Topic number: 5

Laser induced fluorescence spectroscopy of $N_2(A^3\Sigma_u^+, v = 0)$ and absolute density calibration by Rayleigh scattering

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The metastable $A^3\Sigma_u^+$-state of nitrogen molecule is characterized by high internal energy ($\sim 6\,\mathrm{eV}$), and therefore these metastables are of great importance in the gas discharge ignition and sustainment. Especially in spatially restricted atmospheric pressure discharges (e.g. DBD) the surface processes gain more influence, and the metastables may significantly contribute to the exoemission of electrons.

To determine the metastable density experimentally, the laser induced fluorescence (LIF) spectroscopy is applied as well known and established diagnostic tool for investigation of low pressure plasmas. For gas discharge plasmas at higher (atmospheric) pressure it is still a challenge because of the increasing de-excitation by collisional quenching and the small dimensions of these discharges.

Therefore, a capacitively coupled radio frequency discharge (CCP) at pressures up to 10 mbar was used to optimize the LIF method at the beginning, and afterwards to continue with dielectric barrier discharges at pressures from 100 mbar to 1 bar.

The CCP setup consisted of two parallel stainless steel electrodes at a distance of 10 mm powered via an rf source at 13.56 MHz at one electrode, whereas the other electrode was grounded. The dye laser, pumped by a frequency doubled Nd:YAG laser at 532 nm, was guided parallel to the electrodes and the fluorescence light was detected via a monochromator perpendicular to the laser beam. The laser excited the $A^3\Sigma_u^+, v = 0$ state to the $B^3\Pi_g, v = 3$ state at 687.4 nm, which afterwards fluoresced to the $A^3\Sigma_u^+, v = 1$ state at 762 nm. The electrical signal from the photomultiplier tube was integrated via a boxcar integrator and compared to the measured laser intensity.

For the determination of absolute number densities the measured time dependent LIF signal was analysed to calculate the original signal strength at the laser pulse without the delay resulting from the effective lifetime τ_{eff} of the excited state and the time constant τ_{PMT} of the photomultiplier circuit, respectively. Both delays result in a signal proportional to the difference of two exponential decays, which is exemplary shown in figure 1 for a pressure of 400 Pa.

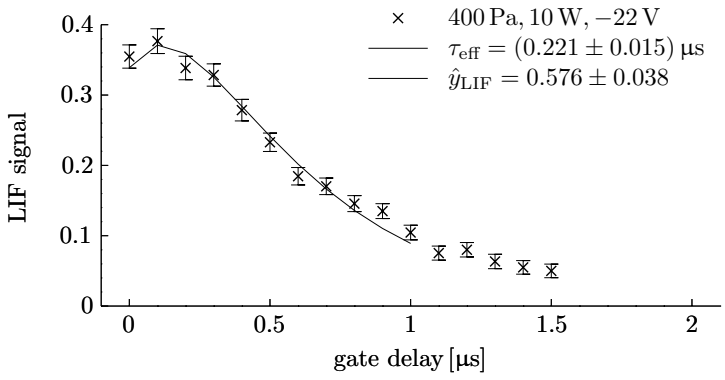

Fig. 1: Time dependent LIF signal for 400 Pa and the corresponding fit to determine the effective lifetime τ_{eff} and the original signal strength $\hat{y}_{\mathrm{LIF}}$.

By the use of a fitting routine we extracted the effective lifetime of the $B^3\Pi_g, v = 3$ state for the different pressures and got a dimension for the original signal strength $\hat{y}_{\mathrm{LIF}}$, if there would not be any delay due to the finite lifetime of the excited state and the delay by the photomultiplier circuit. Nevertheless, the decay rates τ_{eff}^{-1} depended not linear on pressure which indicates a more complicated quenching scheme than only de-excitation by collisions with the background gas. A comparison of the quenching rates from literature [1, 2] with the measured lifetimes showed also a big difference and emphasised the necessity of this procedure.

To get absolute densities we used the method from [3] which was a comparison of the LIF experiment with RAYLEIGH scattering measurements. Therefore, the different cross sections, their dependence on the direction of observation and the differences of the optical detection system at the fluorescing wavelength of 762 nm and the scattering wavelength of 687 nm were taken into account.

An example of the axial density distribution between the powered electrode at 0 mm and the grounded electrode at 10 mm is given in figure 2 for a pressure of 1000 Pa.

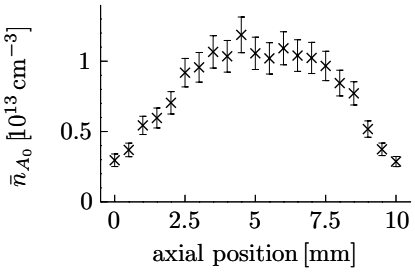

Fig. 2: Axial density profile of the $A^3\Sigma_u^+, v = 0$ state of nitrogen for 1000 Pa, 50 W and -53 V self-bias.

The density profile is, in spite of the electrical asymmetry, symmetric because of the small discharge gap of 10 mm and the still large lifetime of the $A^3\Sigma_u^+$ state, which results in the arbitrary destruction of the metastables at one of the electrodes. The absolute densities vary between 10^{12} cm^{-3} for 40 Pa and 10^{13} cm^{-3} for 1000 Pa, which is in good agreement with previous measurements at low pressure [3, 4].

Reference

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[2] S. De Benedictis, G. Dilecce and M. Simek, 1999, J. Chem. Phys. 110, 2947
[3] B. Krames, T. Glenewinkel-Meyer and J. Meichsner, 2001, J. Appl. Phys. 89, 3115
[4] S. De Benedictis, G. Dilecce and M. Simek, 1998, J. Phys. D: Appl. Phys. 31, 1197