

# Supplemental materials at articles

## “Phenomenon of apokamp discharge”

**Introduction.** There are data on formation of atmospheric-pressure plasma jets and pulse corona discharges in air, as well their radiation characteristics in this supplement. Carried out researches have allowed to determine the conditions under which plasma jets with length up to 4 cm are formed in air of atmospheric pressure. When applying voltage pulse with an amplitude of 250 kV corona discharge with length of separate jets up to 40 cm was formed. In this case, X-ray was registered. It was shown that the emission spectrum in the wavelength range of 200–800 nm of plasma jets and corona discharges ignited at atmospheric pressure air and the pulse voltage applying to a tip electrode, is formed mainly by spectral transitions of the second positive system of a nitrogen molecule. Carrying out of studies on plasma jets and pulse corona discharge has revealed phenomenon of apokamp discharge.

**Formation of plasma jets in the air and nitrogen of atmospheric pressure.** At the present time, atmospheric-pressure plasma jets (APPJ) are object of investigations of large number of research groups. This is due to the plasma jets application prospects in medicine for the inactivation of biological systems and modification of surfaces of various materials (cleaning, etching, thin film deposition, etc.) APPJ is formed in the discharge (glow, spark, arc, radio frequency (rf), barrier, etc.) and is ejected through a narrow nozzle (circular or slit in the cross-section) due to creation of overpressure (above atmospheric) in the discharge zone. Excitation by a glow, a barrier or a spark discharge leads to formation of a non-equilibrium plasma having an average temperature of heavy particles (gas temperature) from 20 to 400 °C and density of charged particles typical of weakly ionized gases (not higher than  $10^{11}$ – $10^{12}$  cm<sup>-3</sup>). Such plasma is called cold and non-thermal at temperatures values closed to “room temperature”.

By now, APPJ sources based on barrier discharge in He, Ar, N<sub>2</sub>, air and in mixture of rare gases with additive of nitrogen and oxygen have been created and studied. To obtain the plasma, in this case, voltage pulses of negative and positive polarity with duration of 0.1–1 μs, amplitude of up to 30 kV and repetition rate of tens kHz are applied across the interelectrode gap. In addition, gas pumping with rate of ones-tens l/min is used. A number of papers refer to the use of rf excitation method. A large number of papers are devoted to the study of the effect of excitation pulse parameters and source design on geometry of the plasma jet and its spectral characteristics. Much less attention is paid to the measurement of the energy characteristics of an APPJ, its temperature, including the temperature profile along the jet’s cross-section. This is especially relevant for modes in which the gas temperature exceeds 100 °C. This usually occurs when replacing the inert gas by their mixtures containing additives of molecular electronegative gas, air or nitrogen. Then, with one hand, a larger amount of chemically active species is formed, but on the other hand, high voltage (> 15 kV) is required. In addition, when using molecular gases, a plasma’s stream becomes sensitive to the gas pumping rate and its temperature increases. Therefore, such source of plasma can no longer be called a low-temperature source.

As a result of our research a stable plasma jet at barrier discharge in the air and nitrogen of atmospheric pressure with minimal gas flow rate (up to 1 l/min) was obtain. Energy, temperature and spectral characteristics of this object were determined. In this case, barrier discharge was ignited in a quartz tube with a 1.5 mm-diameter nozzle. Excitation was performed by voltage pulses from the pulser # 1. This pulser allows to vary the voltage pulse duration in the range of  $\tau = 1$ –1.5 μs, pulse repetition rate  $f$  in the range of 10–90 kHz and the amplitude of the voltage pulses in the range of 5–13 kV. For recording of voltage pulses and pulses of discharge current a resistive voltage divider and current shunt based on low-inductance thin-film chip-resistors were used. In quartz tube flow of nitrogen or air is fed. Monitoring of gas flow rate was carried out with the U30/AR40 flowmeter and pressure sensor PSE 511. A signal from sensor was recorded with digital oscilloscope. The radiation power of APPJ was measured with a photodetector HAMAMATSU H8025-222 (with a maximum of spectral sensitivity at wavelength of 222 nm), placed on the plasma flow axis at various distances  $x$  from nozzle.

