Dramatic Enhancement of OH($A^2\Sigma^+$) Density in a Recombining Hydrogen Plasma

Kanji SHIBAGAKI, Renge ASAKAWA, Daiyu HAYASHI[1)] and Koichi SASAKI[2)]

Department of Electrical and Electronic Engineering, Suzuka National College of Technology, Suzuka, Mie 510-0294, Japan

[1)]*Philips Technology Research Laboratories, Weisshausstr. 2, 52066 Aachen, Germany*

[2)]*Plasma Nanotechnology Research Center, Nagoya University, Nagoya 464-8603, Japan*

(Received 21 June 2008 / Accepted 19 August 2008)

Optical emission spectra in the wavelength range of molecular hydrogen were observed in both ionizing and recombining plasma modes of hydrogen discharges. The optical emission spectrum from the ionizing plasma was dominated by the Fulcher-α system ($a^3\Sigma_g^+ - d^3\Pi_u^-$) of molecular hydrogen. On the other hand, the optical emission spectrum from the recombining plasma was composed of many lines, and was completely different from the spectrum of the ionizing plasma. The many peaks observed from the recombining plasma were assigned to the $X^2\Pi - A^2\Sigma^+$ transition of OH. The OH($A^2\Sigma^+$) is thought to originate from the chemical etching of a glass discharge tube by atomic hydrogen. This is analogous to the interaction between a detached divertor plasma and a graphite plate, and a possible process for the enhancement of OH($A^2\Sigma^+$) density in a recombining plasma is discussed with this analogy in mind.

Keywords: optical emission spectroscopy, molecular spectroscopy, recombining plasma

DOI: 10.1585/pfr.3.055

Recombining plasmas are important in nuclear fusion research since they are closely related to plasma detachment, which is considered to be a promising approach for avoiding excess heat flux to the divertor plate in a nuclear fusion machine [1]. It is well known that in a recombining plasma, highly excited states are populated significantly via three-body recombination, and the optical emission spectrum is considerably different from that of an ionizing plasma [2]. In the case of a recombining hydrogen plasma, the optical emission spectrum of atomic hydrogen has been investigated well, however, the optical emission spectrum in the wavelength region of molecular hydrogen has not been investigated thoroughly. Therefore, in this study, we examine the optical emission spectrum in the wavelength region of molecular hydrogen in a recombining hydrogen plasma.

The plasma source [3] was a linear machine with a uniform magnetic field of 350 G along the cylindrical axis. An rf power source at 13.56 MHz was connected to a helical antenna wound around a glass tube with an inner diameter of 1.6 cm. The glass tube was attached to a stainless steel vacuum chamber. A plasma column with the diameter same as the glass tube was confined radially by the external magnetic field. We used pure hydrogen for discharge in this experiment. The plasma was produced in a pulsed mode with a repetition frequency of 10 Hz and a discharge duration of 4 ms to avoid over-heating of the machine. The diagnostic method was conventional optical emission spectroscopy, which was performed in the downstream region at a distance of 22 cm from the helical antenna using a spectrograph with a focal length of 50 cm. The spectrum was recorded using a charge-coupled device camera with a gated image intensifier (ICCD camera). The gate of the ICCD camera was opened from 3.95 to 3.99 ms after the initiation of the pulsed discharge.

Two types of discharge were observed when the gas pressure was higher than 30 mTorr. Low rf power resulted in a low-density mode discharge with an electron density of the order of 10^{11} cm^{-3}. A high-density mode discharge with an electron density of the order of 10^{12} cm^{-3} was obtained at rf powers higher than ~1.5 kW [3]. On the other hand, when the gas pressure was lower than 30 mTorr, a low-density mode discharge was obtained even at a high rf power of 3 kW.

In the high-density mode discharges at gas pressures lower than 50 mTorr and all the low-density mode discharges, the observed color of the plasmas was pink. In this case, the optical emission spectrum of atomic hydrogen was dominated by H_α emission. On the other hand, we observed blue plasmas in the observation region by adjusting the gas pressure to be higher than 56 mTorr in the high-density mode. Figure 1 shows an optical emission spectrum observed at an rf power of 2.5 kW and a gas pressure of 63 mTorr (high-density mode). The lines shown in Fig. 1 are assigned to the Balmer series of atomic

