Results in Physics 16 (2020) 102928

Contents lists available at ScienceDirect

Results in Physics

journal homepage: www.elsevier.com/locate/rinp

Application of terahertz time-domain spectroscopy in atmospheric pressure plasma jet diagnosis

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ARTICLE INFO

Keywords:
Plasma diagnosis
Terahertz time-domain spectroscopy
Atmosphere pressure plasma jet
Electron density

ABSTRACT

The measurement of plasma density is of great significance in the study of magnetically constrained nuclear fusion and the transmission of electromagnetic waves in plasma. In this paper, the terahertz time-domain spectroscopy (THz-TDS) system is used to diagnose the plasma density of the atmospheric pressure plasma jet. The transmittance properties of the atmospheric pressure plasma jet are measured by a fiber-coupled transmission THz-TDS system. Then the electron density of the plasma jet is calculated based on the measurement results according to the relationship with the complex refractive index. The plasma jets with different electron densities are also investigated by varying the discharge voltages and the ionization gases. The terahertz transmittance spectra in measurement and calculation agree well. This study is of great significance for the applications of THz-TDS in high-accuracy, non-contact plasma diagnosis.

Introduction

Plasma is a kind of quasi-neutral gas composed of the charged particles and neutral particles that exhibits the collective behaviors. It has many unique physical and chemical properties. Atmospheric pressure plasma jet is a convenient and economical low-temperature plasma discharge technology, showing many unique advantages in sterilization, biological mutation breeding, auxiliary combustion, air purification and so on [1–4]. The most outstanding feature of atmospheric pressure plasma jet is that the plasma is not confined between the electrodes or in a containment enclosure. It makes atmospheric pressure plasma jet widely used in material treatment especially in biomedical applications because the size of the sample is free of the limitation of the containment [5,6]. Atmospheric pressure plasma jet is also extensively used in laboratory to study plasma characteristics due to its simplicity and stabilization. As one of the most important characteristics of plasma, the electron density is of great significance for studying the interactions of electromagnetic waves with plasma, the balance and stability of magnetic confinement plasma [7,8]. Several measurement methods of the electron density have been developed for different application conditions, including Langmuir probe method, Thomson scattering, optical emission spectroscopy and microwave interferometry [9–12]. However, these measurement methods still have several respective limits in the applications.

Terahertz time-domain spectroscopy (THz-TDS) technique is a coherent detection technique that directly measures the complex refractive index of samples by processing the amplitude attenuation and phase delay of terahertz wave with a high signal-to-noise ratio [13–18]. It is a good tool to measure the properties of plasma. Compared to Langmuir probe method, THz-TDS is a non-contact measurement that does not affect the plasma state. Compared to Thomson scattering method, THz- TDS has a higher signal-to-noise ratio and better stability and reliability. And compared to optical emission spectroscopy, THz-TDS is easier to process and supports a faster detection. A series of researches have proved the feasibility of terahertz time-domain spectroscopy for low pressure plasma diagnosis since 2003 [19–21]. In 2010, Kitahara et al. utilized THz-TDS to measure the electron density measurement of the low-pressure argon inductively coupled plasma [22]. They also proved that THz-TDS performs higher accuracy under high RF powers and high voltages than Langmuir probe method in 2011 [23]. In 2016, Jang et al. used a THz-TDS system with the air plasma THz source measure the high-density (10^{19}–10^{20} m^{-3}) inductively coupled plasma [24]. In 2017, Meier et al. improved the signal-to-noise ratio of THz-TDS by redesigning the antenna structure and increasing the frequency of the lock-in amplifier and demonstrated that THz-TDS performed in plasma diagnosis as well as microwave interferometry method [25]. However, the researches above mainly focus on the low pressure inductively coupled plasma whose characteristics are different

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https://doi.org/10.1016/j.rinp.2020.102928
Received 2 November 2019; Received in revised form 3 January 2020; Accepted 5 January 2020
Available online 09 January 2020

from the atmospheric pressure plasma jets. THz-TDS technology in atmospheric pressure plasma jets diagnose has rarely be reported.

