Temporary-resolved measurement of electron density in small atmospheric plasmas

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A new method for temporally resolved measurements of absolute values of plasma density applicable for wide spectrum of small-size atmospheric plasmas and utilizing Rayleigh microwave scattering on the tested plasma object is proposed. The absolute electron density measurements in an atmospheric plasma jet revealed presence of two consecutive breakdowns during the half-wave of the discharge-driven high voltage. The ionization mechanisms of both breakdowns are considered. © 2010 American Institute of Physics. [doi:10.1063/1.3389496]

Recent progress in the creation and application of small size atmospheric plasma sources1 requires the development of appropriate tools for their diagnostics. Very promising and important applications of atmospheric plasmas refer to the new emerging fields of plasma medicine and nanotechnology.2,3 Traditional methods cannot be applied for the measurements of such small plasmas,4 and at the present time it is characterized mainly by fast cameras and spectroscopy,5,6 giving only a qualitative picture of processes involved. The Rayleigh microwave scattering (RMS) technique was first proposed theoretically by Schneider7 and then applied for temporally resolved measurements of laser-induced avalanche ionization in air and resonance-enhanced multiphoton ionization in argon (plasma size—tens of microns in diameter and few millimeters in length).8,9 In our recent work,10 we applied the RMS technique to resolve temporal dynamics of cold atmospheric plasma jets. However, no absolute measurements of plasma density were carried out. In this letter, we report on a new method for absolute plasma density measurements in small atmospheric plasma objects using the RMS technique (Fig. 1) and consider wide spectrum of miniature atmospheric plasmas where this approach can be applied. Absolute plasma density is measured in an atmospheric plasma jet [Fig. 1(a)] and mechanisms of ionization are discussed.

The experimental microwave system is schematically presented in Fig. 1(a). Two microwave horns with centers of exit sections located at (x, y, z)=(6 cm, 0, 0) and (0.15 cm,0) were used for radiation and detection of microwave signal [see Fig. 1(a)]. Microwave radiation was linearly polarized along the z-axis (12.6 GHz). More details on the experimental system can be found elsewhere.8,5 The detection of the scattered signal was accomplished using a homodyne scheme by means of I/Q Mixer, providing in-phase (I) and quadrature (Q) outputs. The amplifiers and the mixer used in the experimental setup operated in linear mode for the entire range of scattered signals. The amplitude of output signal U, referred to the mixer output signal, was determined as

\[
U = \sqrt{I^2 + Q^2}.
\]

Now let us consider the relation between the output signal \( U \) and physical properties of scatterer. According to Ref. 7, radiation from cylindrical scatterer oriented along the electric field of incident radiation (wave front is considered to be flat on the scatterer length) and having radius \( r_s \) much less than \( \lambda \), \( \delta \) (where \( \lambda \) is the microwave wavelength and \( \delta \) the skin-layer thickness) is similar to Rayleigh scattering of light and in the far field (at distances \( R \gg \lambda \) and scatterer size) can be considered as radiation from a point dipole. In this case the amplitude of scattered wave \( E_s \) can be written as follows:

\[
E_s \approx \frac{\sin \phi}{R} I_0 l_0 \omega.
\]

where \( \phi \) is the angle between the dipole vector and direction of observation, \( I_0 \) the electrical current excited in the scatterer, \( l \) the length of the scatterer, \( \omega \) the angular frequency of microwave radiation, and \( R \) the distance from the scatterer to

