Experimental and theoretical study on emission spectra of a nitrogen photoionized plasma induced by intense EUV pulses

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Abstract. Spectral lines of low-temperature nitrogen photoionized plasma were investigated. The photoionized plasma was created in the result of irradiation N_2 gas using laser plasma EUV radiation pulses. The source was based on a 10J/10ns Nd:YAG (λ = 1064 nm) laser system and a gas puff target. The EUV radiation pulses were collected and focused using a grazing incidence multifoil EUV collector. The emission spectra were measured in the ultraviolet and visible (UV/Vis) range. It was found that the plasma emission lines in the lower region of the UV range are relativley weak. Nonetheless, a part of the spectra contains strong molecular band in the 300 - 430 nm originated from second positive and first negative systems band transitions of nitrogen. These molecular band transitions were identified using a code for study the diatomic molecules, LIFBASE. The vibrational band of Δv = 0 and ±1 transitions were significantly populated than of that with Δv = ±2 and 3 transitions. A comparison of the calculated and measured spectrum is presented. With an assumption of a local thermodynamic equilibrium (LTE), the vibrational temperature was determined from the integrated band intensities with the help of the Boltzmann plot method and compared to the temperature predicted by SPECAIR and LIFBASE simulations. A summary of the results and the variations in the vibrational temperatures was discussed.

1 Introduction

Photoionization of molecular nitrogen is performed and emission spectra in the result of the photoionization was measured. The plasma was produced by a laser-produced plasma (LPP) extreme ultraviolet (EUV) source. The experimental source was based on a Nd: YAG laser system delivering a 10 J, 10 ns pulse at the fundamental wavelength, λ =1064 nm. The laser power density in such regime can be easily achieved an order of 10^{12} W/cm^2 in the interaction region. Such intensity range can be enough to laser-produced xenon plasma and therefore the EUV radiation collected was used to ionize nitrogen gas up to first few ionization states. Mostly, spectral lines from molecular band transitions in the ultraviolet and optical wavelengths region were the dominant features in the spectra. The molecular emission spectra were found corresponding to the second positive (2PS) ($C^3\Pi_u$-$B^3\Pi_g$) and first negative (1NS) ($B^2\Sigma^+{}_u$-$X^2\Sigma^+{}_g$) systems of the molecular nitrogen. The identification of the molecular band transitions was performed using software for diatomic molecules simulation, LIFBASE [1]. The plasma parameters and its condition can be studied based on these molecular band transitions by evaluating vibrational temperature with an assumption of the local thermodynamic equilibrium. The vibrational temperatures provide insight to the molecular vibrational processes while the rotational temperature usually represents the gas temperature and it is responsible of reaction rates data of many molecular processes. In thermodynamic equilibrium condition, determination of the rotational and vibrational temperatures is often done by assuming the vibrational levels based on the Boltzmann distribution function. For nitrogen plasma, the band transitions from the upper electronic to the ground electronic states in 2PS and 1NS systems and their characteristics make possible to estimate the vibrational temperatures from emission intensities. Though, spectroscopic investigations on nitrogen plasmas or nitrogen-containing plasmas have been studied using the band spectra recorded in the various plasmas at different experimental conditions. Of these, include a compact helicon plasma source [2], nitrogen-containing plasmas [3] as well as in highly constricted plasma and applying the fitting methods [4]. Further, the plasma emission lines from 2PS are well-known and the best candidate used for estimation of the vibrational temperature in a nitrogen or nitrogen-containing plasma. Provided that if an assumption of the thermodynamic equilibrium between the ground and excited electronic states is achieved [5]. In this case, the required condition is the radiative rates are dominated by the collision rates to ensure the LTE condition in plasma. Knowledge of these population rates will provide more information

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about the plasma emissions processes and control conditions. However, if the plasma is far from LTE, determinations of vibrational temperature is more sophisticated unless the high-resolution spectra were used. In this case, the vibrational temperature inferred from the Boltzmann plot methods still give insight into the plasma condition. Though, the 2PS of nitrogen molecule was widely used to study the gas temperatures in nitrogen and nitrogen-containing plasmas [6-10].

In this work, the emission lines of nitrogen photoionized plasma created by laser plasma EUV radiation pulse is investigated. The electron vibrational temperature is determined using the integrated line intensities of the band transitions in the second positive system of nitrogen. This can be done with the help of the Boltzmann plot of vibrational and rotational levels. In this case, especially for the molecular spectroscopy, much attention must be paid to ensure the high-resolution of the molecular band spectrum which enables us to accurately determine the band head wavelength of a transition. On the other hand, the determination of the vibrational and rotational temperature can be predicted using software for the molecular spectral simulation, LIFBASE. In this case, the temperature was set manually in acquiring the simulated. The more accurate data on electron vibrational or rotational temperature can be utilized by direct fitting of the measured spectra [2]. The most applicable software used for this purpose is the commercial software SPECAIR [11].

