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Temporal Evolution of Gas Temperature in the Afterglow of Pulsed ICP Discharge Using Rayleigh Scattering

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Abstract: ***A Rayleigh scattering technique was adopted in the current work to measure for the first time (to our knowledge) the temporal evolution of the gas temperature in the afterglow of low-pressure pulsed ICP discharge operated with argon and Ar/O_2 (10 % O_2) gases. For all gas compositions used in this work and for averaged applied RF power of 100 Watt, the gas temperature was found to be constant in the afterglow time period having a value of 350 K. By using averaged RF power of 500 Watt, the trends of the temporal evolution of the gas temperature and their values were found to be a function of the gas composition and its pressure. For gas pressure of 20 mTorr of pure argon and Ar/O2 (10% O_2) admixture, the trends of gas temperature evolution were exponentially decaying with decaying time constant of 2.5 & 19 microseconds, respectively. For 100 mTorr of Ar/O_2 (10% O_2) a striking trend of the temporal evolution of the gas temperature was observed where the gas temperature was increasing in the afterglow period which might be ascribed to the increase of the electron temperature in the afterglow due to the transition of the deposited RF power from inductive mode to capacitive mode enhanced by vibrational –rotational deexcitation of molecular oxygen.***

Keywords: Rayleigh scattering, Pulsed discharge, Gas temperature, ICP discharge

1. Introduction

Inductively coupled plasma (ICP) sources can produce high-density and uniform plasmas in low-pressure gases without the need for external magnetic fields. Such sources are used extensively for thin film deposition [1],[2], etching [3]-[6] and for gas laser pumping [7]. Compared to conventional continuous wave (cw) ICP sources, pulsed ICP sources have already shown remarkable advantages and potential in plasma etching and deposition processes. By switching the discharge power on and off, the composition of radicals in the gas phase can be better controlled, leading to improved central selectivity in etching [8] and film composition in deposition [9], and damage induced by plasma processes can be suppressed or eliminated [10]. In plasma etching, less plasma induced damage, more anisotropic etching, and higher selectivity [3] can be achieved by properly selecting pulse parameters such as pulse shape [11], the duty cycle and the repetition rate [12]. To optimize the selection of these pulse parameters, accurate measurement of the time-resolved plasma properties (e.g. electron density & temperature and gas temperature) is often needed.

In plasmas; gas temperature can be measured by different techniques such as analyzing the emission of molecular bands (such as second positive system of N_2 [13]) or by analyzing atomic line profile to estimate Doppler broadening of the emitted atomic line in case of using atomic gases. The accuracy of the previous techniques is questionable since there are many factors should be put into our accounts in deducing the gas temperature. In the first technique there should be equilibrium between the rotational temperature and translation temperature of the gas which might not achieved in low pressure discharges. While in the second technique the effect of pressure broadening and instrumental broadening of the measured line profile should be estimated to deduce the correct Doppler broadening width. Laser Rayleigh scattering is alternative technique to measure the gas temperature with high accuracy. In literature there is few studies for measuring the gas temperature of ICP discharge operated with reactive gases such as O_2 and to my knowledge there is no study is found in the literature for measuring the gas temperature of low-pressure pulsed ICP discharge in the afterglow period.

In the current work, laser Rayleigh scattering technique is adopted to measure the gas temperature evolution in the afterglow of pulsed ICP discharge operated with argon and Ar/O_2 admixture. This article is organized as follows: after this introduction the basic of Rayleigh scattering thermometry is highlighted in section 2. Section 3 is devoted for the experimental setup. The results will be presented and discussed in section 4 while the most important finding of the current work will summarized in section 5.

2. Laser Rayleigh Scattering Thermometry

Laser Rayleigh scattering is an effective diagnostic tool for the determination of the temperature of gas mixtures providing the composition is known [14]-[16]. Rayleigh scattering is the elastic scattering of photons by heavy particles (atoms, ions and molecules). The inelastic scattering of photons by molecules, which can undergo a rotational or vibrational transition, is called Raman scattering. Rayleigh scattering has a simple origin: the electrons in atoms, molecules or small particles radiate like dipole antennas when an oscillatory electromagnetic field is applied. In a gas, the motion of the molecules leads to microscopic density fluctuations that randomize the phases of the secondary waves and causes the scattering to be incoherent in all but the forward direction. Away from the forward direction, rapidly changing interferences occur which average to remove any coherent effects resulting in a scattering intensity that, in the far field, is only proportional to the number of scatterers.

