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Spatial distribution of the neutral gas temperature in rf and dc magnetron sputter discharges determined by fitting rotational spectra of the N_2^+ using a two-temperature fit

V. Linss*

TU Chemnitz, Institut für Physik, 09107 Chemnitz, Germany

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Abstract

The spatial distribution of the neutral gas temperature in a dc and rf magnetron sputter discharge (boron target) with pure nitrogen was estimated by fitting the rotational spectra of the first negative system of the nitrogen molecule ion. The rotational levels of the excited nitrogen molecule ion are populated mainly by two mechanisms. Only one of them leads to a rotational level distribution corresponding to the neutral gas temperature. Therefore, fitting of the spectra assuming a single Boltzmann distribution of the rotational levels often leads to unreliable and too high rotational temperatures and the spectra have to be fitted using a two-temperature model. Beside the neutral gas temperature the value of the second temperature as well as the contribution of the higher rotationally excited molecules to the spectra are studied in more detail. The dependence of the spatial distribution of the neutral gas temperature on pressure and discharge power was investigated. Additionally, the results are compared to rotational temperatures obtained by fitting the spectra assuming a single Boltzmann distribution of the rotational levels. These rotational temperatures often do not reflect the neutral gas temperature and wrong dependences would be concluded.

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1. Introduction

The neutral gas temperature is an important process parameter in low pressure discharges. On the one hand, chemical reactions may be thermally driven. On the other hand the neutral gas may contribute to the thermal substrate load, and in particular for thermally sensitive substrates the knowledge of the neutral gas temperature is essential. The neutral gas temperature is often determined by evaluating the shape of rotational spectra of diatomic molecules present in the plasma. A great variety of molecules has been used: H_2 [1], C_2 [2,3], Bi_2 [4], CN [5,6], BH [7,8], CH [9], NH [9], SiH [10], SiO [11], OH [12,13], NO [14], O_2 [15,16]. But one of the probably most used is the nitrogen molecule and its ion. The rotational level distribution of a certain electronic and vibronic state can be measured by several methods, among them optical emission spectroscopy (OES), laser-induced fluorescence, Raman scattering [17], Fourier-transformed infrared spectroscopy [4], resonance-enhanced multiphoton ionization [18] or broad-band mode-locked cavity-enhanced absorption spectroscopy [19]. Under certain experimental conditions the rotational temperature reflects the neutral gas temperature more reliably than the translational temperature measured by Doppler broadening [20].

Only if the lifetime of the excited molecule is long enough for thermalisation the rotational level population represented by the emission band reflects the neutral gas temperature. This is the case for high enough pressures with high collision rates (even if the lifetime is significantly reduced due to collisions) [12,21] or for metastable

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* Present address: ASMEC Advanced Surface Mechanics GmbH, Bautzner Landstr. 45, D01454, Radeberg, Germany.

E-mail address: linss@physik.tu-chemnitz.de.

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molecule states such as the $N_2(A^3\Sigma_u^+)$ [22]. However, in low pressure plasmas the light emission usually takes place before the thermalisation of the rotational level population of the electronically excited state. Thus, the observed population of the rotational levels is determined by the excitation mechanisms and the rotational level population of the electronic states from which the upper electronic state of the chosen molecule band is populated. For a correct temperature determination all excitation channels have to be considered [23–25]. This fact is frequently neglected in the literature, where only direct population by electron impact is considered [26,27] or the rotational level distribution of the excited molecule is simply assumed to be a Boltzmann distribution according to the gas temperature [28,29]. In Ref. [30] it was shown that for the 0-0 band of the first negative system (FNS0-0) of the N_2^+ at least two excitation channels have to be considered in order to obtain a reliable measure for the gas temperature. One excitation channel results in a rotational level distribution according to the neutral gas temperature. The second excitation channel is connected with rotational excitation and thus the rotational temperature of so generated molecules is much higher than the gas temperature. The two-temperature model will be summarised in Section 3.

