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Intensity ratio of spectral bands of nitrogen as a measure of electric field strength in plasmas

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Abstract

The ratios of intensities of the spectral bands of molecular nitrogen corresponding to transitions $N_2^+(B\,^2\Sigma_g^+, v = 0) \rightarrow N_2^+(X\,^2\Sigma_g^+, v = 0)$, $N_2(C\,^3\Pi_u, v = 0) \rightarrow N_2(B\,^3\Pi_g, v = 0)$ and $N_2(C\,^3\Pi_u, v = 2) \rightarrow N_2(B\,^3\Pi_g, v = 5)$ as a function of the applied electric field strength were measured for air in the pressure range of 300 to 10^5 Pa. The non-self-sustaining dc discharge in a parallel-plane gap was used for excitation of gas molecules. The reduced field strength was varied in the range of (150–5000) $\times\ 10^{-21}$ V m^2. The measured ratio of intensities as a function of electric field strength is compared with the theoretical estimates made by other authors. The obtained intensity ratio versus field strength curves can be used for field strength estimation in plasmas if the nitrogen molecules are excited dominantly from the ground state directly by the electron impact.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In plasma applications and research, an estimation of the electrical field strength in the plasma is a frequent task. A spectroscopic method based on the measurement of the intensity ratio of nitrogen spectral bands can be used for this purpose if the excitation of nitrogen molecules from the ground state by electron impact is the dominant process in the plasma. Usually, the intensity of the most intensive spectral band of the second positive system (SPS) of the nitrogen molecule N_2 and that of the first negative system (FNS) of the ionized molecule N_2^+ are compared [1–5]. These bands correspond to transitions $N_2(C\,^3\Pi_u, v = 0) \rightarrow N_2(B\,^3\Pi_g, v = 0)$ and $N_2^+(B\,^2\Sigma_g^+, v = 0) \rightarrow N_2^+(X\,^2\Sigma_g^+, v = 0)$, shortly denoted as SPS(0,0) and FNS(0,0). The wavelengths of these bandheads are 337.1 nm and 391.4 nm, respectively. We denote below the ratio of the intensities of these bands by $R_{391/337}$.

For determination of the field strength, the ratio $R_{391/337}$ must be known as a function of the electric field strength. The results of theoretical calculations of that ratio as a function of reduced field strength, E/N, where E is the field strength and N is gas number density, are presented in the literature [2, 4, 6]. The calculated ratios $R_{391/337}$ in these publications differ from each other remarkably, as is pointed out in papers [3, 7] and can be seen in figure 6 of the present paper. A measurement of $R_{391/337}$ as a function of E/N was lately performed for air under pulsed excitation conditions [7]. The shape of the experimental curve follows the theoretical predictions but experimentally ascertained values are about 3–6 times higher than the theoretical ones.

The following circumstances forced us to continue experimental investigations of the behaviour of relative intensities of the bands of the nitrogen spectrum depending on the field strength:

- The above-mentioned discrepancy between the experimental and the theoretical values of $R_{391/337}$ calls

for additional measurements. Changes in measurement method and apparatus are recommended.

- Our previous measurements of $R_{391/337}$ were carried out in the E/N range of 135–285 Td. There is a need to extend the range of the investigated field strength.
- The intensity ratio $R_{391/337}(E/N)$ presented in the paper [7] was measured in air at the atmospheric pressure. It is not obvious that the same field dependence is valid for lower or higher pressures. Thus, measurements at different pressures should be carried out.

