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Vibrational and Rotational Temperature Spectroscopic Determination of NO Molecule in N_2-O_2 Microwave Discharge

マイクロ波放電 N_2-O_2 混合気体放電プラズマ中の NO 分子の振動回転温度の決定

Hao Tan, Atsushi Nezu, Haruaki Matsuura and Hiroshi Akatsuka
譚　浩,　根津　篤，松浦　治明，赤塚　洋

Dept. Energy Sciences, Tokyo Institute of Technology
2-12-1-N1-10 O-Okayama Meguro-ku 744, Tokyo 152-8550, Japan.
東京工業大学　〒152-8550　東京都目黒区大岡山　2－12－1

By using an optical emission spectroscopy measurement method, the vibrational and rotational temperatures of nitric oxide molecule in N_2-O_2 microwave discharge are obtained. We detect the radiation lies in γ-band (195 – 340nm) of nitric oxide, which comes from the A $^2\Sigma^+$ to X $^2\Pi$ electronic transition. We also developed a theoretical calculation for the vibration and rotation spectrum, with which we can fit the experimental results theoretically, so that the vibrational and rotational temperature can be determined. We also carried out some experiments to examine the cooling process and N_2 ratio dependence, according to which we discussed the molecular combination and excitation reactions under different plasma conditions.

1. Introduction

Nitrogen oxides (NO_x) have been a hot topic in the last decades because they always touched people's nerves in the main problems of the environment and human health.

As we know, in an N_2-O_2 mixture plasma, there exists many kinds of atomic and molecular species such as N_2, O_2, NO, N_xO, NO_x, their ions, and free electrons. Among all these particles, they go through collisions and chemical reactions, and as a result, it is possible to diagnose several radiation peaks from different molecule radiating systems of different molecules at the same time, which can provide us a better approach to the plasma dynamic process both at time-resolved and spatial accuracy.

Therefore, in order to observe the NO γ-band radiation and study the vibrational and rotational properties of NO, we carried out a spectroscopic method and examine it experimentally. Also, the comparison between NO and N_2 is also observed and considered.

3. Experimental Method

Figure 1 shows a schematic diagram of our present experimental setup. N_2-O_2 mixture plasma is generated by using a rectangular waveguide whose one end is connected to the gas entrance and the other is the outlet to a vacuum chamber by a rotary pump. A 26 mm inner diameter quartz tube is aligned in the vertical direction of the waveguide so that it can provide a free cooling down area for plasma without the microwave effect. The pressure is monitored by a membrane manometer which also shows the ultimate pressure of the chamber is 0.02 Torr. The microwave frequency is 2.45 GHz and the power is set at 600 W. The plasma is generated in the quartz tube, where the total discharge pressure is 0.5 – 2 Torr. The discharge species is a gas mixture of nitrogen and oxygen (purity 99.5%) with respective flow controllers.

