Influence of Microwave Electric Field on Spatial and Time-Variation of Hβ Spectra in Pulsed-Microwave Atmospheric Pressure Plasma

T. Murase¹, A. Kamata¹, T. Ishijima², H. Toyoda¹,²

¹ Department of Electrical Engineering and Computer Science, Nagoya University, Nagoya, 464-8603, Japan
² Plasma Nanotechnology Research Center, Nagoya University, Nagoya, 464-8603, Japan

Atmospheric pressure plasma produced by a slot antenna with a pulsed microwave excitation was investigated to understand an electric field influence on hydrogen Balmer-β (Hβ) spectral profile using space- and time-resolved optical emission spectroscopy. Parallel- and perpendicular-polarized Hβ emission line profiles with respect to the microwave electric field direction were measured by a polarization spectroscopy technique. Clear differences of Hβ spectral profiles were observed at the early stage of plasma ignition, especially in the vicinity of the slot antenna, revealing the influence of the microwave electric field on Hβ line profiles.

1. Introduction

Atmospheric pressure nonequilibrium plasmas such as corona discharges[1], dielectric barrier discharges (DBD)[2], plasma jet[3,4] have been given much attention due to its high chemical reactivity with low process temperatures in the atmosphere, and are widely used in various application areas such as materials processing, decomposition of harmful substances, and sterilization. Generally, to produce nonequilibrium plasma, pulsed high voltage is used to suppress the increase both in the gas temperature and the electrode temperature, which induces arc discharge transition.

For better understanding of chemical reactions, detailed knowledge of both temporal dependence of plasma parameters and its spatial variation are necessary. In such purpose, diagnostics of pulsed atmospheric pressure plasma are indispensable to optimize the performance for their applications. So far, optical emission spectroscopy (OES) is known to be a powerful, intrusive, and a cost-effective technique. As one of OES measurement techniques, electron density, one of typical and important plasma parameters, has been evaluated from hydrogen Balmer-β (Hβ) line width [2, 5-11], except for very high-density (>10²³ m⁻³) plasmas where Hβ spectrum shows asymmetry profile or central dip profile [11]. This measurement is based on Stark broadening of the Hβ line width due to micro electric fields in the vicinity of excited H atoms, and relationship between the line width and the plasma density has been discussed. In some discharge conditions, however, there is a possibility that the line profile is influenced not only by the plasma density but also by externally-applied electric fields for the discharge. From this idea, line splitting of Hβ and evaluation of electric fields in low pressure discharges [12-14] and in atmospheric pressure discharges [8, 15] have been reported with use of a polarization measurement technique [16]. This technique utilizes the difference of the Stark splitting between parallel and perpendicular polarized Hβ line profiles with respect to the electric field and is very sensitive to detect electric field.

Based on this technique, we have investigated Hβ line profiles in a pulsed-microwave atmospheric pressure plasma and have demonstrated that Hβ line profile is influenced by time-varying microwave electric fields [17]. In this work, clear difference in the line width has been observed between differently-polarized Hβ emissions, indicating that the line width is influenced by the electric field. In this study, time-resolved OES with a high spatial resolution is applied to investigate spatial variations of the electric field in the pulsed-microwave atmospheric pressure plasma.

2. Experimental Apparatus

Figure 1 shows schematic of experimental apparatus. Ar and small amount of H₂ are introduced into a waveguide line through independent mass flow controllers at fixed flow rates of 1000 sccm and 16...
sccm, respectively, under open-air condition. A nonequilibrium atmospheric pressure microplasma is produced inside a slot antenna (0.6 mm in gap distance) at the end of the waveguide line, using pulsed 2.45 GHz microwave power (pulse width: 70 μs, repetition frequency: 10 kHz, peak power: 150 W). A water cooling system is installed on the slot antenna plate to remove heat from the plasma. Emission image of the plasma is focused on an entrance slit of an optical multi-channel analyzer (OMA) through a relay lens of 4 cm diameter and a microscope. To obtain the spatial variation of the emission within the gap of the slot antenna, the entrance slit of the OMA is arranged so as to be perpendicular to lateral direction of the slot antenna. To evaluate a microwave electric field from Hβ spectral profile measurement, a polarizing filter is placed in front of the entrance slit. Here, direction of the polarization is defined as “parallel” and “perpendicular” with respect to the direction of the microwave electric field generated across the slot antenna gap, as shown in Fig.1. The OMA has three gratings (150, 1800, and 3600 lines/mm) and a CCD detector (576x384 pixels, 22.5x22.5 μm) with an image intensifier. The CCD is cooled by a Peltier cooling system and is purged by dry air to prevent frost on the detector. To synchronize the exposure timing of the image intensifier with the desired measurement time of the plasma production, both the microwave generation and the image intensifier are controlled by a pulse generator. Figure 2 shows schematic diagram of the microwave discharge operation and the timing pulses for the OES measurement. In this study, \( T=0 \) is defined as the time of the plasma ignition by the microwave. Definition of the measurement time \( (T) \) is also indicated in Fig.2. The gate width of the image intensifier \( (T_w) \) is 0.1 μs.