A measurement of the intensity ratio $R_{391/337}$ is complicated because of considerable differences in wavelengths and intensities of SPS(0, 0) and FNS(0, 0) bands. The transmission of the optical system (lenses, fibres, monochromator, photomultiplier etc) depends on the radiation wavelength λ. Thus, a calibration of the optical system (determination of the transmission factor as a function of λ) is necessary. If instead of SPS(0, 0) another band, SPS(2, 5), which corresponds to the transition $N_2(C\,^3\Pi_u, v = 2) \rightarrow N_2(B\,^3\Pi_g, v = 5)$ with bandhead at $\lambda = 394.3$ nm, is used, then the spectral calibration is not so essential. The response of an optical system does not change significantly in the narrow wavelength interval from 389 to 395 nm that covers the spectral bands of interest. The SPS(2, 5) band must be recorded anyway, because it is partly overlapping with the FNS(0, 0) band, whose relative intensity is the subject of interest. As the intensity distribution within the SPS(2, 5) band must be recorded, so it is reasonable to use this band for field measurements. Below we denote the intensity ratio of the nitrogen bands FNS(0, 0) and SPS(2, 5) as $R_{391/394}$. Usage of the ratio $R_{391/394}$ instead of the ratio $R_{391/337}$ has an additional advantage: the spectrum in the narrow wavelength interval from 389 to 395 nm can be recorded with sufficient resolution by a single shot using a modern spectrum recorder with a CCD array.

In this paper, the intensity ratio of the rotational bands of the 0–0 vibrational transition of FNS and 0–0 and 2–5 vibrational transitions of SPS of nitrogen as a function of the reduced field strength is recorded experimentally. We exploited the steady non-self-sustaining discharge in contrast to the pulsed discharge used in our previous work [7]. The aim of the study is to find the relationship between electric field and radiation that could be used for field strength estimations in atmospheric and lower pressure air discharges.

2. Experimental set-up and measuring procedure

The sketch of the experimental set-up is presented in figure 1.

The non-self-sustaining discharge was excited between parallel plate electrodes. The anode was made of brass. A thin semitransparent aluminium coating evaporated on a quartz plate served as a cathode. The distance, d, between the electrodes was adjustable with an accuracy of 0.01 mm. The electrodes were installed in a vacuum chamber equipped with quartz windows. Ambient air was dried with silica gel, filtered out of the dust and directed into the chamber. The airflow rate through the chamber was maintained at 1 litre min^{-1} during all the measurements. The pressure in the chamber was controlled by valves. It was measured with the help of a piezoelectric and micro Pirani transducer. The accuracy of the transducer

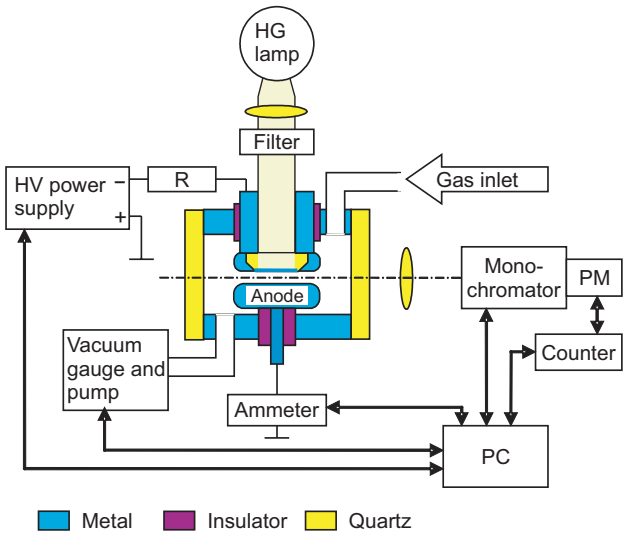

Figure 1. Sketch of the experimental set-up.

was 1% at pressures above 10 Torr and 10% at pressures below 10 Torr.

The anode was grounded via a digital ammeter that could provide the current resolution of 0.1 nA. The cathode was stressed via a current-limiting resistor $R = 100\,\text{M}\Omega$. The initial electrons were liberated from the cathode by the UV radiation ($\lambda = 253$ nm) of a low pressure mercury lamp supplied with an interference filter. The diameter of the illuminated area of the cathode was 18 mm.

The electric field strength was set by adjusting the distance between the electrodes and/or the applied voltage. The radiation from the discharge was focused with an achromatic quartz lens of 75 mm in focal length on the input slit of a monochromator. The discharge was imaged onto the plane of the slit with the reduction of 2. The linear dispersion of the monochromator MDR-23 was 1.3 nm mm^{-1}. Both the slits of the monochromator were 0.15 mm in width. A photomultiplier PM (PMH-100-4, Hamamatsu) in the photon-counting mode was used for detection of the radiation. The photocathode of the PM was cooled down to $-20\,^{\circ}\text{C}$. As a result, the dark count rate was reduced to about 40 s^{-1}. The counting time was set equal to 1 s.

