Measurement of Vibrational and Rotational Temperatures of N₂⁺ of Nitrogen Discharge Plasma by First Negative System

First Negative Systemによる窒素プラズマ中のN₂+の振動温度,回転温度計測

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Band Spectrum of the first negative system of the N₂⁺ is measured by optical emission spectroscopy measurement to examine the vibrational and rotational temperatures of B² Σ state. The experimentally measured spectra are compared with the ones calculated theoretically to determine the temperatures of the generated plasma. We generated a microwave discharge nitrogen plasma in a cylindrical quartz tube (26 mm inside diameter) with its discharge pressure 1.0 Torr. It was found that $T_{\rm vib} \approx 1.8 - 3.4$ eV and $T_{\rm rot} \approx$ 0.13 - 0.34 eV at N₂⁺ B ² Σ ($\Delta v = 1$), whereas $T_{\rm vib} \approx 0.69$ -0.87 eV and $T_{\rm rot} \approx 0.08$ -0.18 eV at C³ Π ($\Delta v =$ 4) of N₂. $T_{\rm vib}$ and $T_{\rm rot}$ of N₂⁺ were higher than those of N₂.

1. Introduction

Nitrogen plasma plays an important role in material or electronic engineering.^[1] For example, in semiconductor industry, nitrogen plasma is applied as a nitrogen radical source at practical level.^[2] Therefore, we have studied characteristics of nitrogen plasmas and examined the dependence of their plasma parameters on discharge conditions.^[3]

In previous spectroscopic studies, we treated only neutral molecules. Therefore, as a next step, we focus on vibrational (T_{vib}) and rotational temperatures (T_{rot}) of nitrogen molecular ions.

In this study, we analyzed the first negative system (1stNS) from N_2^+ and the second positive system (2ndPS) from N_2 by optical emission spectroscopy (OES). And we examined T_{vib} and T_{rot} of N_2^+ to understand the relationship between temperatures of ions and those of neutrals.

2. Experimental setup

Figure 1 shows the block diagram of the experimental setup.^[4] Nitrogen plasma was generated using a rectangular waveguide with a cavity and a quartz tube (26 mm inside diameter). The quartz tube was aligned in the direction of the electric field of the waveguide, and its one end was inserted into a vacuum chamber. The microwave frequency was 2.45 GHz and the output power was set at 500 W. The discharge pressure was set at about 1.0 Torr. We measured the plasma at four points. The distances were 0, 60, 100, 140 mm from the intersection of the quartz tube and the waveguide to the direction of the gas flow.



Fig.1. Block diagram of microwave discharge apparatus and measurement system

3. Theoretical calculation

Generally, the line intensity of radiative transition of molecules is given as a product of a vibration-dependent part and a rotation-dependent part.

$$I_{v''J'}^{v'J'} = I_{v''}^{v'}I_{J''}^{J'}$$
(1)

If we assume the Boltzmann distribution for the vibrational and rotational states of the upper state, $I_{\nu''}^{\nu'}$ and $I_{I''}^{J'}$ can be written as follows:

$$I_{v''}^{v'} \propto q_{v'v''} v^4 \exp\left(-\frac{E_{\text{vib}}}{kT_{\text{vib}}}\right) \tag{2}$$

$$I_{J''}^{J'} \propto S_{J'J''} \exp\left(-\frac{E_{\rm rot}}{kT_{\rm rot}}\right),\tag{3}$$

where v is the frequency of the emitted light, k is

the Boltzmann constant, E_{vib} and E_{rot} are the vibrational and rotational energies of the upper state, respectively, $q_{v'v''}$ is the Franck-Condon factor, and $S_{J'J''}$ is the Hönl-London factor. ^{[5], [6]}

In this study, we analyzed the structures of the 1stNS (B ${}^{2}\Sigma_{u}^{+} \rightarrow X {}^{2}\Sigma_{g}^{+}$) of N₂⁺ and 2ndPS (C ${}^{3}\Pi_{u} \rightarrow B {}^{3}\Pi_{g}$) of N₂ to estimate the vibrational and rotational temperatures.

Applying eqs. (1) - (3), we can describe the unique rotation-vibration spectrum as a function of vibrational and rotational temperatures. Thus, we can determine these temperatures by fitting the spectra observed experimentally with the calculated ones.

Figure 2 shows an example of comparison between the experimentally observed spectrum and the calculated one (z = 100 mm, P = 1.0 Torr).



Fig.2. Example of comparison between spectra calculated theoretically and observed experimentally.

4. Results and Discussion

Figure 3 shows $T_{\rm vib}$ and $T_{\rm rot}$ of N_2^+ and N_2 obtained from 1stNS and 2ndPS, respectively. It was found that $T_{\rm vib} \approx 1.8 - 3.4$ eV and $T_{\rm rot} \approx 0.13$ - 0.34 eV for B² Σ ($\Delta v = 1$) of N_2^+ , whereas $T_{\rm vib} \approx 0.69 - 0.87$ eV and $T_{\rm rot} \approx 0.08 - 0.18$ eV for C³ Π ($\Delta v = 4$) of N_2 .

For both N_2^+ and N_2 , T_{rot} decreased as the plasma flowed to the downstream direction. T_{rot} is an approximate value to the gas temperature if rotational-translational relaxation is sufficient. Thus, it is considered that T_{rot} decrease as the plasma flows to the downstream region.

It was found that $T_{\rm vib}$ and $T_{\rm rot}$ of N_2^+ were higher than those of N₂. It is considered that this is partly because $T_{\rm vib}$ and $T_{\rm rot}$ of N_2^+ are influenced by ion temperature.

5. Conclusions

We measured the band spectra of the nitrogen-molecular ion $(N_2^+ 1stNS)$ and the neutral nitrogen molecule $(N_2 2ndPS)$, determined the

vibrational and rotational temperatures of $N_2^+ B^2 \Sigma$ and $N_2 C^3 \Pi$ states to find relationship between T_{vib} and T_{rot} of ion's and those of neutrals.

It was found that $T_{\rm vib} \approx 1.8 - 3.4 \text{ eV}$ and $T_{\rm rot} \approx 0.13 - 0.34 \text{ eV}$ for $B^2\Sigma$ ($\Delta v = 1$) of N_2^+ , whereas $T_{\rm vib} \approx 0.69 - 0.87 \text{ eV}$ and $T_{\rm rot} \approx 0.08 - 0.18 \text{ eV}$ at $C^3\Pi$ ($\Delta v = 4$) for N_2 . $T_{\rm vib}$ and $T_{\rm rot}$ of N_2^+ were higher than those of N_2 .



Fig.3. Vibrational and rotational temperatures measured experimentally (a) vibrational temperature of N_2 ($C^3\Pi$) and N_2^+ ($B^2\Sigma$) (b) rotational temperature of N_2 ($C^3\Pi$) and N_2^+ ($B^2\Sigma$).

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