Chlorine atom and molecule dynamics in an inductively coupled plasma in pure Cl_2

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Two-photon laser-induced fluorescence (TALIF) was used to measure the absolute densities of Cl atoms and Cl_2 molecules with high spatial and temporal resolution in an inductively-coupled plasma in pure Cl_2 for pressures ranging from 2 to 50 mTorr and RF power (13.56 MHz) of 20 to 500W. The TALIF method of Ono et al. was used to detect the atoms, but a new scheme was developed to put the densities on an absolute scale, using 355nm laser photolysis of Cl_2 gas to produce a well determined density of Cl atoms. Firing the photolysis laser during plasma operation produces an increase in the TALIF signal proportional to the (local) Cl_2 density, allowing this quantity to be measured. In this way the total gas density (nCl + nCl2) was determined, and found to me considerably below the gas density without plasma, indicating that the gas is heated to high temperatures in the plasma.TALIF was also used to monitor the decay of the Cl atoms in the afterglow of a pulsed plasma, allowing the Cl surface recombination on Al_2O_3 to be studied.

1. Introduction

Two-photon laser-induced fluorescence (TALIF) is a powerful technique to measure relative atom densities with high spatial and temporal resolution. Niemi et al. [1] proposed a scheme for putting TALIF measurements of H, O and N atoms on an absolute scale by comparing the signal to that generated from a known density of a rare gas (Kr or Xe). Unfortunately in the case of Cl there is no TALIF transition in any rare gas at a close wavelength, so a such a scheme cannot be used. We have developed an alternative technique based on photolysis of a known density of Cl_2 molecules at 355nm. The Cl_2 absorption cross-section at this wavelength is known with high precision ($15.9 \times 10^{-24} m^2$), and photon absorption leads to dissociation of the Cl_2 molecule with 100% yield, producing two Cl atoms with a velocity of $1700 ms^{-1}$. Using an unfocussed tripled Nd:YAG laser for the photolysis provides a well characterised photolysis beam with a diameter much greater than that of the TALIF laser focus, so that the Cl_2 dissociation fraction can be calculated with high precision. In addition Cl_2 molecule densities were determined by measuring the additional TALIF signal created when the 355nm laser is fired during plasma operation. Finally, decays of the Cl density in the afterglow were measured by firing the TALIF laser at variable delays into the afterglow of a pulsed plasma.

2. Experimental

The plasma was produced in a cylindrical chamber (hard anodised Al, 10cm high, diameter 55 cm) excited by an external planar spiral coil at 13.56 MHz through an alumina window. Cl atoms were excited by two photons at 233.2 nm (supplied by a pulsed Nd:YAG pumped dye laser with frequency doubling) focussed at the reactor centre, and detected by 726 nm fluorescence as described by Ono et al.[2]. For the calibration, the reactor was filled with Cl_2 gas at room temperature (without plasma). The counter-propagating 355nm photolysis beam overlaps the TALIF beam at the reactor centre. We calculate that at the photolysis beam centre the photolysis yield is 6.7%, so that the Cl atom density created is 13.4% of the pre-laser Cl_2 molecule density).

3. Results and discussion

Fig 3 shows the Cl density as a fraction of the Cl_2 density (before plasma). This fraction first increases with power, then saturates. The highest fraction is observed at the lowest pressure, reaching 20% at 50mTorr 500W. Increased RF power causes faster electron-impact dissociation of Cl_2, but also causes significant gas heating, lowering the total gas density at constant pressure, which is the principal cause of the saturation well below 100%.

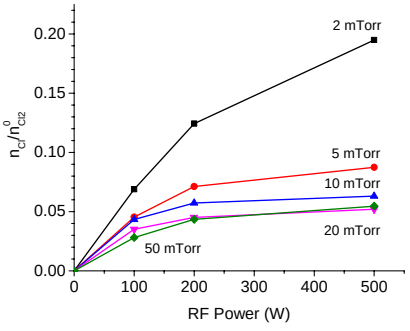

Fig 1. Cl TALIF signals from the plasma and from laser photolysis

3. References

[1] Niemi K, Schulz-von der Gathen V and Dobele H F *Journal of Physics D*- 2001 **34** 2330-2335

[2] Ono K, Oomori T, Tuda M and Namba K *Journal of Vacuum Science & Technology A* 1992 **10** 1071-1079