論文 / 著書情報
Article / Book Information

題目(和文)	窒素・酸素混合気体マイクロ波放電プラズマ中の一酸化窒素励起状態(A 2Σ+, B 2Π, C 2Π)に関する実験・数値分光学的研究
Title(English)	Experimental and numerical spectroscopy examinations of NO (A 2Σ+, B 2Π, C 2Π) excited states in N2-O2 mixture microwave discharge
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出典(和文)	学位:博士（学術）, 学位授与機関:東京工業大学, 報告番号:甲第10252号, 授与年月日:2016年3月26日, 学位の種別:課程博士, 審査員:赤塚 洋,奥野 喜裕,沖野 晃俊,肖 鋒,脇 慶子
Citation(English)	Degree:, Conferring organization: Tokyo Institute of Technology, Report number:甲第10252号, Conferred date:2016/3/26, Degree Type:Course doctor, Examiner:,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

専攻： Department of	創造エネルギー　　専攻	申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of　（ 学術 ）
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		指導教員（副）： Academic Advisor(sub)	

要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

This research focuses on low-pressure and low-temperature gaseous plasma, which often involves atmospheric and ionospheric physics. We investigate the NO molecule in N_2-O_2 mixture microwave discharge because it is generated in oxidation processes in chemical and biological systems, which not only causes environmental problems but is also toxic to organisms. We observe this kind of plasma by optical emission spectroscopy measurements to obtain the spectra ranges at 200-850 nm. The plasma generates and flows in a quartz tube. In the experiment, we diagnose the spectra at different positions along the tube and different plasma conditions (electron density and temperature, total pressure, and partial pressure ratio).

We theoretically **calculate the** γ-band radiation spectrum of the electronic transition from NO A $^2\Sigma^+$ to X $^2\Pi$ state. By matching the calculated spectra with the experimental spectra, the molecular parameters in the plasma can be obtained. From the observed spectra of N_2-O_2 mixture microwave discharge at 2 Torr and 400 W output power, we found that the vibrational and rotational temperatures of NO molecules range from 0.42-0.76 and 0.13-0.21 eV, respectively, while the vibrational and rotational temperatures of N_2 range from 0.45-0.75 and 0.12-0.17 eV, respectively. From these results, besides a normal cooling process as the plasma flows to the downstream along the tube, we also find an unusual increase of the NO vibrational temperature when O_2 partial pressure ratio approaches to 100%. This indicates that the different generating processes of NO A $^2\Sigma^+$ state contribute to a different vibrational temperature distribution, and when O_2 partial pressure ratio changes, different processes dominate the generating process of NO molecules. According to some previous studies about the generating processes of NO A $^2\Sigma^+$ state in N_2-O_2 discharge, electron impact excitation from X $^2\Pi$ state and the molecular collision of X $^2\Pi$ state with N_2 $A^3\Sigma_u$ are two most important processes. However, this analysis is still quantitative.

In order to quantify the analysis above, we improve our kinetic model to calculate the reaction rates and number densities of different species in N_2-O_2 mixture plasma. In this research, we include NO $A^2\Sigma^+$, B $^2\Pi$, C $^2\Pi$ excited states in this model and modify the self-consistency condition to achieve a higher accuracy on the calculation boundary conditions. By measuring the NO γ-band (A $^2\Sigma^+$ to X $^2\Pi$), β-band (B $^2\Pi$ to X $^2\Pi$), and δ-band (C $^2\Pi$ to X $^2\Pi$) spectra, we find good agreements between the simulated and experimental results of the NO A $^2\Sigma^+$ state number densities. On the other hand, the radiation bands of β and δ are observed only when the

oxygen partial pressure is less than 3% as other results obtained by many researches on this topic, but still this reason is not sufficient. We discuss the de-excitation processes for the NO B $^2\Pi$ and C $^2\Pi$ states in this low-pressure plasma and propose an assumption that the de-excitation processes involve collision with O_2 X $^3\Sigma^-_g$ for these two levels are much faster than their generating processes, which can explain the observed spectral disappearance.

In order to prove this assumption, we carried out some experimental and calculation results. As we expected, if our assumption is right, the number density of the NO C $^2\Pi$ state in the plasma should affect the molecular properties of NO A $^2\Sigma^+$ state. Our results show that, as the ratio of the number density of NO C $^2\Pi$ and A $^2\Sigma^+$ increases, the vibrational temperature of the NO A $^2\Sigma^+$ state increases synchronously, which is a positive correlation. This indicates the NO C $^2\Pi$ state experiences a de-excitation collision and turns into A $^2\Sigma^+$ state, which finally leads to the vibrational temperature increase of NO A $^2\Sigma^+$state. Because in this kind of N_2-O_2 plasma, NO C $^2\Pi$ state is generating through the atomic collision between N and O, which can lead to the molecules at a high vibrational level distribution. Considering NO C $^2\Pi$ state de-excitation is too fast to change its vibrational distribution, as a result, the part of NO A $^2\Sigma^+$ state generated from C $^2\Pi$ state has a higher vibrational temperature than the others. Hence, there should be three processes dominate the generating NO A $^2\Sigma^+$ state in N_2-O_2 plasma, the electron impact excitation, the collision of NO X $^2\Pi$ state with N_2 A$^3\Sigma_u$, and the collision of NO C $^2\Pi$ state with N_2 O_2 X $^3\Sigma^-_g$, which is the most important finding in this study. Meanwhile, it is not a direct evidence.

Besides the low-pressure N_2-O_2 plasma, our finding can also explain the appearance of the NO β-band and δ-band at atmosphere. Not all molecules at B $^2\Pi$ and C $^2\Pi$ states can live for a de-excitation collision, therefore, some of them will de-excite to NO X $^2\Pi$ state by spontaneous electronic transitions. To further develop this investigation, more experimental data for determining the rate coefficients of molecular collision excitation and de-excitation should be included.