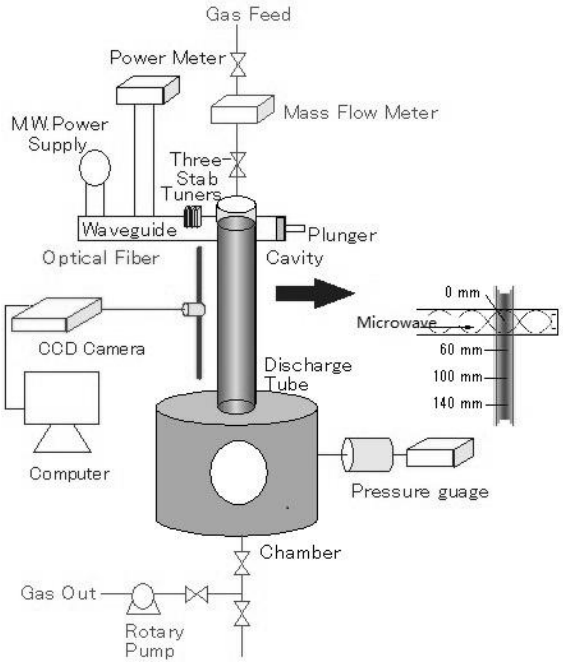

Fig. 1. Schematic diagram of experimental setup

4. Results and Discussion

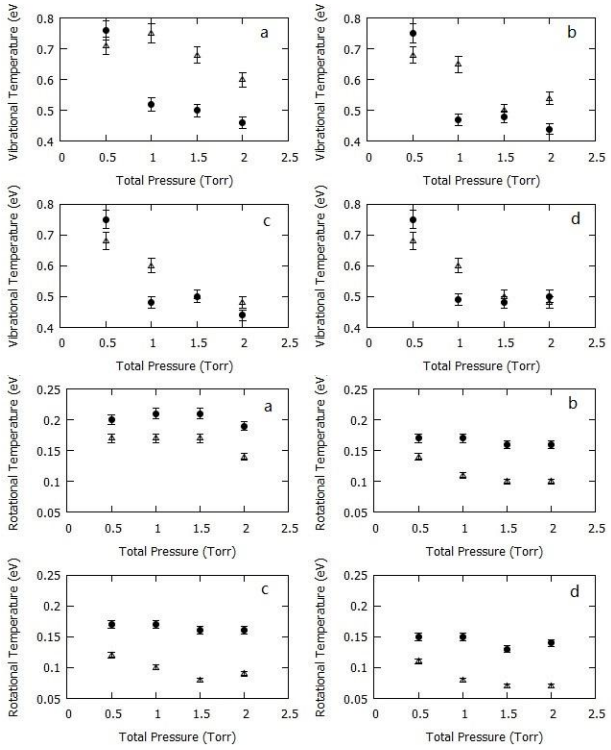

Fig. 2. Fittings of vibrational and rotational temperatures of NO (solid dots) and N_2 (hollow triangle) (a: 0 mm, b: 60 mm, c: 100 mm, d: 140 mm).

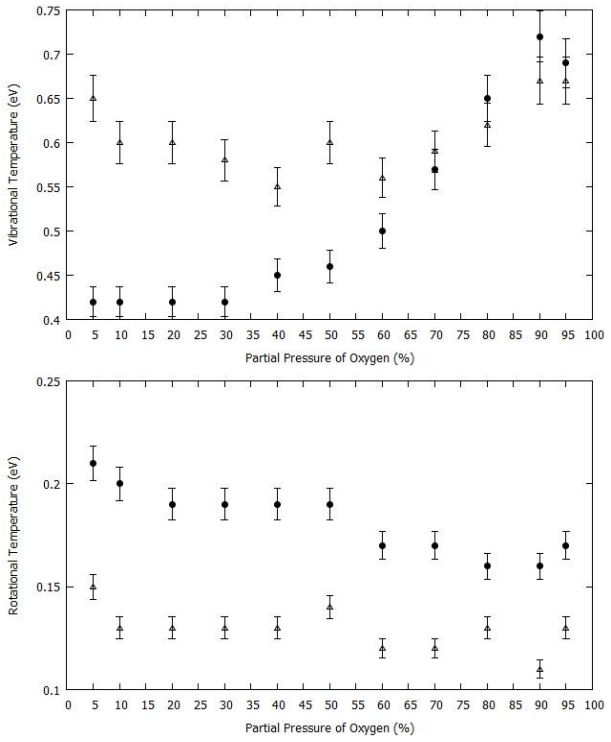

Fig. 3. Vibrational and rotational temperatures of NO $A^3\Sigma$ state (solid dots) and N_2 $C^3\Pi$ state (hollow triangle) plotted against partial pressure of oxygen in a plasma feeding gas.

Figure 2 shows the dependence on the discharge pressure of the vibrational and rotational temperatures of the N_2 2^{nd} positive system, with a discharge gas mixture of N_2:O_2 = 1:1. We measured the spectra at 0, 60, 100 and 140 mm with the pressure from 0.5 to 2 Torr. From Figure. 2, we can find that both NO and N_2 molecules experience a relaxation process towards downstream, and both vibrational and rotational temperatures increase. We can find that the relaxation rates of the results at 100 and 140 mm, are obviously small compared with the ones of 0 and 60 mm. From the results of Fig. 2, we can see that in this nonequilibrium plasma, both NO and N_2 are easily to get higher vibrational energy than rotational energy, which indicates that vibrational energy transformation is more incident during collisions.

Figure 3 shows vibrational and rotational temperatures of NO A $^2\Sigma^+$ state and of N_2 C $^3\Pi_u$ state plotted against partial pressure of oxygen with the total discharge pressure 2 Torr. We found an irregular vibrational temperature increase as the partial pressure ratio of oxygen increases for the rotational temperature of NO A $^2\Sigma^+$ state, while that of N_2 C $^3\Pi_u$ state has its minimum for O_2 ratio 40 – 60%, which is also one of major findings in our present study.

There are two general mechanisms which can explain the results above described: (i) unequal quenching or relaxation rates on different vibrational quantum levels of NO A $^2\Sigma^+$ by O_2 and O, which can lead to an irregular vibrational population at different vibrational quantum levels, and (ii) as the N_2 concentration increases, the collisions between N_2 and NO also decrease, so that the electronic excitation reaction loses its weight in the whole NO A $^2\Sigma^+$ excitation kinetics. If we consider the previous studies on N_2-O_2 mixture plasma, the excitation process of NO A $^2\Sigma^+$ relates closely to O_2 partial pressure. When O_2 or N_2 is the majority of the discharge species, dominate excitation process of NO A $^2\Sigma^+$ changes, respectively. Thus, the vibrational temperature property also changes. Since N_2 is introduced as the base discharge gas, the vibration-vibration energy transition with NO occupies a small quotient. As a result, the vibrational and rotational temperatures of N_2 do not change a lot.

References

[1] B. F. Gordiets, C. M. Ferreira, V. L. Guerra, J. M. A. H. Loureiro, J. Nahorny, D. Pagnon, M. Touzeau and M. Vialle: IEEE Trans. Plasma Sci. **23** (1995) 750.