SPATIAL AND TIME CHARACTERISTICS OF LOW TEMPERATURE RF PLASMAS FOR MATERIALS PROCESSING

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1 INTRODUCTION

Radio frequency (rf) plasmas are utilized in many applications in material processing, such as semiconductors and integrated circuits fabrication and surface modification by etching or coating. Plasma processing has replaced older techniques, such as wet chemistry, because the latter could not provide the necessary characteristics as process demands increased. A good example of this is the reduction of the feature size in semiconductors. The present critical dimension for semiconductor processing is 0.8 μm and is anticipated to be ≤ 0.25 μm by the year 2000. Semiconductor manufacturing needs ensure that each wafer going through the same treatment as much as possible. This requires investigations for control plasma diagnostic development. Although there has been a rapid growth in the application of rf discharge plasma in the last decade, knowledge of processes in the rf plasma is still far from satisfactory.

Optical emission spectroscopy have been employed for characterizing the low temperature rf discharge. By detecting light from the electronic transitions of atoms and molecules it is possible to identify and monitor the different processes in the plasma without perturbing the process. Furthermore, this technique could provide immediate feedback for automatic adjustment of individual process parameters.

The number of studies investigating the spatially and time varying optical emission from rf plasmas at frequencies near 13.56 MHz are limited. De Rosny et.al. [1] have observed the time dependent emission from a short lived state of Si in a pure SiH₄ rf plasma. Donnelly
and coworkers [2] reported the effects of frequency on the time varying optical emission from chlorine containing rf discharges. Bletzinger and De Joseph [3] investigated the temporally resolved optical emission from a nitrogen plasma as a function of position between the rf electrodes. More recently Tochikubo et. al. [4,5], Kokubo et. al. [6] and Köhler et. al. [7] reported spatially and temporally resolved optical emission data for neutral and ionic transitions that occur in argon plasmas. Tochikubo et. al. [4,5] calculated the relative net excitation rates by deconvoluting the optical emission profile, Kokubo et. al. [6] developed the diagnostic technique of the rf discharge by improving the spatiotemporally resolved optical emission spectroscopy, while Köhler et. al. [7] performed Boltzmann calculations to derive excited state populations for comparison with the time modulated emission intensity. Petrović et. al. [8] space and time resolved emission spectroscopy applied to obtained the information on the kinetic of rf discharge. Recent modeling of low pressure rf glow discharges has begun to predict many of the time varying aspects of high frequency rf plasmas [9 – 13]. However, the presently available spatially and time resolved optical emission data is insufficient to fully validate the conclusions derived by plasma modeling, either because the data do not cover an adequate range of experimental conditions or because the experimental conditions are not adequately defined. Additionally, the experimental data presently in the literature consist only of relative emission intensities, except Tochikubo and Makabe [14], which do not allow for comparison with calculations of absolute excitation rates or absolute population densities.

The emphasis of the present experimental work is therefore to measure spatially resolved optical emission intensities and the absolute time and spatially resolved optical emission intensities from ”well defined” argon plasmas over a wide range of pressures (6.7 to 133 Pa) and applied rf voltages (75 to 200 V). All experiments have been performed on a GEC RF Reference Cell [15]. This is a parallel plate discharge chamber with 102 mm diameter electrodes separated by 25 mm. The top electrode is grounded and the bottom electrode is powered by a 13.56 MHz rf power supply isolated with a blocking capacitor. Results of these measurements are presented for both an atomic transitions and an ionic transitions.

The optical emission studies have been performed in National Institute of Standards and Technology, Gaithersburg, USA, RF Plasma Laboratory.
2 EXPERIMENTAL