The relative spectral sensitivity of the entire optical system was determined using a tungsten lamp and a deuterium lamp, whose spectral characteristics were known.

The instrumental function of the system was recorded for every distance d between the electrodes. For that purpose the discharge gap was illuminated with a mercury lamp and the spectral response of the light recording system, $F(\lambda)$, was registered for mercury line λ_M 404.7 nm.

The values of pressure, discharge current and counting rate of PM were measured simultaneously for each recorded wavelength, λ.

3. Experimental conditions

3.1. Discharge current

The discharge current was kept low to avoid a distortion of the initially homogeneous field by a space charge of current

carriers in the discharge gap. According to the 1D Poisson equation the maximum value of the space charge field strength $E_\rho = (e/\varepsilon_0)\int_0^d n(x)\mathrm{d}x$. Here e is the elementary charge and $n(x)$ is the number density of charged particles in the gap. In our case, most of the electrons and all of the positive ions are created in the discharge gap due to the electron impact ionization of molecules. The number density of electrons increases exponentially with the distance from the cathode, and nearly all of the positive ions and electrons are thus created near the anode. Therefore, the drift length of most electrons is much shorter than that of positive ions: the electrons reach the anode quickly but almost all of the positive ions must pass the full distance from the anode to the cathode. Taking into account that the mobility of electrons exceeds that of ions by about two hundred times, one can conclude that the number of ions in the discharge gap exceeds the number of electrons by several orders of magnitude at any time instant. Thus, only the contribution of positive ions to the field distortion should be considered. The contribution of negative ions is negligible for two reasons. First, at field strengths above 150 Td the attachment is unimportant in comparison with the ionization [8,9]. Second, most of the charged particles are created close to the anode and thus the residence time of the negative ions in the gap is much shorter than that of the positive ions. Thus, $\int_0^d n(x)\mathrm{d}x \cong N_+/S$, where N_+ is the total number of positive ions in the discharge gap, and S is the area of the electrode surface occupied by the discharge. The total current i through the gap has two components: an electron component and an ion component. Because the path length of most electrons is much shorter than that of positive ions, the ion component prevails in the gap and the expression for the total current can be written as follows: $i \cong (e \cdot N_+)/T_+$ where T_+ is the drift time of positive ions. Assuming that the field distortion is small, we can write $T_+ \approx d/v_+ = d/(E \cdot \mu_+)$. Here v_+ and μ_+ are the drift velocity and the mobility of positive ions, respectively. Combining the above-presented relationships, we get an expression for the space charge field:

$$E_\rho = \frac{i \cdot d}{S \cdot \varepsilon_0 \cdot E \cdot \mu_+} = \frac{i \cdot d^2}{S \cdot \varepsilon_0 \cdot U \cdot \mu_+}, \qquad (*)$$

where U is the voltage applied to the gap. Values for mobility μ_+ were taken from the literature [8]. The mobility varies with the pressure, p, as $\mu_+ = \mu_{+0}(p_0/p)$, where μ_{+0} is the mobility at the normal pressure p_0. Due to the simplifications, the above formula $(*)$ overestimates the value of E_ρ. However, it enables us to determine the upper limit of the discharge current. In experiments, the current value was adjusted to satisfy the condition $E_\rho/E \leqslant 1\%$.