In this paper, the electron density of atmospheric plasma jet is measured by a fiber-coupled THz-TDS system. The plasma jet is generated by helium or neon discharge in a quartz tube under atmospheric pressure. The refractive index is measured by a transmission THz-TDS system. And then the electron density is calculated based on the refractive index. Different discharge voltages are utilized to change the electron density of the plasma. It's shown that with the increase of the discharge voltage, the electron density increases linearly. Moreover, the plasma jet of helium performs lower electron density than neon with the same discharge voltage because of the higher ionization potential. The measurement results agree well with the theoretically calculation results. This work promises THz-TDS technology is an effective diagnostic method for plasma density.

Theoretical model

According to the optical principles, the propagation characteristics of THz wave in plasma has a significant correlation with the effective refractive index of plasma. It is necessary to consider the effect of the oscillation and collision of electrons with neutral particles in plasma. The thermal motion of the particles in plasma deviates the plasma from electrical neutrality. In order to maintain macroscopic neutrality, electrons and ions in plasma are driven by Coulomb force for free oscillation. The frequency of the oscillation is defined as the plasma frequency $\omega_p = \sqrt{\omega_{pi}^2 + \omega_{pe}^2}$, where $\omega_{pe} = \sqrt{\frac{n_e e^2}{\varepsilon_0 m_e}}$ and $\omega_{pi} = \sqrt{\frac{n_i e^2}{\varepsilon_0 m_i}}$ are the electron and ion oscillation frequencies, respectively, n_e and n_i are electron density and ion density, ε_0 is the vacuum permittivity, m_e and m_i are the electron mass and ion mass, e is the electron charge [26]. Ion oscillation frequency can be ignored, because the mass of ion is much larger than electron. So the plasma frequency can be expressed as

$$\omega_p \approx \omega_{pe} = \sqrt{\frac{n_e e^2}{\varepsilon_0 m_e}} \tag{2.1}$$

The collision between electrons and neutral particles is characterized by the collision frequency ν_{en}, which is determined by the factors as temperature, pressure and neutral particle density.

The permittivity of unmagnetized cold plasma is given by $\varepsilon = \varepsilon_0 \varepsilon_r$, the relative permittivity ε_r can be expressed as [27]

$$\varepsilon_r = 1 - \frac{\omega_p^2}{\omega^2 + \nu_{en}^2} - i\frac{\nu_{en}}{\omega}\frac{\omega_p^2}{\omega^2 + \nu_{en}^2} \tag{2.2}$$

where ω is the angular frequency of the incidental electromagnetic wave. The complex refractive index can be expressed as $n = \sqrt{\varepsilon_r} = n + i\kappa$, where n is the refractive index, κ is the extinction coefficient. So the electron density can be inferred from n measured from THz-TDS.

Transmission THz-TDS obtains the optical properties of sample by analyzing the amplitude attenuation and phase shift of the terahertz wave after transmitted through plasma. For a monochromatic plane wave with the frequency ω propagating along the z-direction, the time dependent terahertz field can be expressed as [28]

$$E(z, t) = E_0 exp\left\{i\left(\omega t + \frac{n\omega}{c}z\right)\right\} \tag{2.3}$$

where the separable time dependent term $E_0(t) = E_0 exp(i\omega t)$ represents the wave form before propagating through the sample. After propagating through the dielectric of thickness d, the terahertz field is modified by the absorption and dispersion, the frequency dependent field is given by

$$E(\omega) = E_0 exp\left\{i\frac{n(\omega)\omega}{c}d\right\}\hat{A}\cdot exp\left\{-\frac{\kappa(\omega)\omega}{c}d\right\} \tag{2.4}$$

Considering the Fresnel reflection situation, when a beam penetrates the interface between two media with different refractive indices n_1 and n_2, the transmittance coefficient at normal incidence is given by

$$T_{1\to 2} = \frac{2n_2}{n_1 + n_2} \tag{2.5}$$

Fresnel reflection occurs when terahertz wave enters and leaves the plasma. The total transmittance coefficient caused by Fresnel reflection is

$$T = \frac{4n(\omega)}{(n(\omega) + 1)^2} \tag{2.6}$$

So the terahertz field after plasma can be described as

$$E_{sam}(\omega) = E_0(\omega) T exp\left\{i\frac{n(\omega)\omega}{c}d\right\}$$
$$exp\left\{-\frac{\kappa(\omega)\omega}{c}d\right\} \tag{2.7}$$