Providing the pressure and species concentrations remain constant or are known, the temperature may be determined from this signal via the ideal gas law.

The total Rayleigh scattering signal collected from a radiating volume for a laser beam of intensity I_I incident on a gas mixture can be written as:

$$P_{Det} = \eta I_I n V (\sigma_R)_{mix} \quad (1)$$

where η is the optical collection system efficiency, n is the mixture gas density, V is the observation volume and $(\sigma_R)_{mix}$ is the Rayleigh scattering cross section of the gas mixture given by the mole fraction weighted sum of all species present in the observation volume:

$(\sigma_R)_{mix} = \sum_i \sigma_{Ri} X_i$ with X_i is the mole fraction of the i^{th} species. In the current work, the operating gases are Ar or Ar/O_2 mixture. Rayleigh scattering cross sections for Ar and O_2 gases using laser wavelength of 532 nm are $5.4688X10^{-28}$ cm^2 and $5.3958X10^{-28}$ cm^2, respectively, which are almost identical and this means that the mole fraction of O_2 gas in Ar/O_2 mixture is not important to take into consideration when equation 1 is used.

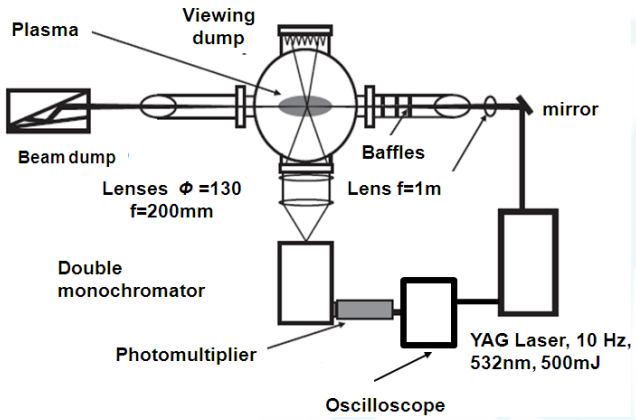

Figure 1: Schematic diagram of Rayleigh scattering experimental setup

In the current work, two Rayleigh scattering signals are measured. Firstly, a calibration point is obtained by measuring Rayleigh signal from the operating gas with known density at room temperature T_1 of 300 K before igniting the discharge and let this signal be P_{det} (T_1). Secondly, measuring Rayleigh signal for the discharge at unknown temperature T_2 (K) and let this signal be P_{det} (T_2) . Assuming the solid angle of collection is small and the entire width of the incident laser beam is included in the observation area, the signals so measured are given by:

$$P_{Det}(T_1) = \eta I_I n_1 V (\sigma_R)_{mix} \quad (2)$$

$$P_{Det}(T_2) = \eta I_I n_2 V (\sigma_R)_{mix} \quad (3)$$

where n_1 and n_2 are the number densities of the operating gas at room temperature T_1 and at the unknown temperature T_2 of the discharge. By using the ideal gas law one can show that:

$$n_1 k T_1 = n_2 k T_2 \Rightarrow T_2 = \frac{n_1}{n_2} T_1 \quad (4)$$

where k is the Boltzmann constant. By taking the ratio of Equations 2and 3 and making use of Equation 4 it is possible to write

$$T_2 = T_1 \frac{P_{det}(T_1)}{P_{det}(T_2)} \quad (5)$$

Special care was taken to ensure that the Rayleigh measurement was not interfered with by stray light.