3.2. Other conditions

The spectra were recorded in the wavelength intervals of 335–338 nm and 390–395 nm with the scanning step 0.04 nm. The distance between the electrodes was varied from 0.15 to 5.0 mm. The pressure in the discharge chamber was varied over the region of 2–740 Torr. The air temperature was between 18 and 26 °C. The value of the current strength was set by adjusting the applied voltage U. The voltage was always kept some per cent below the breakdown voltage for given d and p. Figure 2 represents the values of U and Nd for which the spectra

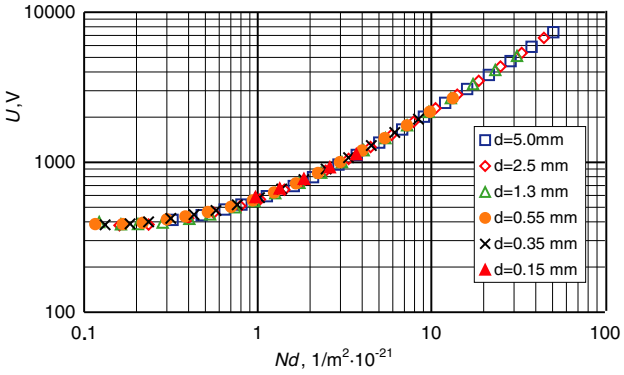

Figure 2. U–Nd region, where the spectrum was recorded.

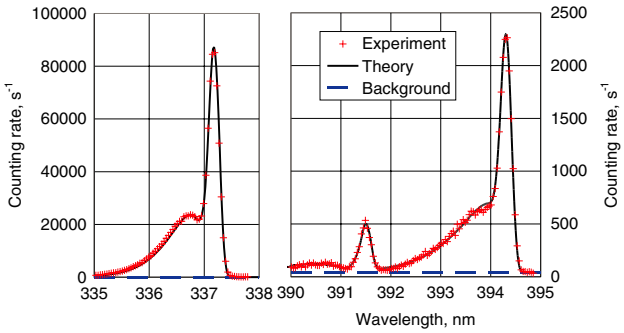

Figure 3. Spectrum of a non-self-sustaining discharge in dry air. $p = 13.2$ kPa, $d = 2.0$ mm, $E/N = 278 \times 10^{-21}$ Vm^2, $i = 1.4\,\mu$A. Points—measurements, black curve—calculated spectrum, dashed line—background.

were recorded. An example of the recorded nitrogen spectral bands is presented in figure 3. The SPS(0, 0) band is located in the wavelength interval of 335–338 nm. The FNS(0, 0) band (small peak—bandhead at 391.4 nm) together with the SPS(2, 5) band (bandhead at 394.3 nm) appears in the interval from 390 to 395 nm.

4. Data processing

If a value of the applied voltage was fixed, the current, i, and the radiation intensity, I, of the discharge changed slowly in time. It was possibly caused by reduction of the cathode emissivity and long-duration fluctuations in the intensity of the Hg lamp. These changes per day were always less than 10% and they were taken into account by scaling the recorded radiation intensity with the current: $I_{\mathrm{scaled}} = I\,(i_{\mathrm{mean}}/i)$, where i_{mean} is a current averaged over the time interval needed for recording of a spectral band. The radiation intensity of the discharge in the investigated spectral bands was proportional to the discharge current.

In order to compare the intensities of the selected bands, we first calculated simulated spectra—the intensity distribution in bands, $S_i(\lambda)$. Indices $i = 1, 2, 3$ correspond to 0–0 and 2–5 bands of SPS and 0–0 band of FNS, respectively. The formulae for these calculations and the values of the transition probabilities and molecular constants are taken from the literature [10–12]. The intensities of the rotational lines are normalized so that the sum of the intensities in a certain band is equal to one.

As a next step we calculate a normalized convolution integral (the experimental profile of the band spectrum):

$$\Phi_i(\lambda) = \int_{\lambda_1}^{\lambda_2} S_i(\lambda - x) F_U(x) \mathrm{d}x,$$