A reference wave propagating through vacuum (n = 1, κ = 0) of the same length is given by

$$E_{ref}(\omega) = E_0(\omega) T exp\left\{i\omega\frac{d}{c}\right\} \tag{2.8}$$

By dividing sample and reference waves in Eqs. (2.7) and (2.8), we get an expression for the frequency dependent complex ratio

$$\frac{E_{sam}(\omega)}{E_{ref}(\omega)} = \frac{4n(\omega)}{(n(\omega) + 1)^2} exp\left\{i(n(\omega) - 1)\frac{\omega d}{c}\right\}\cdot exp\left\{-\frac{\kappa(\omega)\omega}{c}d\right\} \tag{2.9}$$

The refractive index can be directly extracted from the phase shift Φ in (2.9)

$$n = n(\omega) = 1 + \frac{\Phi c}{\omega d} \tag{2.10}$$

So the electron density of plasma can be calculated from the phase shift measured from THz-TDS.

The transmittance of the electromagnetic wave in plasma can be calculated based on the physical optics (PO) method. Derived from Maxwell's equations, the wave equations in plasma can be described as

$$\nabla^2\vec{E} + k^2\vec{E} = 0 \tag{2.11}$$

where k is the wave number in plasma, it can be expressed as

$$k = k_0\sqrt{\varepsilon_r} = \frac{\omega}{c}\sqrt{\varepsilon_r} \tag{2.12}$$

k_0 is the wave number in vacuum, c is the speed of light in vacuum, andω is the angular frequency of the incident wave. Considering the one-dimensional case: the electromagnetic wave propagates in the positive direction along the z-axis, the electric field is parallel to the y-axis direction, and the magnetic field is along the x-axis direction, the wave equation becomes

$$\frac{d^2E_y}{dz^2} + k^2E_y = 0 \tag{2.13}$$

Using Wenzel-Kramers-Brillouin (WKB) approximating $\frac{1}{k^2}\frac{dk}{dz} \ll 1$, which assumes that the plasma parameters change slowly enough with the position, the solution of the wave equation is [27]

$$E_y = E_{y0} exp\left(j\int_0^z k dz'\right) \tag{2.14}$$

After passing through the plasma with the thickness d, the energy becomes

$$I = I_0 exp\left(-2Im\left(\int_0^d k dz\right)\right) \tag{2.15}$$

where I and I_0 are the energies of the THz wave before and after passing through the plasma, respectively. So the transmittance can be expressed

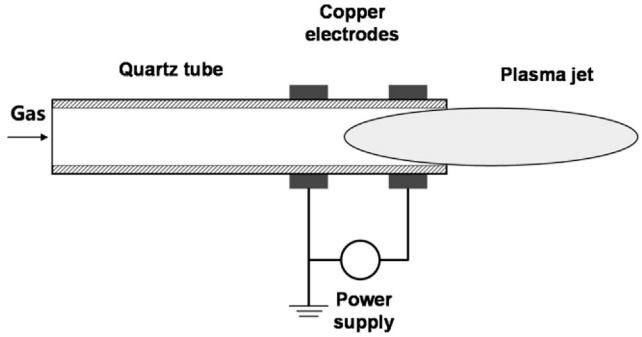

Fig. 1. Schematic diagram of the plasma source.

as

$$T = \frac{I}{I_0} = exp\left(-2Im\left(\int_0^d kdz\right)\right) \tag{2.16}$$

Experimental details

Atmosphere pressure plasma jet

Fig. 1 shows the schematic diagram of the plasma jet generation system. A high-voltage AC electric power is set to apply the voltage to the quartz tube with a 6-mm inner diameter and a 8-mm outer diameter. through the ring-shaped copper electrodes. Plasma is generated with the inert gas flow from the gas supply part. The electric power (CTP-2000K Suman Plasma Cop.) is tunable in frequency with the range of 5–20 kHz and in amplitude with the range of 0–30 kV. The end of the quartz tube is shaped into duckbill in order to broaden the plasma jet to cover the whole THz spot. The discharge part of the system is enclosed in a plastic cylinder wrapped with copper foils to prevent the detected THz signal from the disturbance of the high AC signal. The copper foils are grounded with a wire.

