Short communication

Measurements and Analysis of Spatial Profiles of 777.4 nm Line in a Townsend Discharge in Oxygen

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Abstract

We have presented measurements of spatial profiles of electron induced emission coefficients for the 777.4 nm radiation of oxygen in low current discharges. Measurements were performed by the drift tube technique in low-current self-sustained Townsend type discharges. We have also compared results for the delay distance for electrons in oxygen calculated using Monte Carlo simulation code and results from a semi-empirical formula adjusted to fit the oxygen data. Finally, from the spatial profiles we have determined ionization coefficients. The spatial profiles of emission provide us also with information on heavy particle excitation and on the reflection of electrons from the anode. The absolutely calibrated spatial profiles of emission may be used to separate the effects of electrons and heavy particles in excitation at high *E/N* values and obtain the cross sections for fast neutral excitation.

Keywords: Oxygen, spatial profiles, ionization and emission coefficients, delay distance

1. Introduction

Principal lines used for diagnostics of low pressure non-equilibrium discharges in oxygen are atomic lines as molecular lines are relatively weak. Atomic oxygen emission mainly at the line 777.4 nm (transition O I $2p^3 3s$-$2p^3$ ($^4S^0$)3p) is also often used as an indication of the dissociation of oxygen in the studies of subsequent plasma chemical processes. Kinetics of emission of this line has not been covered sufficiently by the corresponding swarm studies especially at very high *E/N* and considering a possible role of heavy particles with the exception of the still unpublished work of Petrović and Phelps. Those measurements indicated that heavy particle excitation will be considerable for oxygen and have shown the need to analyze the process on a quantitative level but were never submitted for publication. In this paper we show data obtained in a completely different experimental system for spatial profiles of emission of the line 777.4 nm including direct absolute calibration. Those data show the evidence of heavy particle excitation. We also study the non-hydrodynamic development of electron swarms and ionization coefficients at high *E/N*. Preliminary report on this work was given in.[1]

Emission of the OI 777 nm line from oxygen has been used as a diagnostic tool in a number different discharges, most importantly in a number of different applicatrions of equilibrium and non-equilibrium both dc[2] and rf[3] collisional plasmas. Those applications include plasma ashing for production of integrated circuits [4] (and with increased use of the organic low k dielectrics[5, 6] there is a possibility of direct application in plasma etching), treatment of polymers and wool in particular[7] and the data for transport coefficients may be applied directly for modeling of atmospheric discharges such as coronas, where local field approximation may be applicable[8]. In particular diagnostics of oxygen in numerous applications is required as it has possible non-linear modes kinetics due to the presence of the low energy metastables[9].

Emission coefficient ε_m/N of level m is given by [10]

$$\frac{\varepsilon_m}{N} = \frac{S}{j_e} \frac{e}{\frac{\Omega}{4\pi} Q(\lambda) N \cdot \Delta x} \frac{1}{A_l \tau_m} \qquad (1)$$

where S is the emission signal, at the anode, of the line l, j_e is the electron current density at the anode, Ω is the effective solid angle of the detector, $Q(\lambda)$ is the quantum efficiency of the whole detection system, Δx is the width of the entrance slit of the monochromator, A_l is the Einstein's

coefficient for the line and τ_m is the radiative lifetime of the upper level *m*. In (1), we assume that collisional transfers from other excited states is negligible. The emission coefficient as it is usually defined[10] includes the branching ratio or in other words it should be associated with the upper level rather than the line itself as it gives the total emission from the upper level. The excitation coefficient requires analysis of the non radiative quenching and then it describes the total number of excitations to the level rather than emissions. It is customary to normalize the spatial profiles of emission to the absolute excitation or emission coefficients at the anode (where current is carried only by electrons).