where $F_U(\lambda)$ is the instrumental function with unit amplitude. To match the calculated dependences $\Phi_i(\lambda)$ with the experimental points, coefficients α_i and β_i were introduced to denote the intensity of the ith band and its background in arbitrary units. Using the least-square method, the values of α_1 and β_1 were found such that the curve $\alpha_1\Phi_1(\lambda)+\beta_1$ was the best fit for the experimental points of the SPS(0, 0) band. In the wavelength region 390–395 nm, where the bands SPS(2, 5) and FNS(0,0) overlapped partially, the sum $\alpha_2 \cdot \Phi_2(\lambda) + \alpha_3 \cdot \Phi_3(\lambda) + \beta_{23}$ was fitted to the experimental points by choosing appropriate values for α_2, α_3 and β_{23}. Experimental points and curves $\alpha_1\Phi_1(\lambda)+\beta_1$ and $\alpha_2 \cdot \Phi_2(\lambda)+\alpha_3 \cdot \Phi_3(\lambda)+\beta_{23}$ are presented in figure 3 (solid curves). The dashed lines indicate the background levels β_1 and β_{23}. Finally, the intensity ratios $R_{391/337}$ and $R_{391/394}$ were calculated as $R_{391/337} = (\alpha_3/\alpha_1)K$ and $R_{391/394} = \alpha_3/\alpha_2$. Here the factor $K = 1.8$ takes into account the difference in sensitivity of our light detection system at wavelengths 337.1 and 391.4 nm. We did not take into account negligible changes in the sensitivity within the bands.

5. Results and discussion

The intensity ratios $R_{391/337}$ and $R_{391/394}$ are presented in figures 4 and 5 as functions of the reduced field strength E/N for different pressures. A rapid increase with E/N is a common tendency for both intensity ratios $R_{391/337}$ and $R_{391/394}$. In the pressure range of 13 to 100 kPa, the ratio R depends only slightly on pressure and all experimental points seem to lie on a single curve. At lower pressures a decrease in pressure leads to an increase of the ratio R. One can see that ratios R measured at different lower pressures lie on different curves (trendlines), which are parallel to each other in log–log scale (figures 4 and 5). The dependence of the intensity ratio on pressure is, in our opinion, caused by the difference in values of the collisional deactivation coefficients of the states $N_2(C\,^3\Pi_u, v = 0$ and $v = 2)$ and those of the state $N_2^+(B\,^2\Sigma_u^+, v = 0)$ (see table 1).

Therefore, the radiation intensities of the bands change differently with the pressure and, consequently, the intensity ratio of the bands is pressure-dependent.

If the spontaneous emission and the collisional quenching are the only processes of depopulation of an ith excited state, then the fraction of radiating molecules of that state, g_i, is a function of gas number density N:

$$g_i(N) = \frac{1}{1 + \tau_0 N (n_{N_2} k^i_{qN_2} + n_{O_2} k^i_{qO_2})}.$$

In this formula, $n_{N_2} = 0.78$ and $n_{O_2} = 0.21$ are the relative number densities of nitrogen and oxygen in air. Deactivation rate constants k_q are specific for the ith state.

The intensity ratio R, measured at gas density N, can be reduced to standard density N_0 as follows:

$$R_{i/j}\left(\frac{E}{N}, N_0\right) = R_{i/j}\left(\frac{E}{N}, N\right) \frac{g_i(N_0) g_j(N)}{g_i(N) g_j(N_0)}, \qquad (1)$$

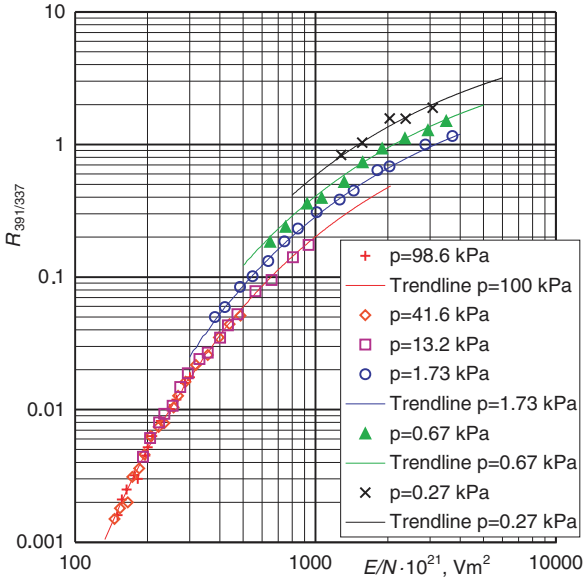

Figure 4. Intensity ratio of $R_{391/337}$ of nitrogen bands measured at different pressures as a function of reduced field strength.