2 Experimental setup

In the experiments, Nd: YAG laser system delivering the maximum energy of 10J/10 ns pulse and 10 Hz repetition rate was used. In the present measurements, energy about 5.6 J per pulse was employed. The laser pulse was focused onto a double stream gas-puff target, created synchronously with the laser pulse. The gas-puff target was created by pulsed injection of high-Z (argon or xenon) gas into a hollow stream of low-Z helium gas using a double nozzle set-up. The nozzle consists of a small central orifice surrounded by a ring-shaped outer nozzle and equipped with an electromagnetic valve system. More experimental detailed description of the double stream gas puff target can be found in [12,13]. Irradiation of nitrogen gas (4 bar) injected into the vacuum interaction chamber synchronously with the radiation pulses from laser plasma EUV source resulted in photoionized plasma. The photoionized plasma emissions and focusing conditions were adjusted in the way to obtain maximum power density in the interaction region. The maximum intensity is estimated around 10^8 W/cm^2 under EUV fluence of ~300mJ/cm^2. The EUV radiation was focused using a gold-plated grazing incidence multifoil EUV collector. The spectral analyzing instrument was an Echelle Spectra Analyzer ESA 4000 spectrograph, equipped with the ICCD Kodak KAF 1001 camera. The spectrometer system allowed for simultaneous measurements of complex spectra within the wide UV/Vis (200-780 nm) spectral range with considerably higher spectral resolution $\lambda/\Delta\lambda \approx 20000$. Schematic view of the EUV photoionization experimental setup is shown in Fig. 1.

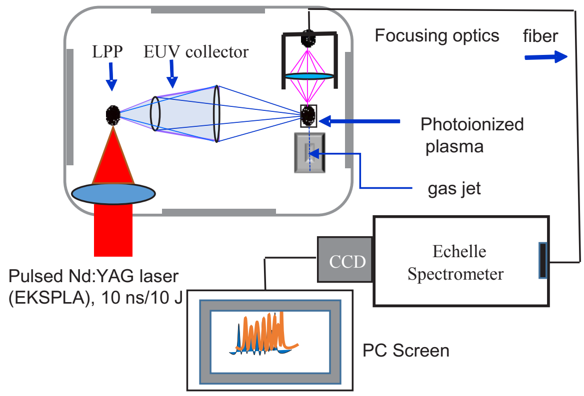

Fig. 1. Schematic view of the experimental setup for the EUV photoionization experiments.

2.1. Vibrational temperature

The spectrum of the generated photoionized plasma is dominated by the emission of second positive N_2 ($C^3\Pi_u$ - $B^3\Pi_g$) and first negative N^+_2 ($B^2\Sigma^+_u$ - $X^2\Sigma^+_g$) systems of molecular features of nitrogen. If the assumption of the upper populations having Boltzmann distributions with both vibrational and rotational temperatures T_v and T_r, respectively, the line intensity of the molecular band system between the upper v',J' and lower v'',J'' rovibrational levels can be expressed as following [14].

$$I(v',J' \rightarrow v'',J'') = \frac{CR_{SP}}{\lambda^4}\frac{q(v',v'')}{Q_{vib}}\frac{s(J',J'')}{Q_{rot}} \times \exp\left(\frac{G(v')}{k_B T_{vib}}\right)\exp\left(\frac{F(v',J')}{k_B T_{rot}}\right) \quad (1)$$

Here, C is the scaling factor related to the collection solid angle and the sample volume of the optical emission, R_{SP} is the spectral response function, $s(J',J'')$ is strength function and $F(v',J')$ rotational spectral term. Q_{vib}, Q_{rot} are the vibrational and rotational partition functions, respectively. $q(v',v'')$ is the Franck-Condon factor of the vibrational transition (v' - v'') [15].

For simplicity, we can obtain the T_v easily by from the Boltzmann plot using integrated band intensities $I(v',v'')$ as a function of the upper vibrational energies of different transition in the second positive system. In such case, as can be seen from equation (1), the total intensity is given for both vibrational and rotational levels. To deduce the vibrational temperature, we can use the vibrational dependent portion as follows [16,17]:

$$\ln\left(\frac{I(v',v'') \times \lambda(v',v'')}{A(v',v'')}\right) = \frac{G(v')}{k_B T_{vib}} + const. \quad (2)$$

Where $I(v',v'')$ is the intensity of vibrational band, $G(v')$ represents the vibrational energy of the upper level, T_v is the vibrational temperature, k_B is the Boltzmann constant and $A(v',v'')$ and $\lambda(v',v'')$ are respectively the transition probability and the wavelength of the band head.

