Oxygen ³P atom and O⁻ ion density and atomic temperature in O₂ DC discharge obtained by Cavity Ringdown spectroscopy

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Abstract: We have used the 630nm ${}^{3}P_{2} \rightarrow {}^{1}D_{2}$ electric-dipole forbidden transition to determine the ground state O (${}^{3}P$) density and translational temperature in a pure O₂ DC discharge in a borosilicate tube. Using Cavity RingDown Spectroscopy (CRDS) we were able to enhance the very weak absorption. From the absorption baseline we were also able to determine the O⁻ ion density.

Keywords: O ³P density, Oxygen DC discharge, CRDS, atomic temperature, negative ion

1. Introduction

Pure O_2 plasma is commonly studied plasma which has applications in etching industries and plasma medicine. O atoms are a key species formed in O_2 plasma, especially for surface processing. It is therefore very important to know the absolute density of O (³P) to test models of O_2 plasma kinetics. Measurements by Two-photon Absorption Laser Induced Fluorescence (TALIF) calibrated against Xe [1], and actinometry with Ar [2] were inconclusive because of their indirect and complex nature (non-linear dependency on multiple parameters). The VUV O atom absorption lines at ~130nm cannot be used in many circumstances because measurements are often conducted under optically thick conditions.

Hence, we have used the weak electric dipole forbidden $^{630nm} {}^{3}P_{2} \rightarrow {}^{1}D_{2}$ transition. But for our system the absorption is very weak (~10⁻⁵) and so we needed cavity enhancement to increase signal-noise ratio [3]. In this case we used a cw tunable diode laser (Toptica DL100L, #1 in fig.1) to do Cavity RingDown Spectroscopy (CRDS).

2. Experimental setup

The positive column discharge (current 10-40 mA) in pure O_2 (0.2-7.5Torr) was established in a 56 cm double-walled (for constant wall temperature) borosilicate tube.



Figure1: Experimental setup for CRDS

The tube ends were enclosed by concave mirrors (#3 in fig.1) with high reflectivity (>99.995%) at 630nm located within adjustable mounts. The 1st order diffracted beam from an acousto-optic modulator (#2 in fig.1) is injected into the cavity, and switched off after the signal reaches a threshold. The position of the first mirror is constantly scanned by a piezo actuator, to allow excitation of individual longitudinal modes of the cavity. The transmitted signal was detected with an amplified photodiode. The 0th order beam is used to measure the wavelength with a wavelength meter.

3. Results and discussion

For single RingDown event the signal decays exponentially. From the exponential decay, we determine the RingDown time. 30-100 averages were taken for each wavelength point.



Figure2: CRDS spectrum at 1 Torr 40 mA with Gaussian fit

The absorption coefficient, *A* is calculated from the decay time in vacuum (τ_0) and with plasma (τ), from:

$$A = c^{-1}(\frac{1}{\tau} - \frac{1}{\tau_0}),$$

where *c* is the speed of light. An example absorption spectrum is shown in Fig. 2, along with a Gaussian fit. The integrated area gives the O atom density (in the ${}^{3}P_{2}$ level), the Doppler width gives the translational temperature. The O atom line lies on top of the O[°] photodetachment continuum, allowing us to also deduce the O[°] negative ion density.

4. Results and discussion

The variation of the absolute oxygen atom density with pressure and discharge current is shown in Fig 3.



Figure3: Absolute oxygen atom density as a function of pressure for different currents

The translational temperature of O atoms is shown in Fig 4. It consistently increases with pressure and current. These measurements are in good agreement with the O_2 b-state rotational temperature (from optical emission) and the O atom translational temperature determined by High-Resolution TALIF. Hence the gas particles in these conditions can be assumed in thermal equilibrium.



Fig. 4: Oxygen atom translational temperature.

The total gas density, N, can then be deduced from the pressure and the temperature allowing the mole-fraction, $[O(^{3}P)]/N$, to be calculated, as plotted in Fig 5. It increases almost linearly with current at all pressures studied, and passes through a peak at around 1 Torr pressure. At these pressures gas phase recombination is small, so that oxygen atoms are predominantly lost by recombination at the borosilicate glass tube walls. At pressures lower than 1 Torr the surface recombination probability is strongly increased due to by energetic ion bombardment [ref J P Booth et al. paper, submitted recently in PSST], leading to the observed sharp drop in O/N.



Fig. 5: Mole fraction, O/N ratio as a function of pressure for different currents

The O^{\circ} negative ion density is shown in Fig. 6. This is only a small fraction of the total gas density (<.001%), and comparable to the electron density. It passes through a minimum around 1 Torr, due to its destruction by associative detachment reactions with oxygen atoms.



Figure6: O⁻ (negative oxygen ion) density as a function of pressure and current.

5. Conclusion

CRDS is a powerful direct method to measure atomic density in oxygen plasmas. It also provides accurate measurements of the gas temperature, as well as the O^{\circ} negative ion density. These parameters are which are key for understanding and modeling of O₂ plasmas.

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7. References

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