In determination of absolute line intensities aimed at determining the excited state densities and at comparisons with calculated excitation efficiencies it is critical to consider spatial profiles of emission. Inevitably broad non-hydrodynamic region develops close to the cathode which makes it impossible to apply the assumed ionization coefficient in the entire gap between the electrodes. Calculation based on ionization coefficient is necessary in order to calculate the electron densities.

Thus a procedure[10] has been developed which separates the region into two, one with local equilibrium (with the local electric field) and the other which is non-hydrodynamic (or non-local) where excitation and growth are assumed to be zero. The width of the non-hydrodynamic region is known as the delay distance and is essential in absolute calibration of the excitation coefficients in low current discharge diagnostics.

2. Experiments

Measurements were performed in a drift tube that has been described earlier.[10–12] We have made measurements for oxygen. The drift tube consists of a pair of plane electrodes, with a diameter of 79 mm at a distance of 14.7 mm, placed inside a close fitting quartz tube. The cathode was made of stainless steel and the anode of graphite so that backscattering of electrons from the anode is minimized. The self-sustained Townsend discharge between 600 Td and 24000 Td (1 Td = 10^{-21} Vm^2) was maintained by running low current discharges at pressures between 290 Pa and 10 Pa, respectively. Discharge between the electrodes is sustained by a DC voltage. The light emitted from the discharge was detected through the quartz window of the vacuum chamber by using a photon-counting chain. Spatial profile was obtained by using 1 : 1 optics with 1 mm the width of the entrance slit of the monochromator mounted on a movable platform. Absolute calibration was achieved by using a standard tungsten ribbon lamp and corresponding properties of the discharge. Determination of absolute excitation coefficients will be the subject of an independent study.

3. Monte Carlo technique

We have used Monte Carlo simulations with electron O_2 cross sections from.[13, 14] The standard MCS code is the same as in.[15] A code for Monte Carlo simulation of electron motion adjusted to include boundary and non-equilibrium effects was developed, tested and applied.[15, 16] In the code we followed electrons released at the cathode until they reach the anode.

4. Results

In Figure 1. we show one example of spectra taken for oxygen for two values *E/N*.

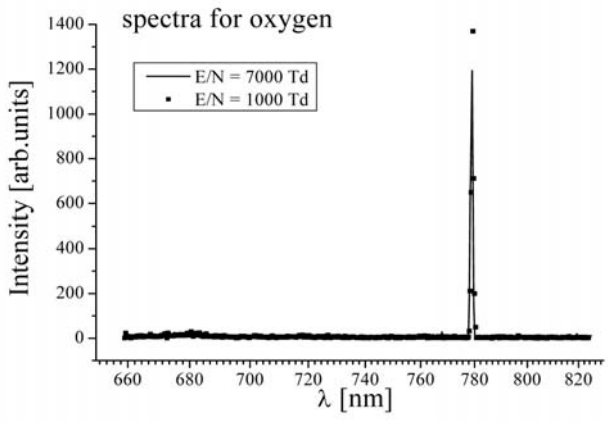

Figure 1. Example of spectra for oxygen obtained in a drift tube at current of 2 μA.

The profiles of emission of the 777.4 nm oxygen line obtained at low *E/N* and normalized to the excitation coefficient of the upper state at the anode and the Figure 2.

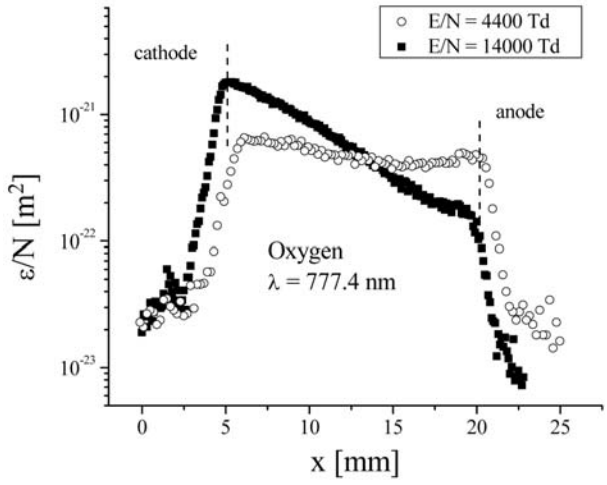

Figure 2. Spatial profiles normalized to the emission coefficients of 3p-3s O I (λ = 777.4 nm) for two E/N values.