Input window of the photodetector was isolated from the flow of heat by means of quartz plate. To measure gas temperature, the thermocouple TPK 011–0.5/3 with the thermoregulator OVEN TRM202 is used. An emission spectrum of plasma was registered through an optical fiber with a transmission spectrum connected with the spectrometer HR2000+ES (Ocean Optics, Inc.) based on multichannel CCD Sony ILX511B (working range of 200–1100 nm, spectral half-width of the instrumental function is approximately 1.33 nm).

Typical waveforms of discharge current  $I$  and voltage  $U$  pulses which take place during the formation of the plasma jet in the air and integral image of glow of APPJ are shown in Fig. 1. To achieve a steady jet expiration

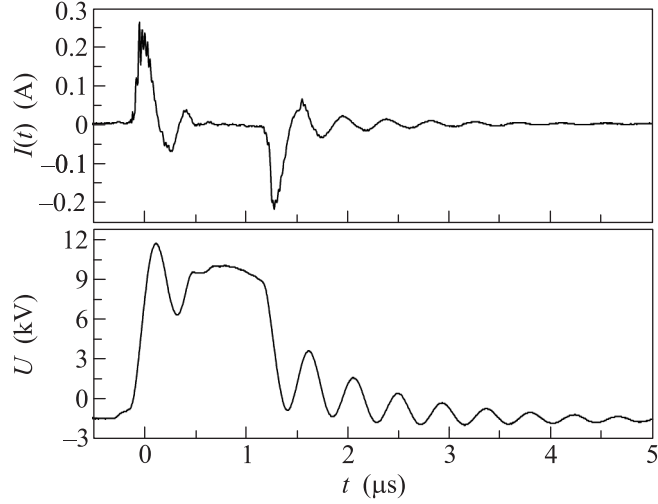


Figure 1: Waveforms of discharge current  $I(t)$  and voltage across the gap  $U(t)$  during formation of diffuse jets with pulser # 1

mode after discharge ignition it is required from 30 to 60 seconds. Typical diameter of the jet is no more than 0.5–1 mm (Fig. 2). At gas flow rate  $g = 0.51/\text{min}$ ,  $\tau = 1\text{--}1.5\ \mu\text{s}$ ,  $f = 45\text{--}85\ \text{kHz}$  and voltage pulse amplitude of

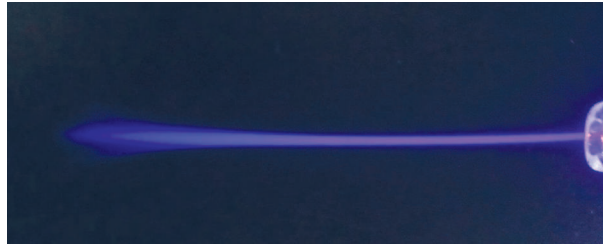


Figure 2: Shape of the plasma jet in the atmospheric pressure air. Pulse repetition rate  $f = 75\ \text{kHz}$ . Jet's length  $L \sim 3.2\ \text{cm}$ . Pulser # 1

up to 13 kV plasma jets with length of 4 cm (in the air) and 3 cm (in the nitrogen) were obtained. Under these conditions APPJ have following feature: at  $g < 0.51/\text{min}$  a jet formation does not occur. Increasing the gas flow rate to  $\sim 0.51/\text{min}$  leads to the formation of jets with the maximum length of 3 cm for nitrogen and 4 cm for air. Further increase in gas flow rate to 5–10 l/min leads to jet's length reduction. Thus, the optimal value  $g \sim 0.51/\text{min}$  is significantly less than those indicated in previous papers.

An emission spectrum of APPJ in the nitrogen and air in the wavelength range 280–450 nm was presented by bands of electronic-vibrational-rotational transitions ( $C^3\Pi_u \rightarrow B^3\Pi_g$ ) of the second positive system of a

nitrogen molecule with characteristic peaks having following wavelengths: 296.2 (3, 1), 315.9 (1, 0), 337.1 (0, 0), 353.7 (1, 2), 357.7 (0, 1), 375.5 (1, 3), 380.5 (0, 2), 399.8 (1, 4) and 405.9 (0, 3) nm (in parentheses vibrational numbers  $v'$  and  $v''$  corresponding to transitions are given). Besides, relative weak lines of transitions of the first negative system of molecular ion  $N_2^+$  ( $B^2\Sigma_u^+ \rightarrow X^2\Sigma_g$ ) at wavelengths 391.4 (0, 0) and 427.8 (0, 1), as well overlapping bands  $D^3\Sigma_u^+ \rightarrow B^3\Pi_g$   $A^3\Sigma_u^+ \rightarrow X^1\Sigma_g^+$  of molecule  $N_2^*$  in the wavelength range 200–280 nm were observed in the emission spectrum. In the air intensity of last mentioned bands was in 3–4 times less than in the nitrogen.