author's e-mail: sasaki@nuee.nagoya-u.ac.jp

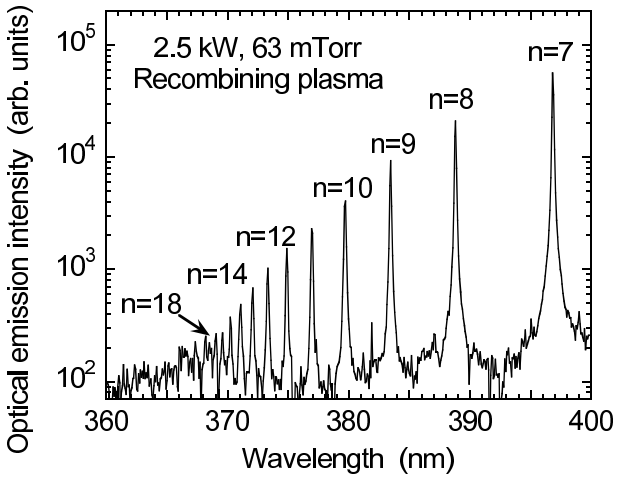

Fig. 1 Optical emission spectrum of atomic hydrogen observed at an rf power of 2.5 kW and a gas pressure of 63 mTorr (high-density mode).

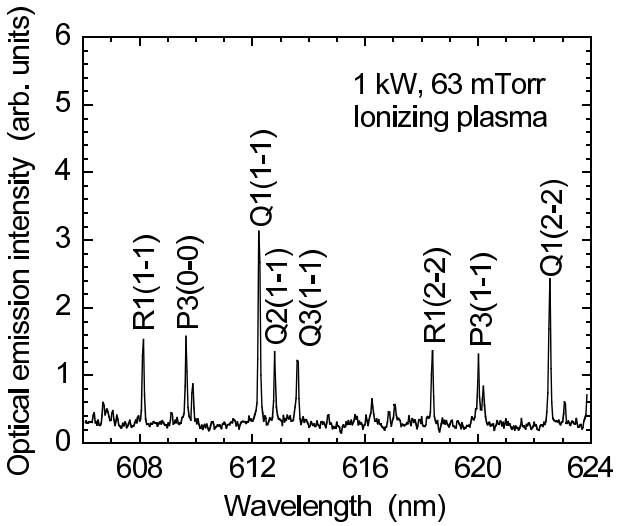

Fig. 2 Optical emission spectrum observed at an rf power of 1 kW and a gas pressure of 63 mTorr (low-density mode).

hydrogen. We detected optical emission intensities from highly excited states having principal quantum numbers up to $n = 18$. The population distribution of excited states deduced from Fig. 1 agreed well with the Saha-Boltzmann equation, indicating that the plasma was a recombining plasma at local thermodynamic equilibrium. The electron density and temperature of the recombining plasma were determined to be $3.2 \times 10^{12}\,\mathrm{cm}^{-3}$ and 0.1 eV, respectively, by data fitting using the Saha-Boltzmann equation [4]. The pink plasmas with intense H_α emissions, which were observed at the low-density mode and low gas pressures in the high-density mode, were ionizing plasmas. The electron temperature of the ionizing plasma measured using a Langmuir probe was ~4 eV.

We shifted the wavelength range of the spectrograph to the Fulcher-α system of molecular hydrogen. Figure 2 shows an optical emission spectrum observed at an rf power of 1 kW and a gas pressure of 63 mTorr (low density mode). The several lines shown in Fig. 2 are assigned perfectly to the transitions of the Fulcher-α system ($a^3\Sigma_g^+ - d^3\Pi_u^-$) [5].

Figure 3 shows an optical emission spectrum in this wavelength range observed from the same plasma as that shown in Fig. 1. It is clear that the spectrum shown in Fig. 3 is completely different from Fig. 2. Some of the lines shown in Fig. 3 are assigned to the transitions of the Fulcher-α system, but the intensities of the Fulcher-α lines shown in Fig. 3 are weaker than those shown in Fig. 2. In other words, the intensities of the Fulcher-α lines in the ionizing plasma produced at 1 kW were stronger than those in the recombining plasma produced at 2.5 kW. It should be emphasized here that the intensities of lines other than Fulcher-α are negligible in ionizing plasmas.

