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Filling the Gaps in Our Understanding of Plasmas in Simple Diatomic Gases- Combining a DC Plasma in Pure O₂ with Multiple Advanced Diagnostics for Experimental Validation of Simulations

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1. Introduction

Despite many decades of study, models of discharges in molecular gases still lack accurate data on many key collisional processes, even for such “simple” and ubiquitous gases as O₂. Good data is lacking for near-threshold electron-impact dissociation, surface recombination, the role of metastable molecules and atoms, of gas heating, of vibrational excitation, of energy transfer processes, and surface thermal accommodation. Ideally, the rate constants of these processes would be individually measured in dedicated experiments, but this is fastidious, if it is even possible. As an alternative approach, we compare comprehensive measurements of internal plasma parameters to simulations for a plasma with relatively simple chemistry, namely a DC positive column discharge in pure O₂. This well-characterized, stable and uniform discharge is optimal for experiment-model comparison. The excellent axial uniformity (uniform reduced electric field) is ideally suited to absorption spectroscopy techniques, which provide accurate absolute densities. Although oxygen plasmas have been studied for many years, new experimental methods, including synchrotron Vacuum UltraViolet (VUV) absorption spectroscopy and laser cavity ringdown absorption spectroscopy (CRDS), provide the densities of all the major species (atomic, molecular, in ground and excited states) with much-improved absolute accuracy. Furthermore, time-resolved measurements in (fully and partially) modulated discharges and with time resolution, allow the kinetics of reactive

species to be probed. The high spectral resolution of CRDS also allows the gas translational temperature to be determined. These comprehensive measurements provide unprecedented insight into the kinetic processes occurring both in the gas phase and at surfaces, and provide a profound test of the models. Whereas the unknown constants in models can generally be adjusted to fit a set of steady-state measurements for a given set of operating conditions, trends with pressure and discharge current, and especially the temporal response to current modulation, are much harder to reconcile with an incorrect model. In practice, model failures often result from simple omission of key processes, or from neglecting the effect of gas temperature on the reaction rates. Due to the relatively simple chemistry occurring in pure O₂, simultaneous measurements of all principal species, as well as the gas temperature, allows the missing reactions to be identified, and even estimation of their rates (and/or activation energies) by adjusting the model to fit the measurements.

2. Experimental

Experiments were performed in a well-characterised and reproducible DC positive column discharge in a temperature-stabilised (at 10 or 50°C) 2cm Ø borosilicate glass tube (internal radius 1 cm) [1]. The electrodes are situated in side-arms, allowing absorption measurements along the axis using wedged MgF₂ windows for VUV experiments, or dielectric

mirrors for CRDS. The electric field is determined from the potential difference between two floating high-impedance probes.

VUV absorption measurements were carried out at the DESIRS beamline at the French national synchrotron, SOLEIL. The Fourier-Transform spectrometer[2] gives spectral resolution up to 10^6 , but for time-resolved measurements it is necessary to use the monochromatic branch[3]. VUV absorption provides the densities of O_2 molecules in the ground (X)[4] and metastable (a, b [5]) states. The oxygen atom resonance transitions are also seen, but are so strongly saturated that they provide no useful information.

The density of ground-state oxygen atoms was determined from the weak, forbidden $^3P_2 \rightarrow ^1D_2$ transition at 630nm. The single-pass absorption of the order 10^{-5} is measured by cavity ring-down spectroscopy, using a cw single-mode diode laser and mirrors with reflectivity of the order 99.99% [6]. Longitudinal mode matching of the cavity to the laser is achieved by oscillating one mirror by piezo actuators. Since the ring-down events occur randomly in time, it is not possible to actively synchronize the measurement to the discharge modulation. However, the relative phase is recorded for each event, allowing full time-profile to be accumulated. In addition to the atom density, the Doppler profile of this transition is perfectly resolved, providing the gas translational temperature directly.

3. Results

An example of the time-resolved CRDS signals during full modulation of the discharge is shown in Fig. 1. The on-resonance curve shows the build-up and decay of the oxygen atom density during the pulse cycle. The jump on plasma extinction is caused by rapid gas cooling (causing the Doppler profile to narrow, and the gas density to increase). Surprisingly, the off-resonance curve is not flat; this can be attributed to O^- negative ion photodetachment (during discharge), and by ozone Chappuis-band absorption. Therefore CRDS provides time-resolved densities of these two species as well. A key parameter is the loss frequency of oxygen atoms due to recombination at the tube wall[1]. This was found to vary

according to the discharge history, a factor that must be allowed for carefully when comparing data sets from different diagnostic campaigns.

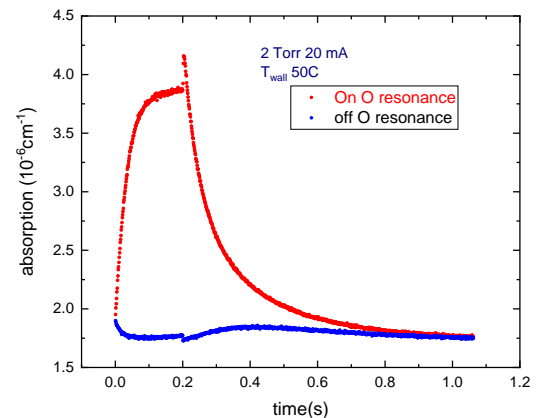


Fig.1 Time-resolved absorption measured by CRDS on the O atom resonance and off-resonance.

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