The measurement results of ultraviolet radiation irradiance of the plasma jet in the nitrogen and air in the axial direction at various distances have shown that under the same conditions radiation power of an APPJ in the air is about 2 times higher than in the nitrogen. Plasma jet's temperature linearly decreased for the both gases with increasing distance from the nozzle, and at a distance of 4 cm from the nozzle was about 150 °C.

**Diffuse “channels” in atmospheric pressure air during the corona discharge.** Integral images of glow of the corona discharge in the air ignited due to applying of voltage pulses with various parameters from three pulsers to a tip electrode are presented in Fig. 3. Corona discharge in Fig. 3a was ignited with pulser

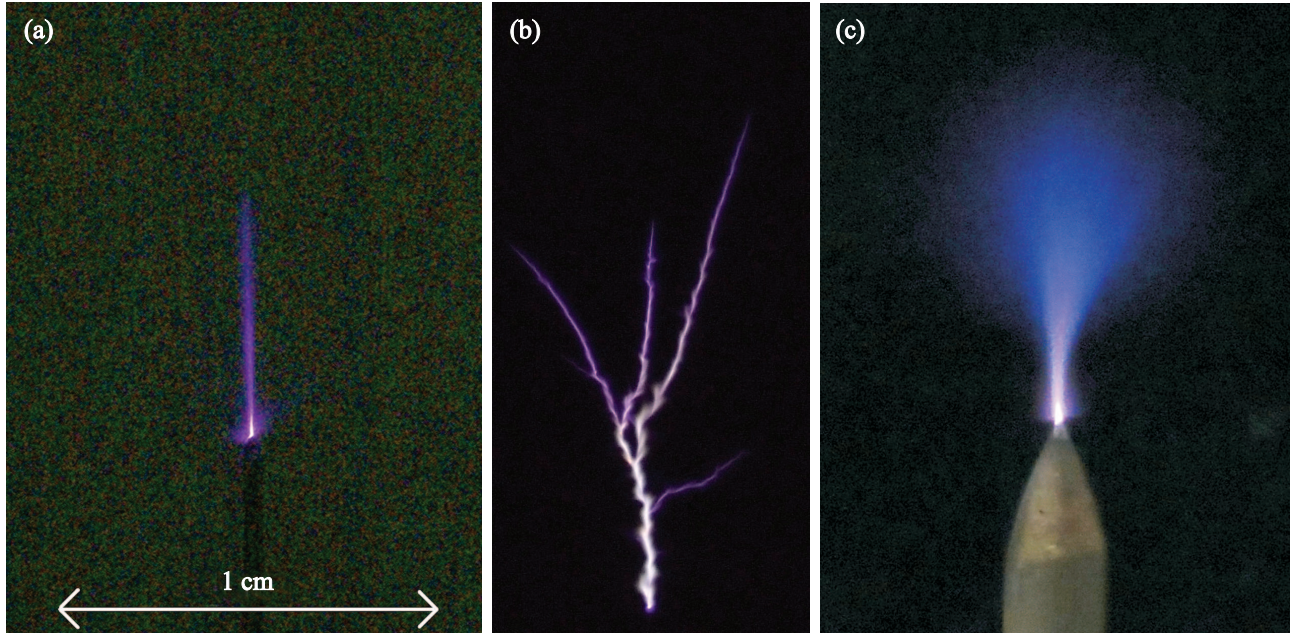


Figure 3: Integral images of glow of corona discharge in the atmospheric pressure air with using pulsers # 1 (a), # 2 (b) and # 3 (c). (a) – Stationary mode at 10 ms-duration burst of voltage pulses having different polarities and amplitude of 10 kV; pause between bursts is 10 ms. (b) – Image height is 40 cm. (c) – Image height is 3.5 cm

like the pulser # 1. This pulser formed voltage pulses with an amplitude of 5–10 kV, pulse repetition rate of 100 kHz and full width at half-maximum of  $\sim 1.5 \mu s$ . The polarity of the voltage pulses after every two pulses of the same polarity reversed. In this case, the burst mode with a duration of one burst of 10 ms and different pauses between bursts was used. High-voltage electrode was tip with diameter of 1 mm and radius of curvature of  $50 \mu m$ . By varying the duration of the individual bursts and delays between bursts, it has managed to form a corona discharge in the form of a thin diffuse “channel” (Fig. 3a). With the reduction of the pause between bursts two jets having larger diameter and smaller length formed instead of one.