![FIG. 1. (Color online) (a) The schematics of RMS experimental setup. (b) Typical scattering signals measured by RMS setup induced by calibrator bullet flying over the microwave horns along z-axis [in-phase (I) and quadrature (Q) components, and total amplitude of output signal \( U \)]. Teflon bullet of 8.5 mm length and 3.2 mm diameter was used. (c) Typical image of plasma jet for \( U_{in} = 3.8 \) kV, He flow of 15 l/min.](image-url)
The current $I_0$ in Eq. (1) is the total current excited in the scatterer channel, including conductivity and polarization currents. For cylindrical scatterers oriented along $z$-axis with cross-section area $S$, the amplitude of the current induced in the scatterer channel is $I_0 = \sigma E_z S$ for conductive scatterers ($\sigma$ is the electrical conductivity and $E_z$ the incident microwave field amplitude at scatterer location), and $I_0 = \varepsilon_0 (\varepsilon - 1) \omega E_z S$ for dielectric scatterers ($\varepsilon$ and $\varepsilon_0$ are dielectric permittivity of scatterer material and vacuum, respectively). Note, the local reduction in electrical field of incident microwave radiation inside the scatterer channel is negligible since the skin layer thickness significantly exceeds the scatterer radius ($\delta \gg r_1$), and depolarization effects are small (due to smallness of depolarization factors of strongly prolate scatterers and relatively low plasma conductivity considered in this work).\(^1\)\(^1\)\(^2\)\(^1\)\(^1\)\(^2\)\(^1\)\(^1\)\(^2\) Thus, for the fixed level of radiated power, microwave frequency $\omega$, and horn positions, and taking into account that the elements used in the detection system were in linear range for the entire range of observed scattered signal amplitudes (i.e., $U \approx E_z$, where $U$ is the output signal from the detection system), Eq. (1) can be rewritten for the amplitude of output signal for two limiting cases of pure conductor and pure dielectric as follows:

$$U = \begin{cases} A \sigma V, & \text{for conductor} \\ A \varepsilon_0 (\varepsilon - 1) \omega V, & \text{for dielectric.} \end{cases}$$

(2)

The coefficient $A$ is determined by the geometry and parameters of the specific microwave system, and is a function of the scatterer’s position in the space $(x, y, z)$. Equation (2) indicates that the coefficient $A$ is the key parameter required for detection of the absolute values of scatterer constants (namely, $\sigma$ for conductors and $\varepsilon$ for dielectrics) from measured output signal of RMS system ($U$). In this work, the coefficient $A$ was determined using calibration on materials with known properties as described below.

Several dielectric materials were used as calibrators; teflon ($\varepsilon=2.1$), alumina ($\varepsilon=9.2$), polyethylene ($\varepsilon=2.25$), and quartz ($\varepsilon=3.8$).\(^1\)\(^3\)–\(^1\)\(^5\) Calibrator bullet lengths varied in the range of 0.4–5 cm and had diameters around 3 mm. The calibrator bullets were shot from the pneumatic gun aligned along with $z$-axis [see Fig. 1(a)], flew over the horns along $z$-axis and were collected in collector. The time moments when bullet lefted the gun exhaust (located at $z_e=7.3$ cm) and reached the collector entrance (located at $z_d=8.9$ cm) were detected by breaking the electrical circuits comprised by two pairs of wires attached correspondingly to the gun exhaust and collector entrance. The typical temporal evolutions of scattered signals are presented in Fig. 1(b). The moments of leaving the gun exhaust and reaching the collector are indicated by the arrows 1 and 2 in Fig. 1(b). Well-reproducible nonzero scattering signal was observed only during bullet flight over the horns and thus it refers to scattering from the calibrator bullet.

Let us consider below the scattering from calibrators located at $z=0$ [corresponds to the moment of time when amplitude of scattered signal was maximal—$t=0.15$ ms in Fig. 1(b)]. In agreement with Eq. (2), the amplitude of scattering signal $U$ increased proportionally to the calibrator length up to $L$ about 4 cm [see Fig. 2(a)]. The deviation from the linear dependence observed for long bullets with $L>4$ cm is caused by curvature of wave front on the length of long scatterer and decrease in electric field amplitude with distance from $x$-axis. The angular variations in scattered signal were small [see Fig. 2(b)], indicating that scattered pattern was close to isotropic in the plane $z=0$, which indicates that the bullet scattered the radiation in Rayleigh regime. The coefficient $A$ was determined from Eq. (2) using measured $U$, known $\varepsilon$ and $V$ of calibrator, and its data is summarized in the Table I. It is seen that deviations of the coefficient did not exceed 10%–15%. Based on this data the coefficient $A = 11 \text{ V} \Omega^{-1} \text{cm}^2$ was used for further analysis.

The calibrated RMS system was applied for measurements of plasma density in atmospheric plasma jet. A plasma jet was produced in the He flowing through the Pyrex syringe equipped with pair of high voltage electrodes [Fig. 1(c)], as reported elsewhere.\(^1\)\(^0\)\(^\text{a}\)\(^1\)\(^6\) The plasma jet was inserted in the space between microwave horns, directed along the path of calibrator bullet flight (along $z$-axis), and centered at $z=0$.