3 Results and discussion

Identification of emission line in plasma has great importance and it is considered as the first step to specify the plasma species and their charge states. Fig.2 shows the measured and simulated emission spectra of molecular nitrogen. The dominant features in the spectrum are the molecular band transitions in 2PS and 1NS systems of N_2 in the ultraviolet (UV) wavelength range. Most of the emission lines were corresponding to the 2PS in the 300-400 nm range. The measured and SPECAIR simulation results were shown a good agreement with relatively different in their intensities, where the experimental emission lines are more strength. Although simulated with the T_v = 3358.9 K obtained from linearity of the Boltzmann plot, the spectra computed using LIFBASE also showed the strongest band transitions, (0-0) and (0-1) in 1NS (B-X).

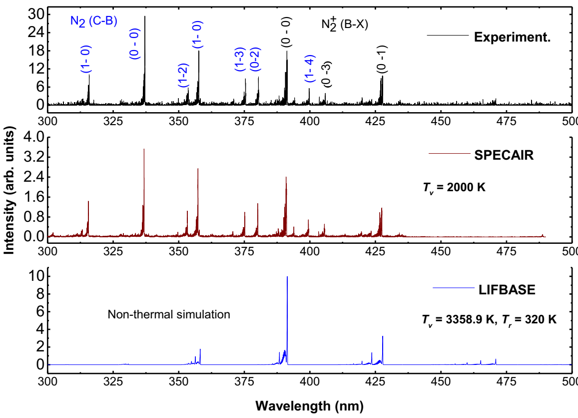

Fig. 2. Measured (top) and simulated UV/Vis emission spectra belong to the 1NS (B-X) and second positive 2PS (C-B) systems of nitrogen induced by the intense laser plasma EUV pulses.

It has been noticed that even for the T_v of 2000 K, the results do not show any significant difference in terms of molecular band transitions rather in the line intensity. The best T_v can be found by fitting the measured spectra with an appropriate fitting function.

In Fig.3, the experimental (top) and simulated (bottom) spectra corresponding to the (1-0) and (1-2) vibrational band transitions of the 2PS ($C^3\Pi_u$ - $B^3\Pi_g$) acquired at a pressure of 4 bar at a pulse repetition rate of 10 Hz with the energy of 5.6 J per pulse. A good agreement was found between the measured and simulated spectra for a temperature of 2000 K. This spectra is mainly part of the spectrum in Fig.2 and has been taken between 354 nm to 359 nm to clearly demonstrate the measured and calculated vibrational spectra.

To evaluate electron vibrational temperature, Fig.4 shows the Boltzmann plot of the integrated band intensities versus the upper vibrational energy levels corresponding to the transitions in 2PS. We have used six transitions in 2PS to infer the T_v because the spectra dominated mostly with molecular band transitions in the 2PS band system, Fig.2. The fitted line to the experimental points gives rise an approximate vibrational temperature of 3358.9 ± 665 K. This value is a bit higher compared to the best value of the T_v (2000 K) that used to obtain the best fit between the simulated and measured spectra with SPECAIR software under the present experimental conditions. Despite the temperature differences, the measured and calculated spectra using SPECAIR were found to be in good agreement. Such discrepancies are expected whether associated with experimental errors or attributed the use of a small number of vibrational transitions, as it is always recommended to include much more lines in constructing the Boltzmann plot. In the other hand, when the determined vibrational temperature 3358.9 K is used to simulate the measured spectrum with LIFBASE software, the computed spectra contain molecular bands mainly from 1NS (B-X), (0-0) and (0-1) emissions.

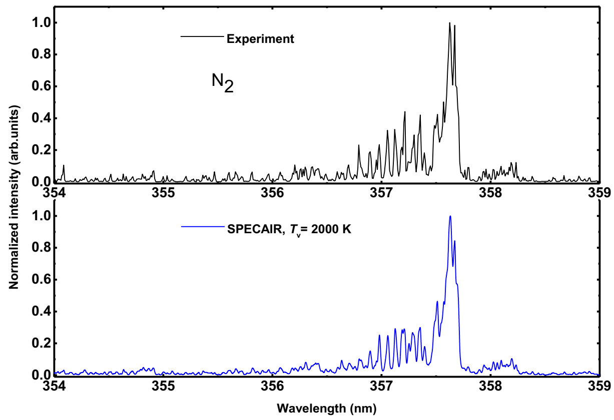

Fig.3. Comparison of measured (black) and calculated (red) emission spectra of a second positive ($C^3\Pi_u$-$B^3\Pi_g$) band transitions in plasma over a wavelength range from 354 nm to 359 nm