Integral image of glow of the corona discharge with length of diffuse “channel” about 30 cm is presented in Fig. 3b. In this case, pulser # 2 based on Tesla transformer was used. Pulser formed high-voltage pulses

consisting of separate trains of duration of 10 ms and following with a frequency of 50 Hz. Every train consisted of a sinusoidal signal with oscillation frequency of 289 kHz. The largest difference between the positive and negative voltage peaks in the train reached 250 kV. High-voltage pulses were applied to 3.6 mm-diameter cylindrical electrode one end of which had a tip shape with radius of curvature of 0.2 mm. Corona discharge in the air of atmospheric pressure was formed at distance of 40 cm and more between potential and grounded electrodes. If interelectrode distance was less low-current spark discharge current value of which did not exceed 10 A occurred. Image in Fig. 3b was taken during one train of pulses (10 ms).

When applying one voltage pulse “channel” (diffuse jet) is formed. It grows from the electrode with a small radius of curvature and can be branched. At small distances from their front diameter of diffuse jets is less than 1 mm. Color and shape of diffuse jets with using pulser # 2 depended on their development time. During the voltage pulse risetime jets is colored blue, then their color can change. Initially at the tip, and then on the its entire length color of the jet, that in this phase of development can already be called a diffuse “channel” becomes white. When reducing the voltage during the fall of a modulated voltage pulse “channel” begins to bend. Diameter of diffuse “channels” at the base increases as their length. New diffuse jets (streamers) having essentially smaller length and brightness can develop from lateral surface of diffuse “channels” (Fig. 3b).

As for plasma jets, an emission spectrum of diffuse “channels” was represented mainly by radiation caused by the spectral transitions of the second positive system of a nitrogen molecule. However, emission spectrum obtained from near tip region (near potential electrode) contains not only bands of the second positive system of a nitrogen molecule, but also relatively intense lines emitted by the electrode material particles. Radiation intensity in the visible region of the spectrum is more than an order of magnitude smaller than the one in the UV region of the spectrum. As well, an important result at the ignition of the corona discharge is registration of the X-ray with film placed in an envelope having a beryllium foil window (Fig. 4). This fact indicates that

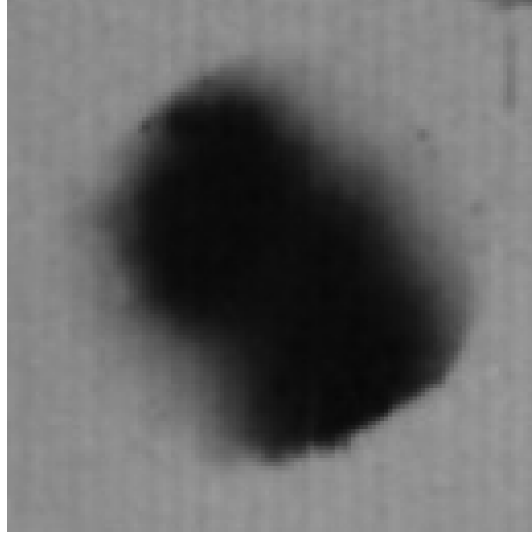


Figure 4: Darkening of the film due to the influence of X-ray emitted during the corona discharge. Exposure time is 11 min. Pulser # 2. The diameter of black colored area is 6 mm

runaway electrons which apparently affect the discharge form are generated during the atmospheric pressure corona discharges.

Integral image of glow of the corona discharge ignited with pulser # 3 (described in main article “Phenomenon of apokamp discharge”) is presented in Fig. 3c. Under the same pulser parameters a length of the expanding diffuse “channel” during the corona discharge was about 3 times less than during the apokamp discharge. In our opinion, this is significant difference between apokamp discharge and corona discharge.