The discharge progress is maintained in dielectric barrier discharge (DBD) mode. In the experiment, the DBD plasma jet has the sizes of approximately 20 mm in length and 6 mm in width.

Terahertz time-domain spectroscopy

Fig. 2 shows the schematic diagram of the THz-TDS system (TAS 7500TS Advantest Cop.). Two femtosecond lasers (1550 nm, < 50 fs), serving as the pump laser and the probe laser, are utilized to excite the photoconductive antennas as the THz generator and detector. The maximal spectral range is from 0.1 THz to 4 THz. An asynchronous optical sampling system (ASOPS) enables high frequency resolution (3.8 GHz) and high-speed THz spectroscopy (16 ms per scan) instead of the classical mechanical delay line. High-bandwidth feedback electronics is utilized to maintain a small and constant repetition rate difference between pump laser and probe laser. The THz detector operates at different positions of different pulses to obtain the whole time-domain waveform. THz wave emitted from the antenna is collimated and focused on the plasma jet by a couple of 90° off-axis parabolic mirrors OAP1 (f = 101.6 mm) and OAP2 (f = 152.4 mm) and then collected to the detector by OAP3 (f = 152.4 mm) and OAP4 (f = 101.6 mm). The whole optical path is enclosed in an acrylic chamber and purges with dry air to reduce the absorption by the water vapor. In this experiment the time sampling is averaged by 1024 points of 262 ps duration.

Results and discussion

In our experiments, the flow rate is controlled to 3.5 L/min by the pressure reducer with a flow meter, in order to reduce the influence of the fluctuation of the jet on the measurement result. The whole system is introduced with the dry air and the humidity is controlled under 5%. Therefore, the effect of water vapor absorption is reduced to a relative lower level.

Fig. 3 shows the time-domain THz pulse signals with and without plasma. The THz beam passing through the plasma exhibits a phase advance, because the relative refractive index is less than 1. The existence of plasma cause a time advance about 0.02 ps. The frequency-domain power spectrum, as shown in the inset of Fig. 3, is obtained by Fourier transform from the time domain signals. And the maximal dynamic range reaches over 50 dB.

Electron density is calculated with the refractive index measured by THz-TDS. The plasma is inhomogeneous in both spatial and time domains but the resolution of THz-TDS is not high enough. So the electron density calculated here is the average electron density during the measurement process Fig. 4 shows the phase shift at different gas pressures and discharge voltages. It is obvious that the amount of the phase shift increases with increasing the voltage, which is caused by an increase in the relative refractive index due to an increase of the average electron density. Fig. 5 shows the average electron density calculated by the measured phase shift as the function of the discharge voltage. The calculated average electron density is in the order of 10^{19} m^{-3} which is in good agreement with the result in the other work [29]. With the increase of the discharge voltage from 1.7 kV to 2.7 kV, the average electron density increases linearly. The rise of the voltage causes that the more electrons in plasma become free electrons, so the gas ionization is increased. In addition, it should be noted that helium