The scattering signal from the atmospheric plasma jet is presented in Fig. 3. Similar to results reported in our previous work,\(^1\)\(^0\) the microwave scattering signal consists of two consequent peaks (will be discussed below in more details). The angular distributions of amplitudes of both peaks were uniform within 25% accuracy as shown in Fig. 3. No systematic shift in the scattered radiation pattern toward the radiating horn as it appears at transition to Mie regime was observed. Thus, we conclude that scattering was close to a Rayleigh regime. Relatively big deviation from the mean value may be caused by nonideal geometrical alignment of the experimental system components.

The temporal evolutions of average plasma density are presented in Fig. 4 for two driven voltage amplitudes, 2.7 and 3.8 kV. Plasma density was obtained from the plasma conductivity by using the following expression: $\sigma = 2.82 \times 10^{-4} n_e \nu_m / (\omega^2 + \nu_m^2)$, $\Omega^{-1} \text{cm}^{-1}$,\(^1\)\(^7\) where $\nu_m$ is the frequency

<table>
<thead>
<tr>
<th>Material</th>
<th>$A$ (V $\Omega^{-1} \text{cm}^2$)</th>
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<tbody>
<tr>
<td>Teflon</td>
<td>11</td>
</tr>
<tr>
<td>Alumina</td>
<td>10.6</td>
</tr>
<tr>
<td>Polyethylene</td>
<td>12.2</td>
</tr>
<tr>
<td>Quartz</td>
<td>12.3</td>
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of electron-neutral collisions (in atmospheric conditions in helium we used $v_{ne} \approx 2 \times 10^9 \text{ cm}(\text{Torr})^{-1} \text{s}^{-1}$), $n_e$ the plasma density (in inverse cubic centimeters), and $\omega$ the angular frequency (in radians per second). The plasma conductivity, in turn, was expressed from Eq. (2) using $A = 11 \text{ V cm}^{-1} \text{ cm}^{-2}$, and the volume of the plasma column, which was determined from visual observations (diameter 0.3 mm and length increased with time in accordance with position of “plasma bullet” detected by intensified charge-coupled device (ICCD) camera according to Ref. 10).

It is seen in Fig. 4 that after discharge initiation the plasma density reaches about $(5-10) \times 10^{13} \text{ cm}^{-3}$ (which agrees well with that predicted for streamer head) and then decays with characteristic times of few microseconds governed by electron attachment. Note, measured plasma density corresponds to the value averaged along the entire plasma column rising with time from the syringe exit. Thus, initial plasma density values refer to that in the streamer head, while following evolution has significant contribution of the decaying plasma column remaining after streamer. The second peak of plasma density appears with a certain delay after decay of the first discharge (about 1 $\mu$s for $U_{HV}=2.7$ kV) and indicates the presence of the second breakdown event. This additional ionization of the channel (corresponds to the current pulse observed in the jet by Rogowski coil reported in our previous paper) can be provided by ongoing increase in voltage (and thus, the electric field) after streamer passage (see voltage waveform on Fig. 4), which leads to additional breakdown when threshold field is reached. Increase in high-voltage amplitude leads to faster achieving of critical breakdown field, which causes reducing of delay between first and second peaks observed experimentally at $U_{HV}$ increase (see Fig. 4). The ionization of ambient air molecules by long-living metastable Helium atoms (Penning ionization) remaining on the way of streamer passage can serve as a source of primary electrons initiating the avalanches for the second breakdown in contrast with photoinitiation mechanism in the case of initial streamer breakdown.17

In summary, a method for measurements of absolute value of plasma density in small atmospheric plasmas was proposed. The experimental microwave system was first calibrated with calibrator bullets having known physical properties, and than applied for the atmospheric plasma jet. Analysis of angular distribution of scattering from plasma jet indicates that plasma scatterer was in Rayleigh regime. Plasma density in the streamer head was about $(5-10) \times 10^{13} \text{ cm}^{-3}$. It is important to note that proposed here approach can be utilized in many areas of applied plasma physics, namely, for laser-induced ionization of air, atmospheric inductively coupled plasma (ICP) torches, rf microdischarges, and dielectric barrier discharges.

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