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Time Resolved Spectroscopy of RF Plasmas: A Comparison of Hydrogen and Deuterium Plasmas

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Abstract. Spatio-temporal resolved emission is a powerful technique for exploring the neutral, ionic and electron characteristics of rf driven plasmas. Here this is illustrated by measurements of emission from capacitively coupled H₂ and D₂ rf driven plasmas. These exhibit two pronounced emission regions, one associated with the normal sheath edge and the other near the driven electrode. Time-resolved electrode and plasma potential measurements show this near-electrode emission is associated with field reversal in the electrode sheath. The close similarity of the emission structure in the two isotopes, and the general lack of such structure in He plasmas indicates that the structure and hence field reversal are not solely due to ion mass effects but may also involve the details of the electron collisionality of the gas.

1. INTRODUCTION

Visual observation of a typical rf plasma provides the most obvious plasma diagnostic tool. For example, from an observation of change of the colour or the spatial variation in intensity experienced operators can diagnose a vacuum leak or a change in the character of a surface. Often such colour or intensity changes occur immediately after ignition or after a change of operating conditions or substrate. However the emission of light is a complex process depending on the detailed chemical and physical nature of the plasma and so can reveal much more detailed information. The abundance of atomic and molecular species in the ambient gas and its plasma products in the ground and excited states influence the emission. Most emission occurs as a result of collisions of the atom or molecule with an electron, so the electron density and energy distributions are important. Carefully designed experiments can therefore provide detailed information about these plasma species and parameters. Here the emphasis will be on the observation of plasma emission to study the behaviour of plasma electrons.

By observing only light at a wavelength specific to a particular constituent species and under conditions where it can be assumed that the density of that species is spatially and temporally uniform, the emission is indicative of the behaviour of the electrons exciting that species. In rf driven plasmas the electrons may be modulated with the frequency of the electrode voltage. In most technological plasmas this frequency is 13.56 MHz i.e. a period of 73.7 ns. Fast photon counting techniques allow emission intensity variation measurements with a resolution in the nanosecond region, permitting a study of the high energy portion of the electron energy distribution through the rf cycle. There have been many previous studies of spatio-temporal resolved measurements of emission from such plasmas [1-12]. We have exploited the fast gating (< 2 ns) capabilities of recently available ICCD detectors to study emission. Since the plasma can be imaged onto the detector array, this greatly facilitates spatially resolved measurements. In our previously reported measurements we have compared and contrasted spatio-temporal emission of H₂, He and Ar atoms [13, 14]. The most obvious difference in the excitation data from the three gases was the prominent double emission structure observed in H₂. Such emission structure has previously been reported in H₂ and H₂-containing gas mixtures [6, 7, 10, 12-17].

A number of mechanisms to explain the inner near-electrode emission layer have been advanced. These include electrons propagating from the electrode [10] and electric field reversal for a portion of the rf cycle [15]. The latter involves the presence of structure in the sheath electric field i.e. a change in electric field direction over a certain region. Several authors [18-20] have reported measurements of field reversal within rf sheaths in Ar and He but not specifically associated with double emission layers. Many rf sheath models predict a change in electric field direction when the sheath is contracting [18, 21-25]. While in general little attention has been given to these features they were recently seen as evidence of sheath reversal and subsequent plasma heating during sheath contraction in an rf driven nitrogen plasma [24]. Therefore we additionally measured the time dependence of the electrode-plasma potential difference correlated with the time dependence of excitation through the rf cycle [13,14]. These measurements showed that, in contrast to He and Ar, in H_2 the electrode goes positive with respect to the plasma i.e. there is a period during the rf cycle when electrons are accelerated towards the electrode. This electric field reversal is at that phase of the rf cycle where the near electrode emission in H_2 occurs.

The physical process that leads to this electric field reversal is not yet clear. We felt our measurements supported a postulate that it arose from the relatively high electron collision rate in hydrogen, others have associated this effect with the lighter ion mass of hydrogen or the presence of negative ions. In preliminary ion mass studies we noted almost identical behaviour in H_2 and D_2 [13]. In an attempt to throw further light on this issue here we report a more detailed study of plasmas with the two hydrogen isotopes.

2. EXPERIMENTAL DETAILS

Figure 1: A schematic diagram of the apparatus

A schematic diagram of the plasma and optical emission systems are shown in Fig. 1. The present measurements were made in the UK GEC (Gaseous Electronics Conference) rf reference reactor [26]. This is a parallel plate device with two 102 mm diameter electrodes separated by 25.4 mm. The upper, aluminium electrode and the stainless steel vacuum vessel were grounded. The plasma was created by capacitively coupling rf voltages at 13.56 MHz, to the lower, stainless steel electrode. In the present

measurements peak-to-peak rf voltages of up to 1200 V were applied producing plasma input powers of up to 150 W and discharge currents of up to 3 A. The operating pressures, ranging from 33 to 133 Pa (250 to 1000 mTorr), were measured using a capacitance manometer.

Time resolved light emission measurements were obtained by imaging the central plasma onto an Oriel Instruments Instaspec V fast-gated integrating charge coupled device (ICCD). A lens with a focal length of 200 mm and aperture f 4.5, placed 250 mm from the ICCD and 1.1 m from the plasma centre was used. This imaged a vertical section of the plasma region, which included both the electrodes, and a horizontal section which extended ~15 mm either side of the central axis. A 657 nm filter with a HWHM bandpass of 11 nm was used to select the 656 nm Balmer- α line of H_2 and D_2 .