3. Experimental Setup

The experimental setup is similar to that described in reference. [17]. A schematic diagram of the experimental setup to measure Rayleigh scattering signal is shown in figure 1. Briefly, a stainless steel discharge chamber of inner diameter of 310 mm was used. The discharge was generated and sustained by a planar, spiral, three-turn water-cooled copper coil antenna. The coil was located behind a 25-mm-thick quartz window. The discharge was maintained between the quartz window and a grounded stainless steel end plate, which were separated by 75 mm. All the measurements were performed midway between the quartz window and the stainless steel end plate. A pulse-modulated RF current of frequency 13.56 MHz was used to produce an inductive electric field that produces the discharge. The RF modulation frequency was 12.5 kHz and the duty cycle was 50%. The input power measured before the matching network was fixed at 100 and 500 W and the reflected power was maintained at approximately 5 W. The gas inlet was at the center of the chamber and the flow was controlled using a mass flow controller. Pure Ar gas (at a pressure of 20 and 100 mTorr and a flow rate of 132 sccm) or Ar/O_2 (10% O_2) mixture gas was used at a pressure of 20 & 100 mTorr at a flow rate of 132 sccm. The laser source was a Nd:YAG laser operated at a second-harmonic wavelength of 532 nm. The laser had a pulse energy of 500 mJ, a pulse duration of 10 ns and a beam divergence of 0.5 mrad, and was operated at a repetition rate of 10 Hz. The beam was injected into the chamber in a direction perpendicular to the chamber axis and focused by a 1000-mm-focal length lens that produced a focused beam inside the chamber with a diameter of approximately 1 mm. The scattered radiation was observed at an angle perpendicular to the chamber axis and laser beam. The scattered photons from the plasma were collected by a pair of lenses and focused onto the entrance slit (slit width of 0.3 mm and slight height of 4 mm) of a double monochromator, the output of which was detected by a photomultiplier. The photomultiplier was coupled to digital storage oscilloscope Tektronix TDS 2000C, which was synchronized with the Nd:YAG laser. The Nd:YAG laser was synchronized with a desired time in the afterglow using a delay generator connected to the output of the matching box. The measurements of scattered Rayleigh signals were obtained by registering those signals during scanning the double monochromator over wavelength range of 1.5 nm around the laser wavelength. In this scanned range, both of the laser Thomson scattering signals of the free electrons and Raman scattering signals of molecular gas is much weaker than Rayleigh scattering signals. Unlike laser Thomson scattering signals that measured in reference [17] Rayleigh scattering signal was obtained by accumulating a signal from 256 laser shots while it was 20000 laser shots in the case of laser Thomson scattering measurements.

4. Results & Discussion

To measure Rayleigh scattering signal with high accuracy the stray light signal must be measured first without any gas in chamber. Stray light is the scattering light from the windows and the edges of the chamber. After that the chamber should be filled with the operating gas at the desired pressure and a total signal which is the sum of Rayleigh signal and stray light is acquired at room temperature for calibrating the system. The Rayleigh signal at room temperature is then estimated by subtracting the stray signal from the total measured signal. Figure 2 shows the measured spectrum of stray signal at base pressure of 10^{-5} Torr (red circle symbols curve) and the measured spectrum of total signals (black square symbols) when the chamber was filled with 100 mTorr of Ar/O_2 (10% O_2) admixture. The spectrum of Rayleigh scattering signal was estimated (spectrum with blue triangle symbols of figure 2) by subtracting the spectrum of the stray signal from the spectrum of the total signal. All spectra that shown in figure 2 were fitted by Gaussian distribution and all of them had a width of 0.39 nm. Figure 3 shows a spectrum of Rayleigh scattering signal measured at 5 microseconds in the after glow of 20 mTorr Argon discharge at average discharge power of 500 watt. It can be seen from this figure that the spectrum was fitted well with Gaussian distribution of width of 0.39 nm which indicated that there was no Doppler broadening of the measured spectrum. The area under the curve of the spectrum that is shown in figure 3 and the area under the blue curve of figure 2 are representing both of P_{det} (T_2) and P_{det} (T_1) in equation 5 respectively, and this gives gas temperature of 420 K.

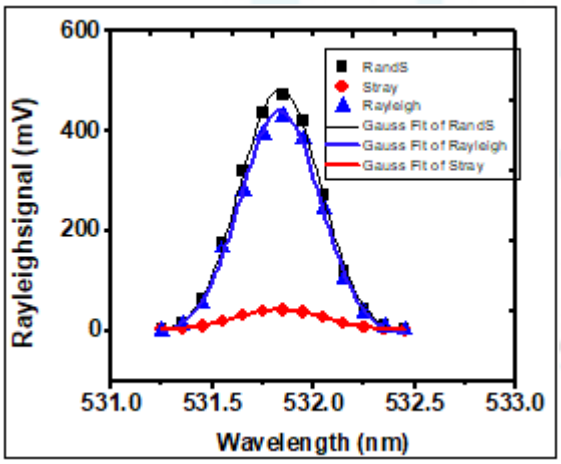

Figure 2: Spectra of total scattering signals (Rayleigh & stray light), stray light signal & Rayleigh scattering signal measured from 100 mTorr Ar/O_2 (10% O_2)

