Nitrogen Atom Density Measurements in NAGDIS-T Using Vacuum Ultraviolet Absorption Spectroscopy

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For the development of a high nitrogen atom density source, we produced nitrogen plasmas and measured the nitrogen atom density using vacuum ultraviolet absorption spectroscopy (VUVAS) at high neutral gas pressure (>1.5 mTorr) and discharge power (>500 W) in the NAGDIS-T, which could generate spiral-shaped dissociative recombining plasmas. It was seen that the nitrogen atom density increases with increasing neutral gas pressure and discharge power, and it reached 6.2×10^{17} m^{-3}. In the low gas pressure case, the estimated atom density was confirmed to be consistent with that measured by the actinometry method.

Keywords: nitrogen atom, plasma, NAGDIS-T, vacuum ultraviolet absorption spectroscopy, actinometry

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Nitrogen atoms (N) are expected to be used in various applications such as $\mathrm{NO_x}$ removal [1], material nitriding, and the molecular beam epitaxy method [2]. A high nitrogen atom density (n_N) environment is required; however, it is difficult to achieve due to the high binding energy of nitrogen molecules (8.80 eV) [3]. For the development of a high n_N source, a torus-type plasma generator NAGDIS-T [4] is used in this study. This device aims to create a high n_N environment through dissociative recombination by generating a spiral plasma with a long connection length of magnetic field lines, which realized high-density nitrogen molecule plasma (N_2^+) generation in the upstream region and its temperature reduction before reaching the wall [5].

In previous studies using the NAGDIS-T, n_N in the ground state was measured spectroscopically by assuming coronal equilibrium [5] and by actinometry [6]. As a result, it was confirmed that n_N was on the order of 10^{17} m^{-3}. In this study, we performed vacuum ultraviolet absorption spectroscopy (VUVAS) to measure n_N in a more direct manner. In Ref. [6], VUVAS was used to compare with actinometry under limited experimental conditions; the present study extends the conditions to higher neutral pressure, p_n, and higher discharge current, I_d, to generate a highern_N.

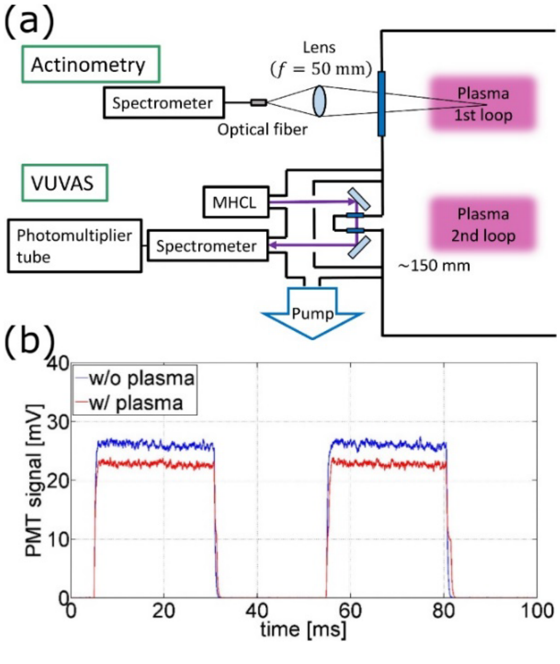

Fig. 1 (a) Schematic diagram of VUVAS and actinometry measurement systems. (b) PMT signals without (blue) and with (red) the nitrogen plasma discharge.

Figure 1 (a) shows a schematic diagram of VUVAS and actinometry measurement systems in the NAGDIS-T. A vertical and toroidal field of 2.7 and 40 mT, respectively, are applied to create a spiral plasma that is generated from the top inside the vacuum vessel. Because the plasma rotates twice in the toroidal direction under this condition, we call them the 1st and 2nd loops from the top. The VUVAS measurement is performed near the 2nd loop of the plasma (3.75 m from the cathode along the magnetic field). A micro-discharge hollow cathode lamp (MHCL), which uses helium gas with a mixture of small amounts of

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O_2, N_2, and H_2 gases for the discharge, is used for the light source [7], and the transmitted light is detected by a VUV spectrometer (NU-Rei Co. Ltd.) and a photomultiplier tube (PMT). VUVAS can detect N atoms that reach around the 2nd loop of the plasma, while the actinometry measures the 1st loop of the plasma. The light emission at the center of the plasma is collected through a lens and transferred to a spectrometer via an optical fiber.

Figure 1 (b) shows typical time evolutions of the transmitted light intensities at 120.0 - 120.1 nm, which corresponds to an N atomic transition, with and without the nitrogen plasma. Here, background nitrogen gas pressures are the same. The light emission from the MHCL is generated in pulses. It is confirmed that the PMT signal decreases when the N plasma is generated, indicating that there are N atoms absorbing the light emission from the MHCL. In addition, we confirmed that no light absorption occurred for oxygen (130.2 nm) and hydrogen (121.6 nm) emissions. For actinometry, 1% Ar is introduced into the nitrogen plasma, and an N emission line at $\lambda = 746.8$ nm and an Ar emission line at $\lambda = 750.4$ nm are measured. Then, n_N is calculated from the ratio of the line emission intensities [6].