In the experiments presented here, the plasma source was the Gaseous Electronic Conference (GEC) Radio Frequency Reference Cell. The details of the design of the cell are given in Refs. 15 and 16. Briefly, the GEC RF Reference Cell is a parallel plate discharge chamber with 102 mm diameter electrodes separated by 25 mm. The electrodes are cylindrically symmetric and their surfaces are horizontal. The top electrode contains 169 holes with 380 μm diameters to provide a showerhead gas inlet. The cylindrical vacuum chamber is constructed of stainless steel and has 8 radially looking cooper gasket side ports at the chamber midplane. Two 203 mm diameter ports are fitted with 136 mm diameter quartz windows for spectroscopic observations. Two additional ports, orthogonal to these, are 152 mm diameter flanges, one of which accommodates a turbo molecular pump for establishing a base pressure of $< 10^{-5}$ Pa. Four 70 mm diameter ports at 45° with respect to the four larger ports are also mounted at the cell midplane. The bottom of the vacuum chamber is constructed so the pumpout of the gas is accomplished by four symmetrically placed 70 mm diameter ports piped into a single exhaust line to a mechanical vacuum pump.

The top electrode is grounded to the chamber on the outside of the vacuum interface. The bottom electrode is powered by a 13.56 MHz rf power supply isolated either with a 0.1 μF blocking capacitor for the time resolved measurements or with a filter box in external circuit for spatially resolved measurements. The filter box is Π network with two 1.85 μH series inductances, 100 pF input and output shunt capacitances and 150 pF middle shunt capacitance, designed as in Ref. 17.

For the data presented here, whole flow rates were 10 standard cubic centimeters per minute (sc cm). Gas pressures were 6.67, 13.33, 33.33, 66.67 and 133 Pa and the peak-to-peak applied rf voltages was 200 V.

The experimental setup for spectroscopic measurements is shown schematically in Fig. 1a and 1b. The spectroscopic apparatus consist of a 1 m focal-length monochromator with a 2400 lines/mm grating (0.416 nm/mm reciprocal linear dispersion). The monochromator is fitted with a quartz prism predisperser at its entrance slit to eliminate scattered light and multiple order effects in the observations. The vertical monochromator entrance and exit slits are 50 or 100 μm wide and 2 mm
Figure 1: Schematic diagram of the GEC RF Reference Cell and experimental arrangement high. The monochromator is also equipped with a cooled 5 cm diameter end-on photomultiplier for detection of the optical emission signal. There is a retracable mirror near the monochromator exit slit so a He-Ne laser may be substituted for optical alignment purposes. The photon flux produced in the plasma is focused on the entrance slit of the monochromator by a combination of three flat mirrors (M1, M2, M3), each 152 mm in diameter and one concave mirror with 650 mm focal length (M4) with the same diameter. These mirrors are arranged to act as a periscope bringing the level of the plasma emission to the same height as the monochromator entrance slit. This arrangement also rotates the image of plasma by 90° producing a spatial resolution of 0.5 mm vertical and 5 mm horizontal. By this rotation the electrode surfaces are imaged parallel to the long dimension of the entrance slit, thus permitting observations close to the electrode surfaces. Because of the periscope, vertical scanning of the plasma emission between the electrodes can be accomplished by translating mirror M2, while horizontal scanning is accomplished by translating the optical table (Fig. 1a). For
absolute intensity measurements the standard lamp was also mounted on the optic table.

3 SPATIALLY RESOLVED OPTICAL EMISSION MEASUREMENTS

For spatially resolved measurements, the photomultiplier tube is connected to a picoameter (Fig. 1a). The signal from the picoameter is fed to a strip chart recorder through an analog interface and to personal computer through IEEE-488 interface. The $Ar I$ 750.4 $nm$ and $Ar II$ 427.8 $nm$ lines were observed in the pressure range of 6.67 to 66.67 $Pa$ and 200 $V$ applied rf voltage. The time averaged intensity distributions along the discharge axis for $Ar I$ 750.4 $nm$ line are shown in Figure 2. The measurements are performed in pure argon rf discharge [18]. The intensity is not symmetric to the midpoint of the discharge axis because the top electrode is grounded and the bottom electrode is powered. Position of 0 $mm$ in Fig. 2 corresponds to the bottom electrode and position of 25 $mm$ corresponds to the top electrode.