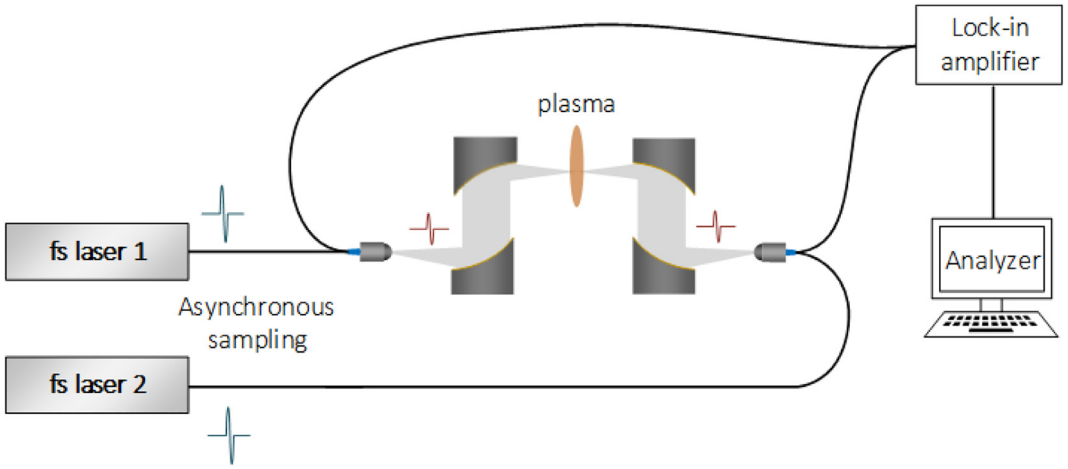

Fig. 2. Schematic diagram of fiber-coupled THz-TDS system.

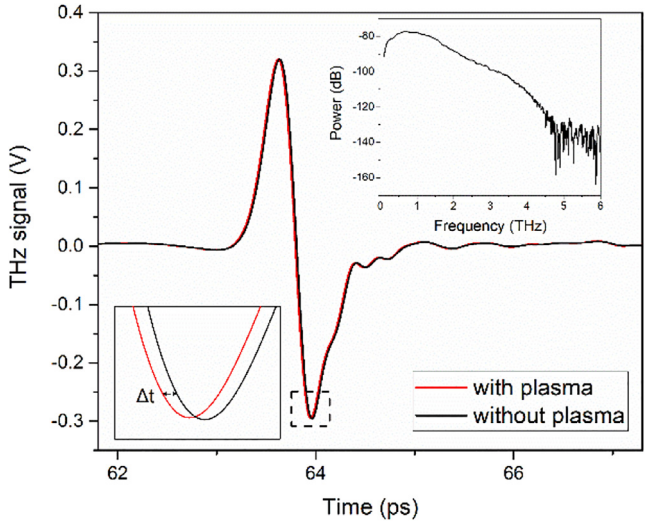

Fig. 3. Time domain spectrum of THz pulse with/without plasma. Power spectrum of is shown in the inset.

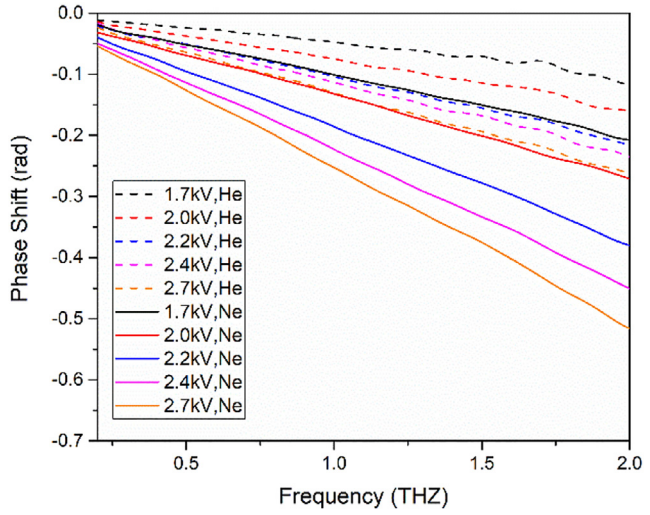

Fig. 4. Phase shift of THz pulses in He and Ne plasma with different voltages.

