Laser Thomson Scattering, Raman Scattering and Laser-Absorption Diagnostics of High Pressure Microdischarges

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Abstract. Laser scattering experiments were performed in high pressure (100s of Torr) parallel-plate, slot-type DC microdischarges operating in argon or nitrogen. Laser Thomson Scattering (LTS) and Rotational Raman Scattering were employed in a novel, backscattering, confocal configuration. LTS allows direct and simultaneous measurement of both electron density \( n_e \) and electron temperature \( T_e \). For 50 mA current and over the pressure range of 300 – 700 Torr, LTS yielded \( T_e = 0.9 \pm 0.3 \) eV and \( n_e = (6 \pm 3) \cdot 10^{13} \) cm\(^{-3} \), in reasonable agreement with the predictions of a mathematical model. Rotational Raman spectroscopy (RRS) was employed for absolute calibration of the LTS signal. RRS was also applied to measure the 3D gas temperature \( T_g \) in nitrogen DC microdischarges. In addition, diode laser absorption spectroscopy was employed to measure the density of argon metastables (1s5 in Paschen notations) in argon microdischarges. The gas temperature, extracted from the width of the absorption profile, was compared with \( T_g \) values obtained by optical emission spectroscopy.

1. Introduction

Microdischarge plasmas or microplasmas are non-equilibrium gas discharges operating at pressures of hundreds of Torr and having dimensions of tens to hundreds of micrometers. Their discharge scaling parameter \( pL \) (\( p \) is gas pressure and \( L \) is characteristic dimension) is similar to that of larger plasmas. Microdischarges have many unique applications, such as UV light sources, plasma display panel cells, ozone sources, and biomedical applications.

Microdischarge diagnostics are important for understanding basic microdischarge operation, however, conventional methods (e.g., probes, microwave interferometry) are difficult or impossible due to the small size and high pressure. In contrast, optical diagnostics are convenient for characterization of microdischarges. In this paper, measurement of basic microplasma parameters (i.e., electron density \( n_e \), electron temperature \( T_e \), neutral gas temperature \( T_g \) and density of excited metastable species) using laser spectroscopy is reported. The techniques include Laser Thomson Scattering (LTS), Rotational Raman Scattering (RRS) and Diode Laser Absorption Spectroscopy (DLAS).

2. Experiment

A direct current microplasma source in symmetric slot-type parallel-plate geometry was chosen for investigation [1]. The interelectrode gap could be varied from \( d = 300 \) to 600 µm and the electrode...
surface area was 5 x 0.5 mm$^2$. In the rotational Raman scattering experiments the discharge was operated in pure N$_2$ (400–700 Torr) at typical voltages of 360 – 400 V between the electrodes at currents of 5 – 30 mA. In laser Thomson scattering experiments the discharge was in Ar. The discharge current was set at $I$ = 50 mA, resulting in a voltage between the electrodes in the range 310-350 V (as pressure was varied). Higher currents resulted in glow-to-arc transitions. A schematic diagram of the experimental apparatus is presented in Figure 1. The microdischarge was mounted on a holder that was connected to an x-y-z linear stage. This arrangement allowed the discharge to be precisely moved with respect to the stationary laser beam and light collection optics, so that 3D spatially resolved measurements could be carried out.

Figure 1. Schematic diagram of the experimental apparatus for laser scattering spectroscopy.

The pulsed, frequency doubled Nd:YLF laser had a wavelength of 526.5 nm, a spectral linewidth of 0.05 nm (FWHM), a pulse frequency of 3 kHz, a pulse duration of 100 ns, and an average power of 6 W. The laser beam was guided into a periscope that focused the beam into the microdischarge with a ~40 µm FWHM diam.

In most laser scattering experiments light is detected at an angle of $\theta = 90^\circ$ with respect to the laser beam axis [2-5]. In a slot-type microdischarge with a long, deep, and narrow gap, the z-axis [1] is the most appropriate direction of propagation of a tightly focused laser beam, as well as for detection of the scattered light. Therefore, a backscattering geometry (i.e., $\theta = 180^\circ$) was used. Since the laser beam did not pass through the objective lens and window used to view Raman scattering, the background radiation from scattering of the beam was minimized. Scattered light was collected by two lenses and focused on a 100 µm diameter pinhole spatial filter with 4X magnification. The transmitted light was then focused by a third lens (with 2X demagnification) onto the 100 µm entrance slit of the Triple Grating Spectrometer (TGS), similar to one used by van de Sande [2, 3]. The TGS provides very efficient spectral filtering of the scattered light at the laser (central) wavelength, while transmitting the light shifted with respect to the laser wavelength. Different combinations of spatial filters and entrance slits were evaluated for rejecting scattered laser light, while maintaining an adequate portion of the Raman spectrum with sufficient resolution. For rotational Raman spectroscopy, a wider spatial filter (750 µm) could be used since the Raman peaks were sufficiently shifted from the laser wavelength. However, the spectral width of the Thomson signal ($\propto \sqrt{T_e}$) is relatively small due to the low electron temperatures ($\sim$3.5 nm for $T_e$ = 1 eV), hence, a 500 µm-wide filter was the best compromise. With the laser stray light reduced by a factor $\sim 10^4$, the third grating dispersed the light over 10.9 pixels/nm on an intensified charge-coupled device (ICCD). The ICCD was gated ON for 300 ns, synchronized with the laser pulse, to reject most of the background emission from the plasma. Spectra accumulated over typically 100 s (3·10$^7$ laser pulses) were recorded and processed by a program that implemented a nonlinear least squares fit to the experimental data. Data points in the spectral range blocked by the spatial filter were not included in the fitting.