The measurements of optical intensity distributions of $Ar II$ 427.8 $nm$ are shown in Figure 3. These measurements are taken under same con-
The intensity of both \( \text{Ar I} \) and \( \text{Ar II} \) lines are highly dependent upon the discharge pressure. As the pressure is increased from 6.67 to 66.67 Pa, the most intense regions of emission shift toward the powered electrode and the formation of well defined sheaths (dark zones near the electrode surfaces) are observed. The bright band of emission is excited by electrons propagating into the discharge from the electrode [1,3]. The propagation distance is a function of the electron velocity, which in turn depends on the electron collision frequency. Changes in pressure therefore will change the distance in which the exciting electrons can propagate away from the electrodes. This effect one can see in Figs. 2 and 3.

Analysis of the spatial variation in the optical signal intensity indicates that is bright band of emission peaks, for a given pressure, closer to the powered electrode in the \( \text{Ar II} \) emission than for the \( \text{Ar I} \) emission. This is more evident in Fig. 4 where the normalized time averaged intensity distributions of the \( \text{Ar I} \) and \( \text{Ar II} \) lines are shown. This Figure also indicates that for all plasma conditions the \( \text{Ar II} \) emission is spatially broader than the \( \text{Ar I} \) profile, with the \( \text{Ar II} \) emission extending closer to both electrodes.

To investigate the effect of changes in plasma parameters on the radial
Figure 4: Normalized, time averaged optical emission spatial profiles for Ar I 750.4 nm (solid) and Ar II 427.8 nm (dashed) lines. The applied rf voltage was 200 V and the gas pressures were (a) 6.67 Pa, (b) 13.33 Pa, (c) 33.33 Pa and (d) 66.67 Pa. The position d = 0 corresponds to the surface of the powered electrode.

Ar emission profile, the horizontal distribution of neutral Ar line at 415.9 nm was observed in a pure Ar plasma as a function of pressure. The plasma was scanned at the electrode midplane in a direction parallel to the electrode surfaces by moving the optical table supporting the spectroscopic apparatus horizontally. Because of side on observations, the measured intensity of the plasma emission contains contributions from many different plasma layers. In order to separate the contributions of the different layers and to obtain real radial distribution of the plasma emission the Abel inversion procedure [19] was used. The normalized signal intensity of the plasma emission as well as the Abel inverted results are presented in Figure 5. The extent of the electrodes is also indicated for comparison. As can be seen the Abel inverted profiles show a...
substantial difference from the horizontally scanned data as the pressure increases. Also the plasma extends outside the electrode region. These results indicate that uniformity of the plasma across the electrodes is seen to be fairly constant at low pressures, but becomes distinctly nonuniform at high pressures.

4 TIME RESOLVED OPTICAL EMISSION MEASUREMENTS

The time varying output from the photomultiplier was recorded with a system utilizing a time-to-amplitude converter (TAC) and a multi-channel analyzer (MCA). A detailed schematic diagram is shown in Figure 6. The timing cycle of the TAC is initiated by a fast pulse derived by a 200 MHz discriminator from a 10:1 voltage probe attached to the bottom of the powered electrode. The timing cycle is stopped by a photon initiated pulse from the photomultiplier system, or by the ending of the preset TAC timing period. For the data presented here, the maximum time measured by the TAC was set to 200 ns (slightly less
than 3 rf cycles). The photon count rates were kept low (< $10^5$ counts/s) by placing calibrated neutral density filters in the optical path for the measurement of the most intense optical signals. Because much less than one photon is detected, on the average, for each timing cycle, there is no discrimination against photons detected later in the timing cycle. The output pulses from the TAC, whose magnitudes are proportional to the time between the trigger from the voltage waveform and the detection of the photon, are sorted into channels by the MCA. This accumulated spectrum represents the time resolved optical emission signal for a single spatial location. Measurements were made at 10 – 15 different positions between the electrodes, depending upon the degree of spatial variation.
in the signal intensity. The MCA was set to record 256 channels providing a timing resolution of 0.78 ns/channel (200 ns/256 channels). The time history of the Ar I 750.4 nm and the Ar II 434.8 nm emission line signals were recorded with respect to the same time on the voltage waveform, therefore their relative phases may be compared. The Ar I line has an atomic lifetime of approximately 20 ns while the Ar II line has an atomic lifetime of approximately 7 ns [20]. These atomic lifetimes are less than the 13.56 MHz period (73.3 ns) and this fact make possible the measurement of time dependent optical emission. Examples of correlations between the measured time dependent optical emission signals and the voltage and current waveforms are shown in Fig. 7 for argon plasma with peak-to-peak rf voltages of 200 V and