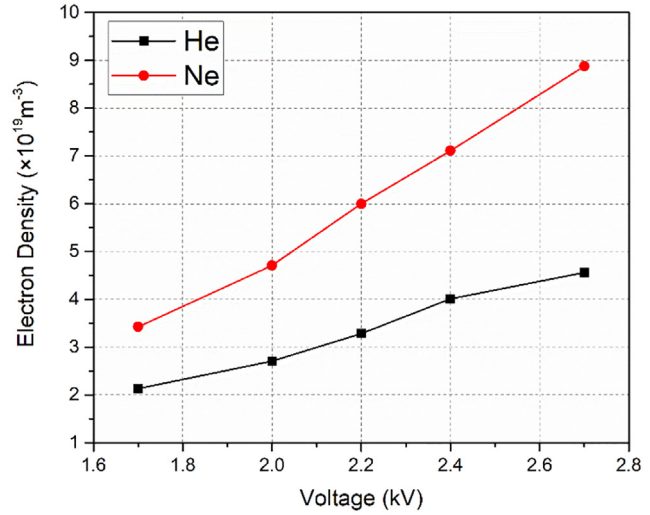

Fig. 5. Electron densities of He and Ne discharge plasmas as a function of discharge voltage.

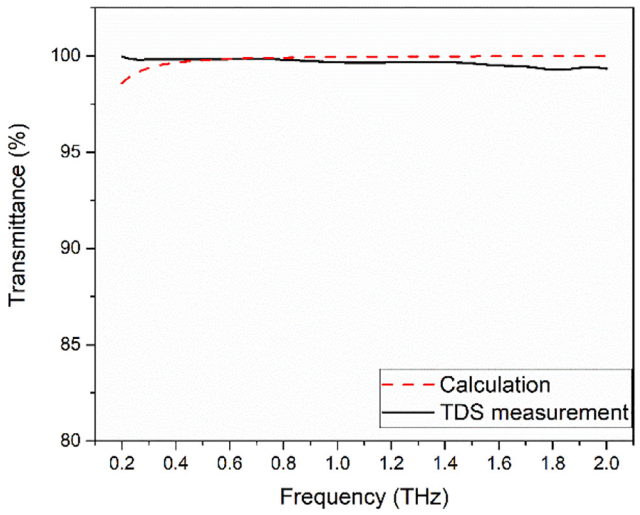

Fig. 6. Transmittance spectra of THz wave in plasma in calculation and TDS measurement.

(He) performs lower electron density than neon (Ne) with the same discharge voltage, because the ionization potential of helium is higher than neon. Therefore, more electrons in neon are likely to get enough energy to become free electrons. In our experiment, a bright neon plasma jet can be observed at a relatively low voltage.

The atmospheric pressure plasma jet in the experiment has a high transmittance for high-frequency THz waves. Fig. 6 shows the frequency-domain transmission spectrum of the He plasma with a discharge voltage of 2 kV. The red dotted line indicates the result of the simulation. The solid black line is the result measured by THz-TDS. The transmittance exceeds 99% in most frequency range. The plasma jet is actually inhomogeneous and approximately cylindrical, so the scattering to THz wave results in the difference between the curves of simulation and measurement. In addition, the high transmittance of THz wave in plasma makes it more sensitive to the power fluctuations of THz-TDS. So the difference of the curves in the low frequency range is mainly because of the decrease of SNR. We also observed the influence of the jet jitter in the experiment. Improving the stability of the jet will improve the accuracy.

Conclusion

In conclusion, we have measured the electron density of the plasma jet under atmospheric pressure by a fiber-coupled THz-TDS system. The plasma jet is generated by helium or neon discharge in a quartz tube. The complex refractive index is directly measured by THz-TDS. And then the electron density is calculated based on the measurement results. The plasma jets with different electron densities by varying the discharge voltages applied to the electrodes are also investigated by THz-TDS system. Results show that the electron density increases linearly with the increase of discharge voltage. The electron density is in the order of 10^{19} m^{-3} in the experiment. Moreover, helium performs lower electron density than neon with the same discharge voltage because of the higher ionization potential. The measurement and calculation results show that the transmittance of THz wave in the plasma jet exceeds 99%. This work promises that THz-TDS technology is an effective diagnostic method for the plasma density.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work is supported by the Equipment Pre-Research Foundation (No. 6140415010202) and the National Natural Science Foundation of China (No. 61705162).

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