The depth of field of the image represented 18 mm of the central plasma region. Vignetting of the image by the electrodes was a small effect in the present measurements. The collection angle of the lens was effectively constant except within 0.5 mm of either electrode, dropping linearly to about 80% of the central value at the electrode. The spatial resolution was 0.5 ± 0.1 mm, determined by comparison with a test image.

The ICCD gate was triggered by the output of a delayed pulse generator which was triggered by a pulse derived from a reference waveform from the matching unit. Software automatically incremented the pulse delay by 2 ns to record data for the next gating step, and the process was repeated until 75 delay steps (150 ns) had been completed, i.e. two complete rf cycles. The time and space resolved images were accumulated within 120 s. The accuracy of the gating pulse with respect to the phase of the matching network reference signal was determined to be ± 0.3 ns.

The time variation of the light emission intensity $I(t)$ was measured but it is the time dependence of the excitation $E(t)$ which reflects the time-dependent behaviour of the electrons, $I(t)$ is related to $E(t)$ via a convolution integral [2]. Here the measured emission signal $I(t)$ is deconvoluted with the radiative lifetime of the excited state (15 ns) to give the excitation $E(t)$. In hydrogen we neglect quenching which introduces at most an uncertainty of 1 ns in timing [10, 16]. The self quenching rate for deuterium will be slower.

The time varying electrode potential relative to the plasma was obtained from dc and ac measurements of the electrode and the plasma potentials [13]. The time-averaged and ac components of plasma potential were measured using a passively compensated Langmuir probe and a capacitive probe respectively. The Langmuir probe was also used to measure time-averaged plasma parameters such as electron density and temperature. The dc component of the electrode potential was recorded directly from the rf matching unit. We used a derivative probe and shunt to determine time resolved electrode potentials (time resolved plasma currents were also recorded by this technique).

To reduce noise our instruments employ averaging over a number of cycles for these optical and electrical measurement techniques so cycle to cycle variation would not be apparent. Never the less single shot measurements of the ac component of electrode current and potential shows only random cycle to cycle variation of ~ 1% with no significant contribution from sub harmonics.

3. RESULTS AND DISCUSSION

The Langmuir probe measurements indicate that the electron density in H_2 plasmas varied from 2×10^9 to $3 \times 10^{10} \text{ cm}^{-3}$ for the present pressure and power conditions. The measured electron densities in D_2 were generally about 50% lower. The kinetics of hydrogen plasmas are complicated and no self-consistent numerical model exists for rf plasmas. For example the plasma contains three types of positive ions H^+ , H_2^+ and H_3^+ but their relative concentration has, to our knowledge, not been measured or calculated. While H_2^+ is created directly by electron impact ionisation of the H_2 molecules they can be rapidly converted to H_3^+ by

which has a very large cross section for low energy H_2^+ ($> 3 \times 10^{-15} \text{ cm}^2$ for $E < 0.1 \text{ eV}$ [27]). Since the dissociation fraction in capacitively coupled plasmas is relatively low ~ 0.005 [28] and protons are produced mainly from the direct ionisation of atoms they are probably the minority ionic species. The densities of the three ionic species are strongly coupled and will vary with plasma conditions and with

isotope. Models of H_2 glow discharges have to date assumed a single H_2^+ species [12]. This lack of information makes it difficult to accurately determine the ion response to the rf fields. Using the measured electron densities and assuming the diatomic ion is dominant then the ion plasma frequency is expected to vary from 9 MHz to 33 MHz in H_2 and from 4 MHz to 16 MHz in D_2 . If the triatomic ion is dominant then the values are 5 MHz to 20 MHz in H_2 and from 3.5 MHz to 13 MHz in D_2 .

Figure 2. Spatially and temporally resolved Balmer alpha excitation from a hydrogen plasma operating with a forward directed rf power of 50 W and gas pressure 250 mTorr (top) and 100 W and 1000 mTorr (middle). The temporal dependence of the driven electrode potential relative to the plasma under the same conditions (bottom).

Figure 3. Spatially and temporally resolved Balmer alpha excitation from a deuterium plasma operating with a forward directed rf power of 50 W and gas pressure 250 mTorr (top) and 100 W and 1000 mTorr (middle). The temporal dependence of the driven electrode potential relative to the plasma under the same conditions (bottom).

The space and time resolved excitation data for Balmer-alpha excitation typical of those obtained in H_2 and D_2 are shown in Figs 2 and 3 respectively. The threshold energy for exciting this state from the H_2 ground state is 16.6 eV. This is probably the dominant mechanism since the dissociation fraction is low.

The results for both H_2 and D_2 are very similar. While the potentials are slightly different the near electrode excitation is seen to occur after the electrode is at its least negative with respect to the plasma. The strong positive peaks in the electrode current waveforms occur at this time. In both cases the electrode is positive with respect to the plasma, indicating a sheath collapse. This is a period during the rf cycle when electrons are accelerated towards the electrode, causing excitation of, and subsequent emission from, gas atoms in the near electrode region. This is an important observation since it appears to invalidate the argument that the near-electrode emission is an ion mass effect. The ion mass effect has been studied in fluid and PIC-MC models[12]. These correctly predicted the emission structure in hydrogen, using H_2^+ as the dominant ion. No calculations have to date been made for D_2 . Helium would have the same or intermediate ionic mass to D_2 depending on which ion state is dominant. The models shows no evidence of near-electrode emission which is largely consistent with our measurements in He[14]. In our helium measurements the only indications of near-electrode emission are at lower pressures and highest powers where associated sheath collapsed is also observed in our potential measurements.

The close similarity of the two emission region structure in the H_2 and D_2 , and general lack of such structure in He plasmas indicates that the structure and hence field reversal are not solely due to ion mass effects but also involves the details of the atomic structure of the gas.

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