In this article, the neutral gas temperature estimation in nitrogen dc and rf magnetron sputter discharges with a boron target using the two-temperature model will be reported on. Additionally, the results of the rotational temperature determination assuming a single Boltzmann distribution will be given for comparison. Furthermore, the rotational temperature of the highly rotationally excited N_2^+ molecules will be discussed.

In this article the commonly used abbreviations for the electronic states of the molecules will be used: $N_2(X)$ instead of $N_2(X^1\Sigma_g^+)$, $N_2(C)$ instead of $N_2(C^3\Pi_u)$, $N_2^+(X)$ instead of $N_2^+(X^2\Sigma_g^+)$ and $N_2^+(B)$ instead of $N_2^+(B^2\Sigma_u^+)$.

2. Experimental set-up

2.1. Magnetron sputter discharges

The experiments were carried out in a PLS570 assembly (Balzers) equipped with a turbomolecular pump. The vacuum chamber had a height and a diameter of approximately 0.75 m. A water cooled circular magnetron source with a 10 cm boron target was used. The discharge was operated in rf (13.56 MHz) or dc mode. The target-substrate distance was 10 cm. Pure nitrogen was used as working gas with a gas flow of 100 sccm controlled by a mass flow controller (MKS Instruments). The pressure was adjusted by the throttle valve position and measured using a baratron gauge. A schematic drawing of the assembly can be found in Ref. [31]. OES spectra were recorded at different distances from the target (0…7 cm) for both the rf and dc magnetron discharge. For each type of discharge the discharge power was varied at a pressure of 0.2 Pa (rf discharge 50…500 W, dc discharge 50…300 W) and the pressure was varied at constant discharge power (rf discharge 0.2…1.5 Pa at 500 W, dc discharge 0.2…2.0 Pa at 200 W). For the selected pressure range the mean free path of the nitrogen molecules is well below the dimensions of the vacuum chamber so that thermal equilibration of the gas can be assumed (product of mean free path and pressure for nitrogen at 300 K is $6.8*10^{-3}$ mPa [32]). Further information about magnetron sputter discharges can be found in Ref. [33] where this type of discharge is compared with other discharges.

2.2. Recording of the spectra

OES spectra were recorded using a spectrometer HR 460S (Jobin Yvon) with a focal length of 460 mm and a LN_2 cooled CCD detector. The grating with 2400 groves/mm yielded a spectral resolution of about 0.05 nm. The relative spectral sensitivity was constant in the recorded wavelength range [34]. Light emission from the plasma was collected by a glass fibre. The angle for light collection was limited to about 1.5° by a small tube of ceramics. The integration time to record a spectrum was between 0.5 and 60 s depending on the spatial plasma density and three accumulations were averaged.

3. Fitting of the rotational spectra

Before fitting the FNS0-0 spectra a linear background was subtracted and the spectra were cut towards lower wavelengths. So, the rotational lines with rotational quantum numbers of the upper electronic state $K'>15$ of the R branch and $K'>40$ of the P branch were not considered in the fit because these lines might be overlaid by the SPS3-6 and the FNS1-1. Then the spectra were fitted using two methods introduced in Ref. [30]. Firstly, a single Boltzmann distribution of the rotational levels was assumed. This method was given the name “conventional fit”. Secondly, the spectra were fitted assuming two contributions with different Boltzmann distributions of the rotational levels referred to as “two-temperature fit”. Before a short summary of the two fitting methods the main excitation channels of the $N_2^+(B)$ state in low pressure plasmas should be mentioned (see also Ref. [30] and references given therein). It can be populated by electron impact from the ground state of the neutral N_2 molecule $N_2(X)$ [35,36] via

$$N_2(X) + e \Rightarrow N_2^+(B) + e + e \tag{1}$$

or stepwise via ionisation and subsequent excitation of the molecule ion. Important ionisation mechanisms in low pressure plasmas are the electron impact on the neutral N_2 molecule but also Penning ionisation during the

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