3. Results and Discussions

Before the space-resolved measurement of the microplasma, optical alignment of the system is optimized. For this purpose, image of the plasma is projected on the CCD using zero\(^{th} \) order light of the grating and positions of a microscope objective lens and the relay lens are adjusted to obtain clear image on the CCD. Figure 3 shows an example of the optimized CCD image. Here, red and white broken lines indicate the edge of the monochromator entrance slit and the edge of the slot antenna, respectively, as a guide of eyes. From this measurement, spatial resolution of the system is obtained to be 3 μm/pixel. Hereafter, \( x \) axis is defined as the direction across the slot antenna with the origin at the center of the slot gap.

At first, total emission intensity profile from the plasma is observed along \( x \) axis. In this measurement, zero\(^{th} \) order images for 50 pulsed discharges are accumulated and then the data along the CCD horizontal direction are again accumulated. Figure 4 shows the optical emission profile along \( x \) axis at \( T=0.25 \) and 1.05 μs, respectively. The electrode edge is located at \( x=±0.3 \) mm. Irrespective of the
measurement time, strong emission intensity from the plasma is observed in the vicinity of electrode edges, while the emission intensity is much lower in the bulk region. Furthermore, decrease in the emission intensity with time is faster for the bulk region compared with that for the edge region.

Figure 5 shows temporal variations of the total emission intensities in the vicinity of the electrode (-0.30 <x< -0.22 mm) and the center (-0.04<x< 0.04 mm). Strong emission intensity is observed at the edge of the slot antenna, especially at T=0.15 μs. With increasing the measurement time, however, intensities both for the center and the edge rapidly decrease within 0.3 μs and slowly decrease as increasing the discharge time. The results of Figs. 4 and 5 suggest that the excitation rate is higher in the vicinity of the electrode and at the early stage of the discharge ignition, and also suggest that the strong electric field exists in the vicinity of the electrode as the origin of high excitation rate.

However, the excitation rate is generally determined by ground state particle density such as Ar or H₂ density and electron density, as well as the excitation rate coefficient. In atmospheric pressure plasma, it is known that the gas temperature has spatial variation due to short thermal diffusion length compared with the characteristic length of the electrode. To give an insight into the spatial variation of the gas density, spatial variation of the rotational gas temperature (Tᵣ) is measured from N₂ band spectrum, supposing that Tᵣ is close to the translational gas temperature (Tᵣ') due to fast energy exchange collisions between rotational and translational ones. Figure 6 shows optical emission spectra of N₂ second positive (C²Πₓ → B₂Πₓ) at T=10.5 μs for x=0 and ±0.25 mm with a spatial resolution of 0.1 mm. The above transition is chosen because the interference of background emissions (Ar, H, OH, CO and CN from air impurities) on the measurement is smaller than other transition lines. It is known that the emission band tail becomes longer with the increase in Tᵣ. From the experimentally-obtained band spectra, Tᵣ and (Tᵣ') was obtained by fitting simulated N₂ band spectrum with the observed one and finding the best-fitted Tᵣ to minimize χ². From the results in Fig. 6, gas temperatures at x=0 and ±0.25 mm are evaluated to be 2600 and 1300 K, respectively. The result indicates that the gas temperature is lower in the vicinity of the electrode due to heat diffusion to the electrodes and that the gas density is higher in the vicinity of the electrodes. From this result, it is concluded that the variation of the gas density is not the origin of the spatial variation of the emission intensity.

Finally, temporal variations of H₉ spectral profiles are measured at different positions, i.e., in the vicinity of the electrodes and at the center of the slot, using the polarization spectroscopy method. From measured H₉ line profiles, deformation of the profile like central dip [9-11] was not observed. Figure 7 shows time-resolved full-width at half-maximum (FWHM) line widths of H₉ spectra for parallel (Δλ∥) and perpendicular (Δλ⊥) polarizations. Difference between Δλ∥ and Δλ⊥ is observed at T<1 μs, indicating influence of strong electric field on H₉ spectral profiles at the initial stage of the microwave plasma ignition. It should be also pointed out that Δλ∥-Δλ⊥ at T=0.15 μs is larger in the vicinity of the electrode than that at the center, indicating that the microwave electric field intensity in the vicinity of the slot antenna is higher than that in the bulk region.
To evaluate the microwave electric field intensity from the measured line profile, following procedure is proposed. In general, H₀ profile is considered as convolution of Gaussean and Lorenzian profiles, (Vorgt profile). When the influence of time-varying the electric field such as microwave electric field is taken into account, the spectrum profile becomes broader. To obtain the contribution of the electric field, Δλ₀, and Δλ₁, for various combinations of Lorenzian, Gaussean FWHMs and time-varying electric field intensities are calculated [17] and tabulated. Next, measured Δλ₀, Δλ₁ and Gaussean line widths are compared with the calculated table and both electric field and Lorenzian FWHMs are obtained. From this procedure, an electric field intensity of 0.8 × 10⁷ V/m is obtained at T= 0.25 μs in the vicinity of the electrode.

4. Summary

Space- and time-resolved OES technique with the microscope was employed to investigate pulsed-microwave atmospheric pressure plasma excited in a slot antenna. Influence of the microwave electric field across the slot antenna gap was discussed from difference of H₀ spectral line widths between parallel and perpendicular polarized emission with respect to the microwave electric field direction. From the space-resolved measurement, strong electric field was observed in the vicinity of the slot antenna. From the time-resolved measurement, monotonic decrease in the electric field with the time evolution after the plasma ignition was observed.

Acknowledgement

This work was supported by Grant-in-Aid for Scientific Research (C) 21540509 by MEXT.

5. References