We will label these plots as spatial profiles of the excitation coefficient to keep the terminology in line with that of Phelps and coworkers.[17] The actual value of excitation coefficient is constant in the region of the exponential growth and equal to the value at the anode.

From the slope of the spatial profile one may obtain the ionization coefficient provided that hydrodynamic stage has been achieved. The data obtained by this technique are perhaps not as accurate as those obtained by the pulsed current growth measurement but the present technique is easier to apply in a broader range of conditions and also at higher *E/N*. In Figure 3.we show a comparison between

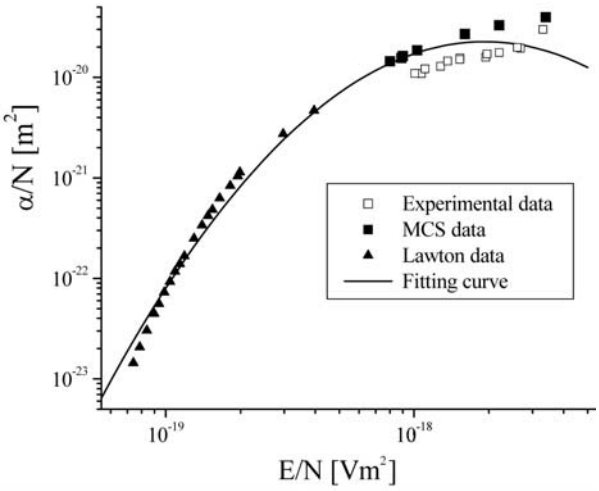

Figure 3. Comparison between experimental, Monte Carlo and Lawton data for ionization coefficients in oxygen[18, 19].

the experimental data from our measurements, present Monte Carlo simulations and the data of Lawton and Phelps [18, 19] for the ionization coefficients of electrons in oxygen. The experimental data are scattered since all the results obtained (at different intensities from three different lines) may be subject to the effects of heavy particle excitation that will reduce the slope except in a narrow region close to the anode. Thus the data should be regarded as a general check of consistency rather than accurate data. The Monte Carlo Simulations presented here were obtained by using a null collision code developed for high *E/N*[15] and the cross sections of[13, 14].

In order to describe the non-equilibrium we have used the delay distance d_0 which is roughly equal to the distance required to achieve the spatial growth of number density described by the equilibrium ionization coefficient. Method determination of the distance d_0 that was used in the present analysis includes fitting the spatial profiles of emission towards the anode and towards the cathode. Distance d_0[10] is obtained as a cross point of two straight lines that fit the recorded profile in the logarithmic scale; one- horizontal line that fits the non-hydrodynamic (not in equilibrium with the local field) region next to the cathode and the second that fits the exponential growth of emission which for the lowest currents appears to be exponential. In other words the value d_0 is the point where we may assume that the growth of the number of electrons may begin with the hydrodynamic value of the ionization coefficient α so that the total multiplication would be the same as in experiment. One may observe, in the spatial profile of excitation, a region next to the cathode where excitation is zero, than a gradual growth of emission and finally a growth with the hydrodynamic ionization coefficient. We extrapolated the hydrodynamic region to the zero value and that point determines the distance d_0 as applied in equation

$$\frac{i(d)}{i_0} = \gamma(e^{\alpha(d-d_0)} - 1) \tag{2}$$

for the breakdown which may be used to analyze the secondary electron yields (γ) from the breakdown voltage versus pressure times the gap distance.

In Figure 4. we compared results for the delay distance for oxygen calculated using Monte Carlo simulation code (solid symbols) and by semi-empirical formula (open symbols)[16] adjusted to fit the oxygen data.