![Figure 7: Measured time dependent optical emission signals and the corresponding voltage and current waveform with applied rf voltage of 200 V and pressure of 133.3 Pa. (a) non-calibrated optical emission of the Ar I 750.4 nm line, (b) non-calibrated optical emission of the Ar II 434.8 nm line, (c) (dotted) measured voltage waveform and (solid) calculated voltage waveform, (d) (dotted) measured current waveform and (solid) calculated waveform. The measured voltage and current waveforms were obtained simultaneously with optical emission data in (a) and (b).]
gas pressure of 133.3 Pa [21]. Figure 7a shows the raw $Ar I$ optical emission signal as recorded by the MCA at a point where the emission signal was a maximum (21 mm from the grounded electrode). The observation time was 3 minutes and a neutral filter with 0.6% transmission was used to reduce the count rate to acceptable levels. The raw data for the $Ar II$ line is shown in Fig. 7b for the same plasma at an observation point 22.5 mm from the grounded electrode (also the location of the maximum emission intensity). The observation time for the less intense $Ar II$ line was 5 minutes and no transmission filters were used. The dotted curves in Fig. 7c and 7d show the voltage and current waveforms, acquired simultaneously by the digital oscilloscope with the data shown in Figures 7a and 7b. The solid curves represent the voltage and current waveforms at the surface of the powered electrode as calculated by the equivalent circuit model [22,23]. The presence of higher harmonics in the electrical waveforms is indicated by the non-sinusoidal character of the curves.

The optical emission profiles for the $Ar I$ and $Ar II$ lines, as shown in Fig. 7 are quite different. The peaks in the time varying $Ar I$ optical emission signal are broader than in the $Ar II$ profile and exhibit an asymmetry on the right hand side of the temporal peaks. These characteristics are due to the relatively long atomic lifetime of the neutral excited state. This also contributes to the significant “background” or non-time-dependent portion of the $Ar I$ signal observed in Figure 7a. In contrast, the $Ar II$ signal profile exhibit narrow temporal peaks and a large time dependent signal due to the short lifetime of the excited state as compared with rf period. Comparison of the optical emission data in fig. 7 also indicates that the $Ar I$ and $Ar II$ profiles are not in phase with each other, with the $Ar I$ profile leading the $Ar II$ profile. The $Ar II$ time dependence is observed to be nearly 180° out of phase with the applied voltage, as was previously reported by Tochikubo et al [4]. Due to the non-sinusoidal nature of current waveform it is difficult to draw correlations between the current and the optical emission signals.

The optical emission signals as a function of observation point and time are shown for a range of plasma conditions in Fig. 8 for the $Ar I$ line (750.4 nm) and in Fig. 9 for $Ar II$ line (434.8 nm). Data were taken for argon pressures of 6.67, 13.33, 66.67 and 133.33 Pa and uncorrected applied peak-to-peak rf voltage of 200 V [21]. Figures 8 and 9 are plots of contour fits to the calibrated optical emission time profiles taken at many observation points between the electrodes.
Figure 8: Optical emission measurements of the spatial profile and temporal evaluation of the Ar I 750.4 nm line. The applied rf voltage was 200 V and the gas pressures were 6.67 Pa, 13.33 Pa, 66.67 Pa and 133.3 Pa.
Figure 9: Optical emission measurements of the spatial profile and temporal evaluation of the Ar II 434.8 nm line. The applied rf voltage was 200 V and the gas pressures were 6.67 Pa, 13.33 Pa, 66.67 Pa and 133.3 Pa.
A time range of 150 ns is shown (~ 2 complete rf cycles), with each square of the grid corresponding to approximately 4 ns on the time axis and 0.5 mm on the d axis. The time \( t = 0 \) was chosen to correspond to the maximum of the voltage waveform. The d axis is defined as the position of the observation point as measured from the grounded electrode along an axis through the center of the rf electrodes. The vertical axis on the plots is the absolute spectral radiances measured from the discharge. The number at the top of vertical axis is the magnitude of the maximum observed signal. For the purpose of the absolute intensity measurements a tungsten ribbon filament standard lamp calibrated for spectral radiance (± 2 %) by the Radiometric Physics Division, NIST [24] was mounted on the optics table. This lamp is used to calibrate the optics/monochromator system to obtain absolute spectral radiance measurements. The lamp was substituted for the plasma source by rotating mirror M1 (see Fig. 1a) to image the lamp filament onto the entrance slit. The calibration measurements were made with the same slit width and photomultiplier voltage as used in the plasma measurements. The intensity of the standard lamp signal was stronger than the observed spectral lines by approximately a factor of 10^3. To prevent the measurement system from saturating, metallic film neutral density filters were placed in the optical path in front of the monochromator slit for measurement of the lamp signals. To avoid both, a second order radiation from the lamp and a significant component of the signal from scattered light, bandpass interference filters were incorporated into the optical path also. The bandpass filters were used only in the standard lamp calibration procedure, and not in the plasma experiment. The metallic film neutral density filters and the bandpass filters could not be characterized using this optical monochromator system, and were therefore individually calibrated for transmission over the wavelength region of interest. The total uncertainty in the measurement of a spectral line radiance is therefore estimated to be ± 6 %.