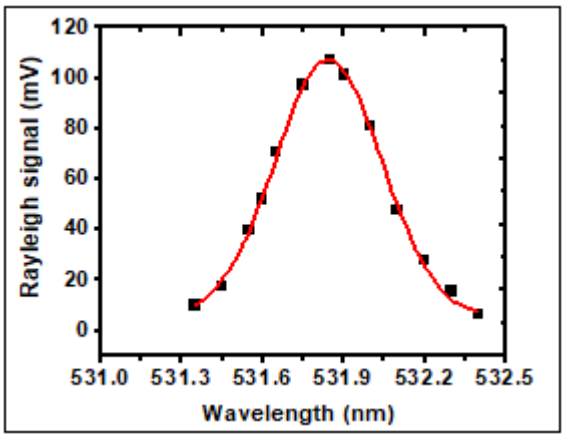

Figure 3: Spectrum of Rayleigh scattering signal measure at 5 microseconds in the afterglow of 20 mTorr argon plasma generated by applying RF power of 500 Watt.

4.1 Gas temperature of argon discharges

The temporal evolution of the gas temperatures of 20 mTorr argon discharge is depicted in figure 4 for two averaged RF powers of 100 & 500 Watt. The trend of the temporal evolution of the gas temperature in figure 4 is following the temporal evolution of the electron temperature measured by Langmuir probe in the same discharge condition where the electron temperature data was published elsewhere [18]. The trend of the temporal evolution of the gas temperature for 100 watt case is almost constant in the afterglow and equal to 350 K which is almost 50 K higher than the room temperature. For RF power of 500 watt case, the temporal evolution of the gas temperature in the afterglow is decaying exponentially from 520 K to 400 K in 7 microseconds with a decay time constant of 2.5 microsecond. This fast decay of gas temperature might be attributed to the decaying of metastable argon atoms density due to Penning ionization [18].

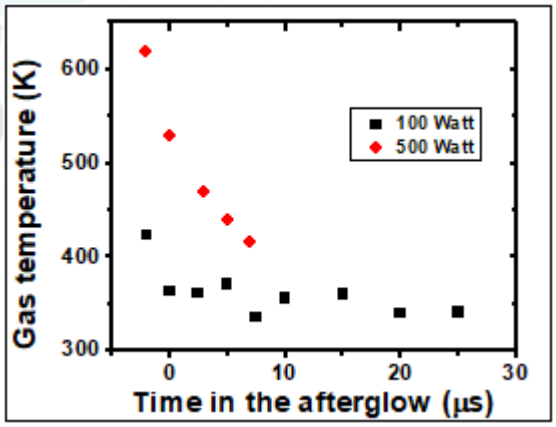

Figure 4: Temporal evolution of gas temperature of 20 mTorr argon plasma generated by applying an average RF power of 100 & 500 Watts.

The temporal evolution of the gas temperature of 300 Watt discharge operated with argon gas pressure of 100 mTorr is illustrated in figure 5. The trend of the gas temperature is monotonically decreased in the afterglow where the decrease of the gas temperature was about 70 K. The gas temperature of 300 Watt discharge operated with argon gas of 100 mTorr pressure is higher than the gas temperature of 500 Watt

discharge operated at lower argon pressure 20 mTorr and this might be ascribed to the higher electron temperature of discharge operated at higher gas pressure [18].

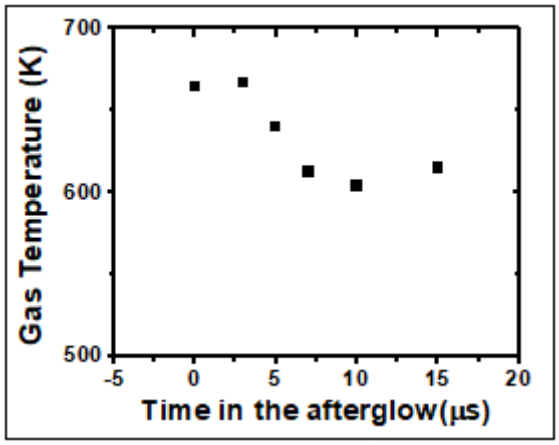

Figure 5: Temporal evolution of gas temperature of 100 mTorr argon plasma generated by applying an average RF power of 300 Watts.