Figure 2 (a) shows p_n dependences of n_N measured with VUVAS at $I_d = 5$ and 10 A and actinometry at $I_d = 5$ A. In the VUVAS measurement, the same assumptions were made as in Ref. [8]; i.e., the gas temperatures used in the measured medium and the MHCL were 300 and 400 K, respectively. Here, it should be noted that VUVAS and actinometry measurements at $I_d = 5$ A were not done simultaneously. Although these measurements were performed with the same discharge current, the discharge voltage was not the same, mainly due to some differences in the surface conditions on discharge electrodes and the wall in NAGDIS-T. Figure 2 (b) shows discharge powers (W_d) as a function of p_n in corresponding three discharge series. It is seen that W_d decreases as p_n increases, because the discharge voltage decreases with p_n. Moreover, W_d in the actinometry measuring discharges at low p_n condition are slightly higher than those during the VUVAS measurement.

In Fig. 2 (a), it is seen that n_N from VUVAS increases with p_n. Also, the n_N at $I_d = 10$ A are roughly two times higher than those at 5 A. At $p_n = 4.74$ mTorr and $I_d = 10$ A, n_N reached 6.2×10^{17} m^{-3}. The actinometry resulted in nearly consistent values with the VUVAS and it decreases with p_n [6].

To considering the p_n effect on the nitrogen atom generation, n_N normalized to W_d was plotted as a function of p_n in Fig. 2 (c). It is found that values of n_N/W_d at a specific p_n are nearly the same in the three different discharge conditions. Further, monotonical increases with p_n are confirmed.

It is thought that increasing p_n and W_d will lead to the creation of a high n_N environment. An increase of W_d contributes to the increase in n_N because high W_d should increase the electron density, which will promote dissociative recombination and direct dissociation. An increase in p_n promotes a decrease in the electron temperature, causing an increase in n_N. The production rate of N via direct dissociation is ~1.6 times larger than that of dissociative recombination at $p_n = \sim 0.6$ mTorr when using the parameters measured in the 1st loop of the plasma. In contrast, at $p_n \sim 1.5$ mTorr, the production rate of dissociative recombination is ~7.6 times larger than that of direct dissociation, primarily because the high energy component of the electron energy distribution function decreases. The contribution of dissociative recombination likely increases with increasing p_n, mainly due to the decrease in electron temperature.

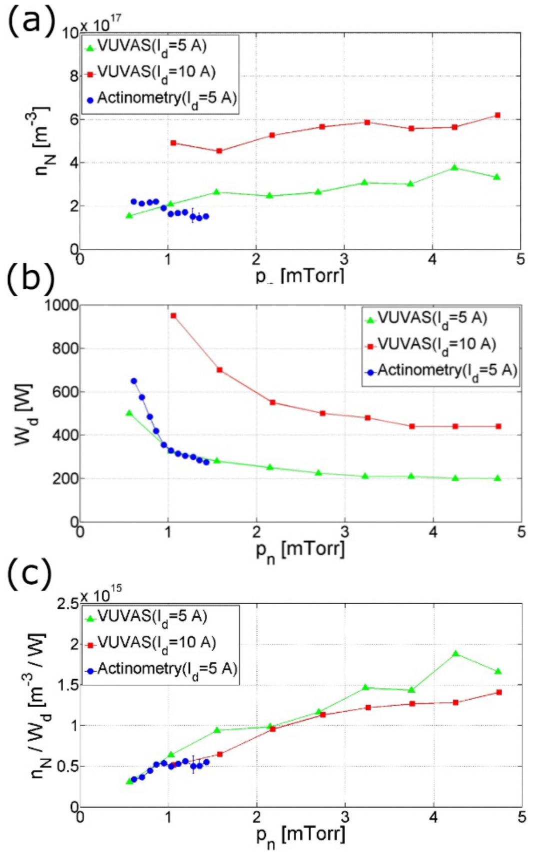

Fig. 2 p_n dependences of (a) n_N, (b) W_d, and (c) n_N/W_d. Here, the n_N were measured with VUVAS at $I_d = 5$ A (green triangle) and 10 A (red square) and actinometry at 5 A (blue circle) in three discharge series.

In this study, we succeeded in measuring the n_N at high p_n (>1.5 mTorr) and high W_d (>500 W) using VUVAS and confirmed that n_N was 2.5 - 6.2×10^{17} m^{-3}. It was also confirmed that n_N increases with increasing p_n and W_d. This is because an increase in p_n causes a decrease in electron temperature, and an increase in W_d causes an increase in electron density, which promotes dissociative recombination and direct dissociation. It would be interesting to further investigate to what degree n_N can be increased by increasing p_n and W_d.

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