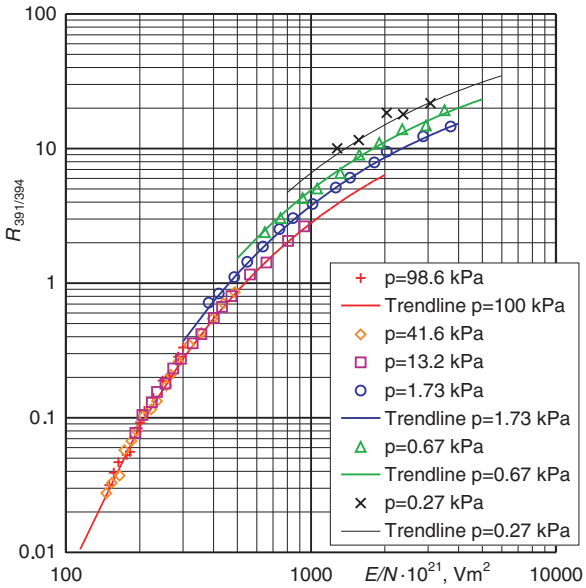

Figure 5. $R_{391/394}$ at different pressures.

Table 1. Radiative lifetimes and deactivation rate constants of some excited states of a nitrogen molecule and ion.

	States	Radiative lifetime τ_0, 10^{-9} s	Deactivation rate constants k_q, 10^{-10} cm^3 s^{-1}	
			k_{qN_2} for N_2 molecule	k_{qO_2} for O_2 molecule
1	$N_2(C\,^3\Pi_u, v = 0)$	42	0.13	3.0
2	$N_2(C\,^3\Pi_u, v = 2)$	39	0.46	3.7
3	$N_2^+(B\,^2\Sigma_u^+, v = 0)$	62	2.1	5.1

Note: data in the table are from the literature [13, 14].

where subscripts i and j stand for different excited states. Figures 6 and 7 represent our results obtained in the pressure range of 0.266–100 kPa and reduced to standard conditions according to formula (1). One can see that all the experimental

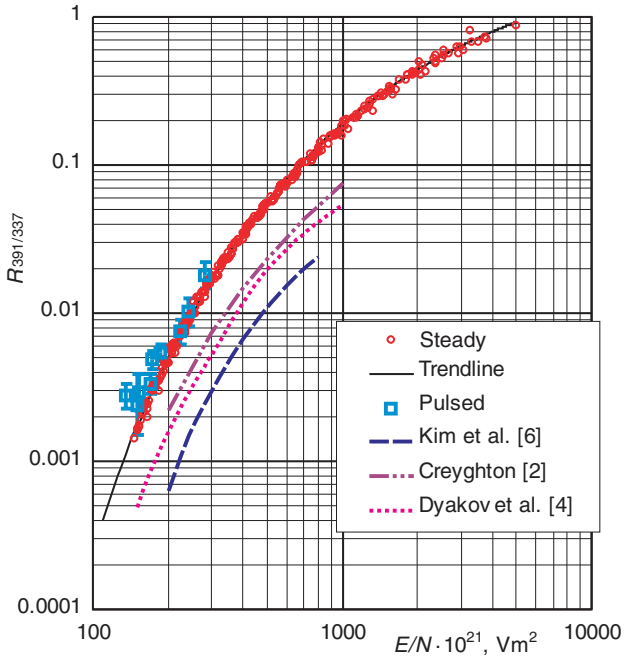

Figure 6. Intensity ratio $R_{391/337}$ of nitrogen bands reduced to standard conditions ($p = 100$ kPa and $T = 0$ °C). Data points—our experiment. Dashed curves—theoretical results of other authors.