4.2 Gas temperature of argon/oxygen discharges

In this subsection we will present the results obtained using a reactive gas admixture with argon gas as an operating gas. The operating gases admixture was Ar/O_2 (10% O_2) at pressure of 20 & 100 mTorr. Figure 6 shows the temporal evolution of the gas temperature of 20 mTorr Ar/O_2 (10% O_2) plasma generated by applying an average RF power of 100 & 500 Watts. The trend of the temporal evolution of the gas temperature at RF power of 100 Watt is the same as that shown in figure 4 for the same RF power where the gas temperature was found to be constant with value of 350 K. This finding indicates that at low RF power there is no effect of adding 10 % of O_2 gas. The trend of the gas temperature evolution in the afterglow when averaged RF power of 500 Watt was applied is exponentially decayed with decay time constant of 19 microseconds as can be seen from the exponential fitting that is shown in figure 6.

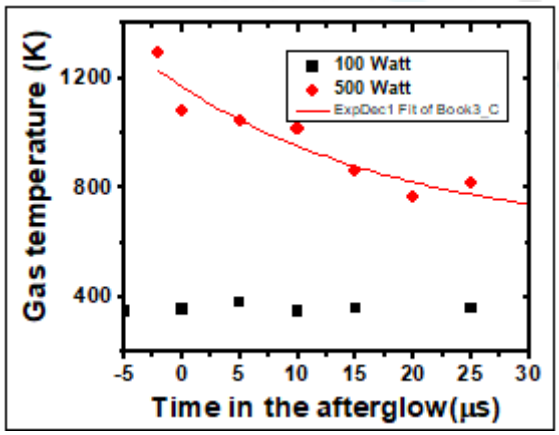

Figure 6: Temporal evolution of gas temperature of 20 mTorr Ar/O_2 (10% O_2) plasma generated by applying an average RF power of 100 & 500 Watts

The temporal evolution of the gas temperature measured when 100 mTorr Ar/O_2 (10% O_2) plasma generated by applying an average RF power of 500 Watt is depicted in figure7. From that figure it can be clearly seen that the gas temperature is monotonically increased in the afterglow which is a striking result. The gas temperature might be ascribed to the increase of the electron temperature in the afterglow, where a transition of the power deposition from inductive mode to capacitive mode is expected when a high averaged RF power is applied, enhanced by vibrational – rotational deexcitation of molecular oxygen [19].

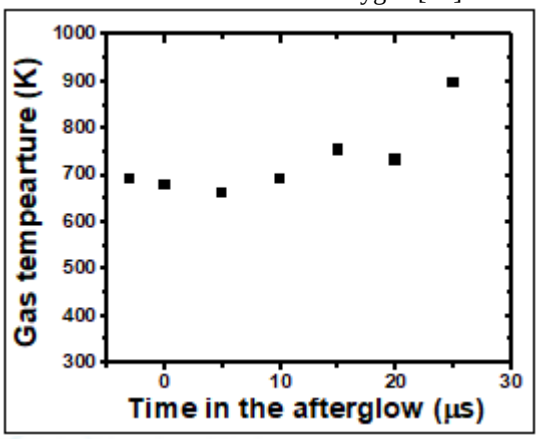

Figure 7: Temporal evolution of gas temperature of 100 mTorr Ar/O_2 (10% O_2) plasma generated by applying an average RF power of 500 Watt.

5. Conclusion

Rayleigh scattering system was constructed and used to measure the gas temperature in the afterglow of pulse modulated ICP argon and Ar/O_2 admixture plasmas generated using RF power range of 100-500 Watts. For low RF power condition typically averaged applied RF power of 100 Watt, the trend of the temporal evolution of the gas temperature is constant over the afterglow time and it had a value of 350 K which is 50 K above the room temperature.

By using averaged RF power of 500 Watt, the trends of the temporal evolution of the gas temperature and their values were found to be a function of the gas composition and its pressure. For gas pressure of 20 mTorr of pure argon and Ar/O_2 (10% O_2) admixture, the trends of gas temperature evolution were exponentially decaying with decaying time constant of 2.5 & 19 microseconds, respectively. For 100 mTorr of Ar/O_2 (10% O_2) a striking trend of the temporal evolution of the gas temperature was observed where the gas temperature was increasing in the afterglow period which might be ascribed to the increase of the electron temperature in the afterglow due to the transition of the deposited RF power from inductive mode to capacitive mode enhanced by vibrational –rotational deexcitation of molecular oxygen.

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