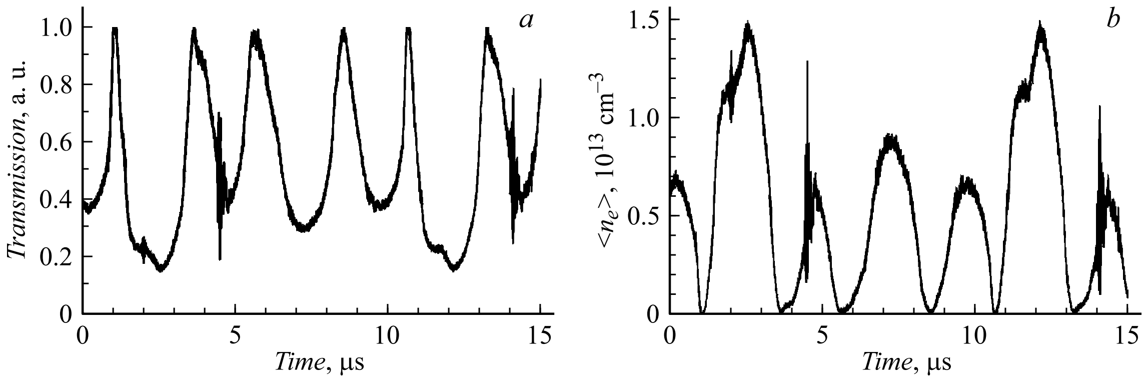

Figure 7. *a* — oscillograms of transmission of waveguide filter with plasma jet; *b* — temporal evolution of the electron density in the plasma of DBD argon jet.

waveguide filter as a function of the electron density in the plasma inhomogeneity. This dependence is shown in Fig. 6 (curve *2*). Comparing curves *1* and *2*, we can conclude that the filter transmission at the same electron density decreases more significantly at lower collision frequency. This can be explained by the fact that, as follows from formula (2), the real part of the conductivity decreases with increasing in collision frequency. A higher concentration is needed to compensate for this decrease. To estimate the influence of the electron collision frequency on the change of the filter transmission and, accordingly, on the value of the determined electron density, calculations were performed at the collision frequency $1.5 \cdot 10^{11}$ and $4.5 \cdot 10^{11}\,s^{-1}$. These dependences are shown in Fig. 6 by dashed lines *3* and *4*.

The obtained dependence (Fig. 6, curve *2*) was used to estimate the time evolution of the electron density in the plasma of DBD argon jet. Fig. 7, *a* shows oscillogram of the waveguide filter transmission when the barrier discharge is switched on (see above for the parameters). It can be seen that during the period transmission passes through minima 4 times, which may indicate maxima of the electron density. Using the dependence of transmission on concentration from Fig. 6 (curve *2*) and the obtained change in time of filter transmission (Fig. 7, *a*), one can obtain the time evolution of the electron density in barrier discharge jet. The result is shown in Fig. 7, *b*.

As follows from Fig. 7, *b*, the maximum value of the average electron density in the jet is $\sim 10^{13}\,cm^{-3}$, which agrees with the known data [1]. It can also be seen that the presented method of microwave waveguide filter has a good time resolution.

Conclusion

Using the microwave waveguide filter, the electron density in plasma jet of the dielectric barrier discharge in argon atmosphere was determined with rather high resolution in time. The waveguide filter of the 10-cm wavelength range is made using a segment of a rectangular waveguide with cross-section of 90×10 mm and length of 500 mm. In the cross-sections of the waveguide, at distances of about half of the wavelength in waveguide, 6 rod diaphragms were placed, which formed 5 coupled resonators. The method of the electron density determination is based on measuring the filter transmission and comparing it with the calculated data. To test the efficiency of the method the gas-discharge sources with known parameters were used. The maximum value of the average electron density in the argon jet of the dielectric barrier discharge was $10^{13}\,cm^{-3}$, which agrees with the known data [1].

The presented method of the electron density determination in DBD plasma jet can serve as an addition to optical methods, and can also be used to determine the electron density in plasma jets enclosed in opaque dielectric tubes, for which optical measurements are excluded.

Conflict of interest

The authors declare that they have no conflict of interest.

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