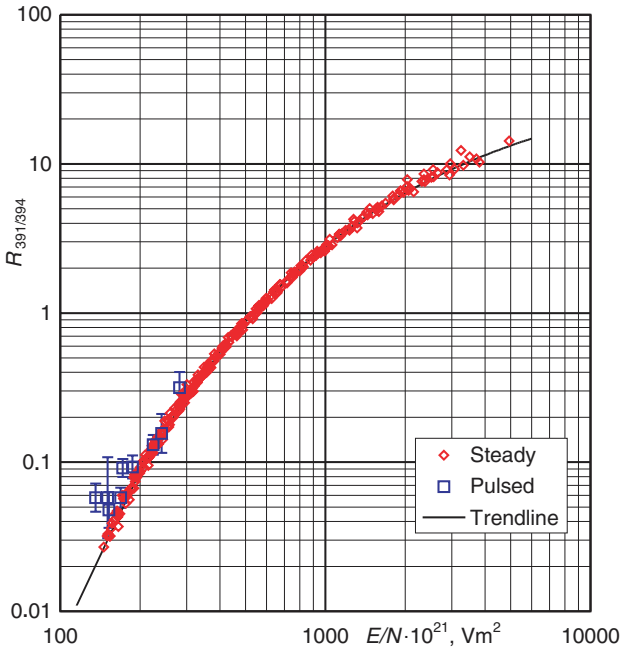

Figure 7. Intensity ratio $R_{391/394}$ reduced to the standard conditions.

points lie well on a single curve in those figures. The shape of the curve $R_{391/394}(E/N)$ is similar to that of $R_{391/337}(E/N)$, but the values of $R_{391/394}$ are about 17 times higher. Similarity in the shapes of $R_{391/337}(E/N)$ and $R_{391/394}(E/N)$ is expected because the general shape of excitation functions of states $N_2(C\,^3\Pi_u, v = 0)$ and $N_2(C\,^3\Pi_u, v = 2)$ is quite similar, and the difference between onset energies of excitation of these states is small compared with the difference of states $N_2(C\,^3\Pi_u, v = 0)$ and $N_2^+(B\,^2\Sigma_u^+, v = 0)$ [15]. From the

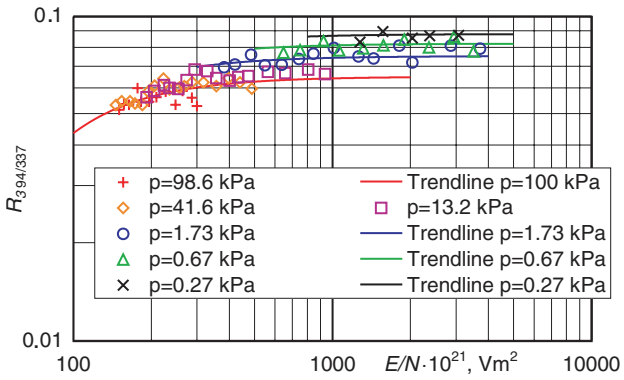

Figure 8. Ratio $R_{394/337}$ at different pressures.

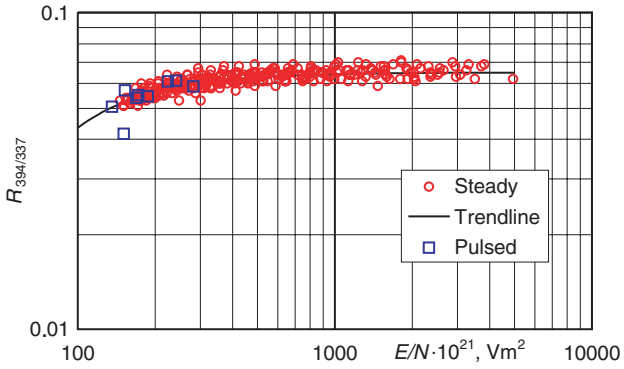

Figure 9. Reduced ratio $R_{394/337}$.

similarity of the shapes of $R_{391/394}$ and $R_{391/337}$ it follows that the ratio of SPS(2, 5) and SPS(0, 0) band intensities, $R_{394/337}$, should not change much with the field strength. This ratio for different pressures is presented in figure 8. In figure 9, $R_{394/337}$ is reduced to standard conditions.

The overall change of reduced $R_{394/337}$ with E/N does not exceed 20% and is close to our measurement uncertainty. However, the systematic increase of $R_{394/337}$ with E/N can be distinguished for lower values of reduced field strength.