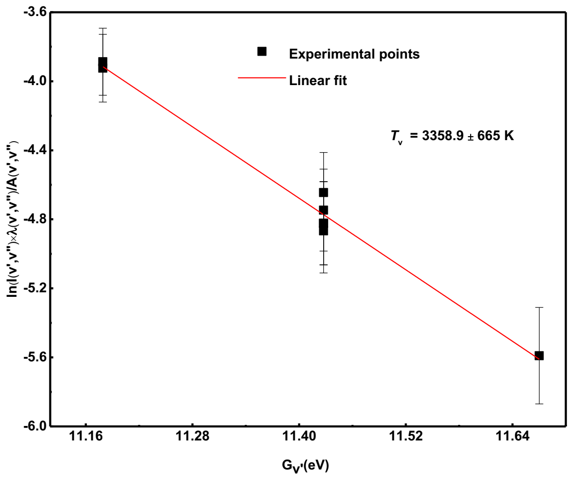

Fig.4. Vibrational temperature determination from the Boltzmann plot of the Δv = 0, ±1,-2, and -3 sequences in second positive 2PS band transitions.

In Fig.5 the vibrational population and its dependence on the T_v are examined using emission spectra in the 1NS. That calculated vibrational populations at a lower T_v, 2000 K, are found to be more vibrationally populated than that calculated at a higher T_v, 3358.9 K. This can be explained as at lower T_v, most of a gas molecule within molecule will be vibrated at their fixed vibrational levels. With Enhancing T_v the individual amplitude of each molecule increases before the molecules undergo the translational motion, this resulted in less populated states at higher vibrational temperatures. Therefore, the vibrational bands with the $\Delta v = 0$ and ± 1 transitions in $B^2\ \Sigma^+_u$ - $X^2\ \Sigma^+_g$ system are significantly populated than of that with $\Delta v = \pm 2$ and 3 transitions. As the plasma in LTE, the gas temperature can be considered as the rotational temperature T_r that measured in the upper states in a molecular transition. In contrast, for non-LTE plasmas, measurements of the gas temperature is possible via analyzing the rotational molecular emission provided that the rotational-translational relaxation is sufficiently fast to equilibrate the gas and rotational temperatures [18].

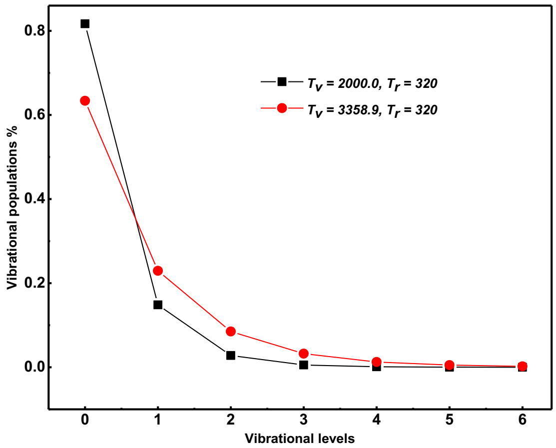

Fig. 5. Vibrational populations of non-thermal mode vibrational levels of 2PS, obtained at the end of the simulations for two different temperatures (T_v,T_r).

4 Conclusions

In summary, laser-plasma EUV source was used to create a low-temperature photoionized nitrogen plasma. High-resolution emission lines from photoionized plasma in the UV/Vis spectral range were recorded using echelle spectral analyzer ESA 4000. The dominant molecular band transitions in plasma were the electronic transitions from 2PS ($C^3\Pi_u$-$B^3\Pi_g$) and 1NS ($B^2\Sigma^+_u$ - $X^2\Sigma^+_g$) systems. The diatomic molecular simulation programs, LIFBASE and SPECAIR have been applied to reproduce the experimental spectra theoretically and interpretation. A comparison of the measured and calculated emission lines is given. With the assumption of the plasma in the LTE condition, the linearity of the Boltzmann plot is obtained using the integrated band intensities in 2PS vibrational transitions. A vibrational temperature of 3358.9 K is inferred from the used Δv = 0,±1,±2 and 3 transitions. The T_v calculated with the aid of the Boltzmann plot is comparable to the results obtained using the intensity ratio of two consecutive vibrational bands of the same sequence in 2PS from a helium microwave gas discharge [19].

The inconsistency between the simulated and measured vibrational temperatures was expected. The differences can be explained due to the departure of the plasma from the local thermodynamic equilibrium and low degree of ionization. The method of direct fitting the experimental emission spectra may yield reasonable results of vibrational temperature in plasma in the case where the Boltzmann plot is not accurate either for low intensity of the band emission or due to the weak radiative transition rates of a line.

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