Two complete cycles are shown in each plot (Figs. 8 and 9) to help in determining the significance of minor features in the contour plots. If a feature is not repeated in both cycles, then it is not reproducible and should not be considered to be real. This is particularly evident for the low pressure \( \text{ArII} \) data shown in Fig. 9 where the signal levels were low thus resulting in noisy data.
5 DISCUSSION

As is evident from Figs. 2 - 5 and Figs. 8 and 9 the spatial and temporal profiles of the optical emission are highly dependent upon the discharge pressure. At 6.67 Pa the spatial profiles are broad with the emission essentially extending across the entire region between the electrodes. Only a small fraction of the optical emission signal exhibits time dependent behaviour at these lower pressures. At higher pressures a larger fraction of the optical signal exhibit a time dependence, which is in agreement with previous results by Seeböck and Köhler [25] for lower frequency discharges.

The bright band of emission peaks are closer to the powered electrode in the $ArII$ emission than for the $ArI$ emission. Formation of bright bands near the grounded electrode is observed for only a few sets of plasma conditions. For the $ArI$ transition, a weak sheath is observed at 66.67 Pa with well defined sheath present at 133.33 Pa (see Fig. 8). If the applied voltage increases from 75 to 200 V the amplitude of the emission near the grounded electrode remains nearly constant while the emission near the powered electrode increases by a factor 2.5. This may be related to the fact that the voltage across the sheath near the grounded electrode is nearly independent of the applied rf voltage as shown by electrical measurements [22] and ion kinetic energy measurements [26]. For the $ArII$ emission, the existence of a bright band of emission near the grounded electrode was only observed for higher pressure (Fig. 9). For these conditions a weak increase in the optical signal is detected near the grounded electrode.

The changes in the temporal and spatial distributions with applied rf voltage are less dramatic than those observed with changing pressure. As the applied voltage increases, the peak emission intensity increases, particularly at the higher pressures. Additionally, the widths of the peaks in the time dependent signal increase with increasing voltage. This may indicate that conditions are favorable for the excitation of the required transitions for emission during a greater portion of the rf cycle at higher applied voltages.

6 CONCLUSIONS

Spatially and temporally resolved optical emission profiles have been measured for argon plasmas with pressures ranging from 6.67 to
133.33 Pa and applied rf voltage of 200 V. The plasma source was the GEC RF Reference Cell [15].

Spatially resolved optical emission spectra may be used to determine sheath thicknesses and to monitor the uniformity of the plasma across the electrodes.

The magnitude of the plasma emission for two lines has been put on an absolute scale by comparison with emission from a standard lamp. This allows for a complete characterization of the optical emission signal from the rf plasma, and provides a more comprehensive data set for comparison with theoretical modeling results.

While much can be learned from optical emission spectroscopy, more research must be done to determine the aspects of the data that are of most importance for monitoring the plasma conditions relevant to etching processes.

7 REFERENCES