The excitation mechanism of $C\,^3\Pi_u$ state in electron beam experiments was investigated by Cartwright *et al* [16]. They demonstrated that excitation of the electronic state $C\,^3\Pi_u$ occurs mainly due to the direct electron impact, although a small cascade contribution from the $E\,^3\Sigma_g^+$ state is possible. The cascade contribution remains within the experimental uncertainty for the lower vibration states [16]. Fons *et al* [15] have carried out a comparative study of the excitation in an electron beam experiment and in a discharge in nitrogen. They demonstrated that the relative population of the $v = 0, 1$ and 2 levels of the $C\,^3\Pi_u$ state in the discharge is nearly the same as in the electron beam experiments. The cascade makes only a minor contribution to the population of these levels. Thus, the excitation mechanism of levels $v = 0$ and $v = 2$ is the same. The similarity of the shapes of curves $R_{391/337}$ and $R_{391/394}$ versus E/N supports this conclusion also in our case. Therefore, we are of the opinion that the ratio $R_{391/394}$ can be used for the field strength determination as well as the ratio $R_{391/337}$.

Experimental points with uncertainty bars in figure 6 are taken from our previous investigation [7], where we measured

the same dependence of the intensity ratio at atmospheric pressure, but instead of the mercury lamp a pulsed KrF* excimer laser was used to liberate initial electrons from the cathode. In this case, the excitation of nitrogen occurred during a time interval of about 30 ns. Pauses between pulses were about 0.1 s. One can see that points obtained under the pulsed excitation lie on the same curve as points obtained under the continuous excitation. The last statement is valid for $R_{391/394}$ and $R_{394/337}$ as well (see figures 7 and 9).

We derived next empirical formulae for relationships between the intensity ratios and the reduced field strength E/N:

$$R_{391/394}\left(\frac{E}{N}, N_0\right) = 46\exp\left[-89\left(\frac{E}{N}\right)^{-0.5}\right], \quad (2)$$

$$R_{394/337}\left(\frac{E}{N}, N_0\right) = 0.065\exp\left[-402\left(\frac{E}{N}\right)^{-1.5}\right], \quad (3)$$

$$R_{391/337}\left(\frac{E}{N}, N_0\right) = R_{391/394}\left(\frac{E}{N}, N_0\right)\cdot R_{394/337}\left(\frac{E}{N}, N_0\right). \quad (4)$$

In formulae (2–4), the reduced field strength E/N is a dimensionless quantity that numerically equals the field strength expressed in units of $10^{-21}\,\mathrm{V\,m^2}$. In figures 4–9, trendlines for the standard conditions are calculated using these formulae. The trendlines for pressures different from the standard one are got using equation (1) in addition. The dispersion of experimental points around these trendlines does not exceed 10%. The measurement uncertainty of $R_{391/337}$ does not exceed 15% and that of $R_{391/394}$ 12%. The measurement uncertainty of E/N is less than 10%.

6. Conclusions

The dependence of the intensity ratio of the nitrogen bands on the field strength in air is determined experimentally in a wide range of E/N values for the first time. The experimental set-up is straightforward: it ensures an excitation of gas molecules under controlled conditions in an undistorted homogeneous field. The results of the present study allow an estimation of the reliability of theoretical calculations. In any case, the discrepancy between our results and theoretical calculations of cited authors exceeds the uncertainty of our measurements. Hence, a more thorough theoretical treatment of the problem is necessary.

The pressure-dependence of intensity ratios of examined transitions is due to differences in collisional deactivation rate constants of different states. Reduction of intensity ratios to standard conditions is possible if the deactivation rate constants are known. Reduced to standard conditions intensity ratio is a unique function of the reduced field strength. We deduced empirical formulae (2–4) for this function. These formulae can be used for estimation of E/N in low-temperature plasma studies in air if the excitation of molecules from the ground state by electron impact is the dominant process. This investigation demonstrates that in air the excitation mechanism of examined states of nitrogen is the same for pulsed and steady state conditions.

Acknowledgments

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