

Diagnosics of N₂-Based Gas Discharge Plasma by Optical Emission Spectroscopy on Atomic and Molecular Processes

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Nitrogen-based gas discharge plasmas have been widely applied to surface modification in material or electronic engineering, while they are also interesting from the fundamental viewpoint, such as chemical compositions of upper atmosphere or ionosphere of the earth. Both for the industrial processes and for basic atmospheric studies, optical emission spectroscopy (OES) measurement is one of the best methods to examine various plasma parameters [1].

We have been studying OES characteristics of nitrogen-based plasmas of microwave discharge with the discharge pressure about 1 Torr. We are measuring 1PS and 2PS band spectra of N₂ molecule to examine vibrational and rotational temperatures (T_v and T_r) of the B $^3\Pi_g$ and C $^3\Pi_u$ states, respectively. The observed 2PS spectrum is fitted quite exactly by conventional calculation [1 – 3], while that of 1PS had not been fitted very well. Hence, we reconsidered the selection rule of 1PS (B $^3\Pi_g \rightarrow A \ ^3\Sigma_u^+$). When we strictly follow the Hund's coupling case (b) ($\Delta J = 0, \pm 1$ and $\Delta K = 0, \pm 1$) for 1PS, the spectrum calculated does not agree with the one observed experimentally (Fig. 1(a)). Meanwhile, if we adopt the basic selection rule only as $\Delta J = 0, \pm 1$, we can calculate the 1PS spectrum as in Fig. 1(b), which agrees with the one experimentally observed quite well [4, 5]. We consider that this is mainly because the rotational temperature of N₂ molecule in the plasma is so high that the Hund's case (b) is no longer a good approximation for the N₂ B state.

We not only determined (T_v , T_r), but also succeeded in extraction of three atomic nitrogen lines in the wavelength region 742 – 747 nm, by subtraction of calculated 1PS band that overlaps the weak atomic nitrogen lines in the N₂-O₂ mixture plasma with low-N₂-dissociation degree [6]. This made it possible for us to apply actinometry measurement of N atom density. It was found that the dissociation degrees of both N₂ and

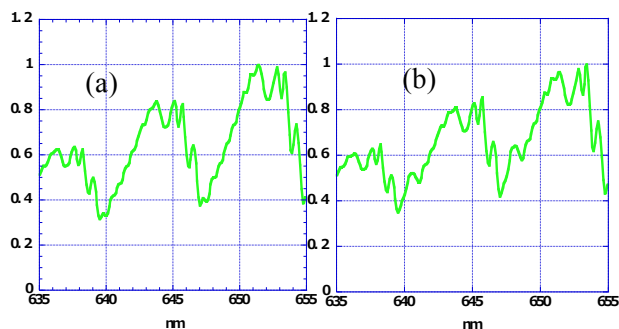


Fig. 1. Comparison of 1PS $\Delta v = 3$ spectrum calculated for $T_v = 0.6$ eV and $T_r = 0.07$ eV, the spectral resolution 0.4 nm. (a) Hund's case "b", (b) only $\Delta J = 0, \pm 1$. (b) can trace small maxima at 640.5 or 648.5 nm, while (a) cannot.

O₂ increase with the molar ratio of nitrogen in the mixed N₂/O₂ discharge gas for the same total discharge pressure [6]. We also constructed numerical kinetic model to describe number densities of excited species in the N₂/O₂ discharge. The experimental results are discussed with our numerical results in terms of elementary processes in the N₂/O₂ discharge plasma [7]. We also examined the effect of the rare-gas admixture on the N₂ dissociation. We found that the dissociation degree of nitrogen slightly decreases by about a factor of 2 – 3 with increasing amounts of Ar and Kr admixtures, whereas it becomes several times higher with increasing amount of He admixture. When we dilute nitrogen with neon, however, we find an anomalous increase in the nitrogen dissociation degree to 10 times or even several hundred times greater than that without the neon admixture even remarkably, at a considerably downstream region of the microwave discharge [8].

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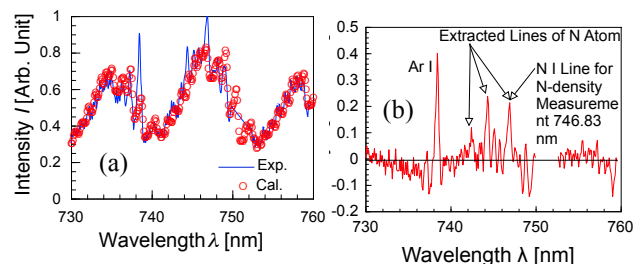


Fig. 2. Schematic diagram of extraction of atomic nitrogen lines from the experimentally observed spectrum. (a) The spectrum fitting of N₂-1PS band spectrum. Line – experimentally measured one, dots – calculated one by our method. (b) The extracted lines of nitrogen atom as subtracted spectrum.