Many of the peaks shown in Fig. 3 are assigned to the $X^2\Pi$ ($v'' = 0$) – $A^2\Sigma^+$ ($v' = 0$) transition of OH (the

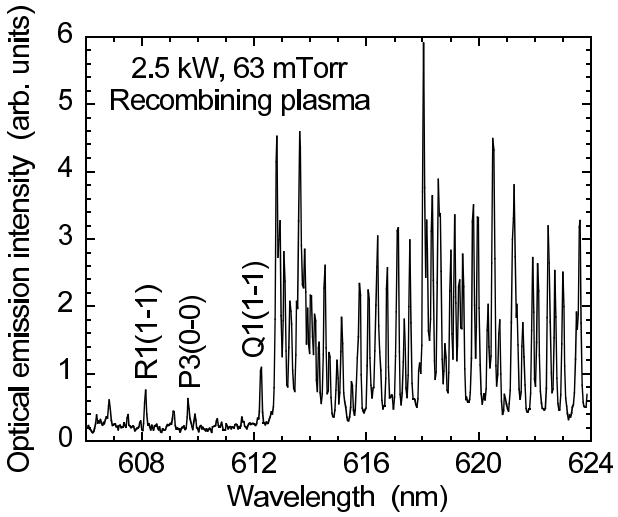

Fig. 3 Optical emission spectrum observed at an rf power of 2.5 kW and a gas pressure of 63 mTorr (high-density mode).

second-order diffraction), even though the peaks are superimposed on a continuum component. The production of H_2O, due to chemical etching of the glass tube surface by H atoms ($SiO_2 + 8H \rightarrow 2H_2O + SiH_4$), is thought to be the most likely origin of OH. Experimental results indicate that the density of $OH(A^2\Sigma^+)$ is enhanced dramatically in a recombining plasma. This can be explained by considering two-step process: $H_2O + e \rightarrow H_2O^+ + 2e$, then, $H_2O^+ + e \rightarrow OH(A^2\Sigma^+) + H$ [6]. The first step is efficient in the upstream ionizing plasma, while the second step has a large rate coefficient in a recombining plasma with a low electron temperature. Another possibility is the desorption of residual H_2O from the surfaces of the stainless steel chamber and the glass tube; but chemical etching of the

glass tube is a more probable source of H_2O, considering the temporal stability of the optical emission intensity of OH. The spectrum of OH shown in Fig. 3 was reproduced by spectral simulation with an assumed rotational temperature of 4200 K. This rotational temperature is higher than the translational temperature of H atoms (~2000 K in the case of an ionizing plasma) in the same plasma source [7], and the difference could be explained by a higher temperature of H_2O^+ than H, and by the exothermic nature of the production process of $OH(A^2\Sigma^+)$.

Chemical etching of the glass tube by H atoms is a close analogy for the interaction between a graphite divertor plate and a divertor plasma, where H atoms chemically etch the graphite plate to produce CH_x [8]. Therefore, the dramatic increase in the $OH(A^2\Sigma^+)$ density observed in the present work may suggest enhancement of the optical emission intensity of CH_x in a detached divertor plasma interacting with a graphite plate. In addition, there is a possibility that optical emissions of molecular hydrogen due to transitions other than the Fulcher-α system would be observed in a recombining plasma, since the electronic excited states are produced via recombination processes. Further investigation of recombining hydrogen plasmas with molecular spectroscopy is an important subject for future work.

This study was performed with the support and under the auspices of the NIFS Collaboration Research Program (NIFS07KCHP010).

[1] S. Takamura, N. Ohno, D. Nishijima and Y. Uesugi, Plasma Sources Sci. Technol. **11**, A42 (2002).
[2] T. Fujimoto, *Plasma Spectroscopy* (Oxford Science Publications, 2004).
[3] M. Aramaki, K. Kato, M. Goto, S. Muto, S. Morita and K. Sasaki, Jpn. J. Appl. Phys. **43**, 1164 (2004).
[4] K. Shibagaki and K. Sasaki, J. Phys. D: Appl. Phys. **41**, 195204 (2008).
[5] Z. Qing, D.K. Otorbaev, G.J.H. Brussaard, M.C.M. van de Sanden and D.C. Schram, J. Appl. Phys. **80**, 1312 (1996).
[6] B.R. Rowe, F. Vallée, J.L. Queffelec, J.C. Gomet and M. Morlais, J. Chem. Phys. **88**, 845 (1987).
[7] M. Aramaki, Y. Okumura, M. Goto, S. Muto, S. Morita and K. Sasaki, Jpn. J. Appl. Phys. **44**, 6759 (2005).
[8] K. Sasaki, T. Maeda, N. Takada, M. Aramaki, M. Goto, S. Muto and S. Morita, Jpn. J. Appl. Phys. **44**, 7614 (2005).