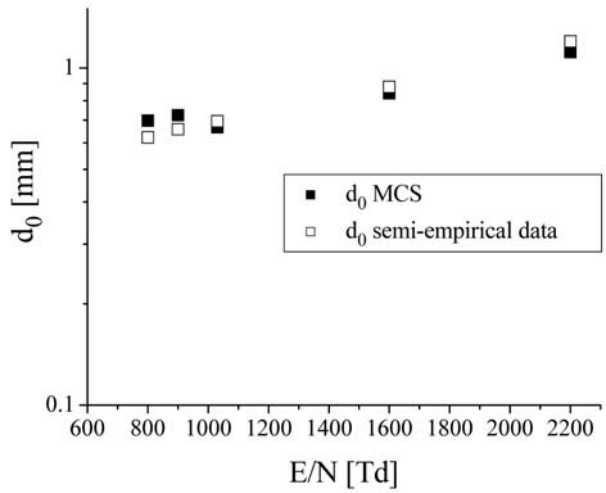

Figure 4. Comparison of delay distance for oxygen between Monte Carlo code and semi-empirical data.

It was not possible to make comparisons of d_0 with the experimental data because of still unexplained high level of emission close to the cathode which is similar to that found in N_2[15] and which covers the non-hydrodynamic region. Thus we regard results of simulations as more reliable. Our previous studies have indicated that in molecular gases a much better value of the delay distance is obtained from the Monte Carlo simulation than from the experiment.

5. Conclusions

We have presented measurements of spatial profiles, emission and ionization coefficients for the excited level of atomic oxygen leading to 777.4 nm emission which is the most prominent feature in the spectrum in non-equilibrium discharges. The range of *E/N* from relatively low all the way to very high was covered and therefore results show a range of features from equilibrium electron excitation to the fast neutral dominated excitation at very high *E/N*. The data are absolutely calibrated and may be used to separate the effects of electron and heavy particle excitation at high *E/N* values and obtain the cross sections for fast neutral excitation.

E/N values higher than 24000 Td could not be covered because of a rapid increase of breakdown voltage at low pressures. Values below 600 Td could not be covered due to oscillations that could not be damped by varying the external circuit elements.

6. Acknowledgements

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7. References

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Povzetek

Predstavljene so meritve emisijskih koeficientov kisika pri 777,4 nm, ob razelektritvah z nizkimi tokovi. Primerjali smo tudi rezultate za zakasnitveno razdaljo elektronov v kisiku dobljene s simulacijo Monte Carlo in s semi-empirično formulo, ugotovljeno na osnovi izmerjenih podatkov za kisik. Meritve smo izvedli s tehniko potovalne komore ob samodejni razelektritvi Townsendovega tipa pri nizkem toku. Potovalno komoro sestavlja par planarnih elektrod premera 79 mm, postavljenih na razdalji 14,7 mm, znotraj kvarčne cevi. Katoda je izdelana iz nerjavnega jekla, anoda pa iz grafita, tako da je povratno sipanje elektronov z anode minimalno. Samodejno Townsendovo razelektritev med $6 \cdot 10^{-19}$ Vm^2 in $2{,}4 \cdot 10^{-17}$ Vm^2 smo vzdrževali z nizkotokovno razelektritvijo pri tlakih med 290 Pa in 10 Pa. Absolutne emisijske koeficiente smo določili z meritvijo optičnega odziva na anodi. Prostorka porazdelitev emisije nam daje podatke o vzbujanju s težkimi delci, o nehidrodinamskem obnašanju razelektritve in o odboju elektronov z anode. Absolutni emisijski koeficienti in njihova porazdelitev omogočajo razločevanje med vzbujanjem z elektroni in težkimi delci pri visokih vrednostih E/N in določitev absorpcijskih presekov za hitra nevtralna vzbujanja.