The synthetic Raman spectra were a convolution of the theoretical Raman spectrum (100 rotational levels of N$_2$ were accounted for) with the measured apparatus function [1]. The apparatus function was
represented by a 0.35 nm FWHM Gaussian. A second, broad Gaussian (2.7 nm FWHM) accounted for a relatively small background, perhaps from scattering off the optics or the face of the ICCD.

The synthetic Thomson spectrum was modelled by a Gaussian function, assuming Maxwellian electron energy distribution function (EEDF) [6]. Absolute calibration (required for extracting the absolute electron density $n_e$) was done using Raman scattering from nitrogen. Before and after a set of experiments, the chamber was evacuated and then filled with pure N$_2$. The calibration constant was found by fitting the Raman signal to a synthetic spectrum at room temperature.

The apparatus used for tunable diode laser absorption spectroscopy is shown schematically in Figure 2. An External Cavity Diode Laser (ECDL) in the Littman configuration working around $\lambda = 801.5$ nm was used as the light source. The width of the laser line (<10 MHz) was much smaller than the typical bandwidth of the absorption line (>1 GHz). Therefore, the absorption profile was recorded by scanning the laser frequency across the absorption line.

The laser beam from the ECDL passed through a beam splitter. Part of the beam was guided to a Fabry-Perot interferometer (1 GHz free spectral range) to perform in-situ calibration of the laser frequency. The rest of the beam was attenuated, to <100 µW, by neutral density filters, directed through the interelectrode space of the microdischarge, and then through an aperture. The aperture suppressed emission from the plasma by reducing the collection solid angle. A set of mirrors was used to magnify (11X) and image the laser light on a linear CCD array, which allowed spatially resolved (across the electrodes) measurement in a single acquisition. The experiment was controlled by LabVIEW software.

3. Results and Discussion

Rotational Raman scattering measurements were carried out in a pure N$_2$ microdischarge with the interelectrode gap of $d = 300$ µm. Figure 3 presents typical net spectra (with the background subtracted) recorded with the plasma ON (top) and OFF (bottom). Note that the background dominates only in the region close to the central (laser) wavelength (~3 nm wide), but it is small compared to the signal in the wings of the spectra. The background-dominated region was excluded from the fit. The gas temperature was determined by fitting the experimental spectrum with a synthetic (model) spectrum, with $T_g$ as the adjustable parameter. The net spectrum with the plasma OFF was fit first to obtain the proportionality constant relating the measured signal intensity to the relative computed intensity. The analysis also confirmed that the model correctly predicted the gas temperature which, for plasma OFF, was the ambient temperature. This same proportionality constant was then used to fit spectra recorded with the plasma ON. The good fit to the

$$ Ar(2p_4) \leftrightarrow Ar(1s_3^3P_2) \quad (\text{Einstein A coefficient} = 9.28 \times 10^6 \text{ s}^{-1} [7, 8]) \text{ at } \lambda = 801.48 \text{ nm was used for absorption measurements.}$$
observed plasma-ON spectra showed that N$_2$ was the dominant species with a number density that varied as $1/T_g$ at constant pressure, as expected.

**Figure 3.** Net (background subtracted) Raman spectra and best fits for $P = 500$ Torr: (Top) – plasma ON ($I = 20$ mA), best fit $T_g = 900$ K; (Bottom) – plasma OFF, best fit $T_g = 320$ K (ambient temperature).

Using this approach, spatially resolved $T_g$ profiles in N$_2$ DC microdischarges were obtained. The results of these measurements can be found in [1].

The LTS measurements were carried out in a pure Ar microdischarge with the interelectrode gap at $d = 600 \mu$m. An example of the net (background subtracted) Thomson spectrum is presented in Figure 4.

Despite the presence of noise, the Gaussian fit was reasonably good (note that the central part of the spectrum (near the laser line) was suppressed by the mask in the TGS and therefore it was excluded from the fit).

**Figure 4.** An example of net (background subtracted) Thomson spectrum. The red line is the best fit resulting in $T_e=1.0$ eV and $n_e=9 \times 10^{13}$ cm$^{-3}$.

Due to the rather long duration of the laser pulse (~100 ns), gating the ICCD did not provide sufficient suppression of the background plasma emission. The problem of background plasma emission also precluded the use of photon counting to improve the signal-to-noise ratio (the ICCD noise was relatively small compared to the plasma emission). These problems were compensated by signal averaging over a very large number (0.3 million) of laser pulses.

The electron temperature was extracted from the width of the Gaussian fit [6].

The values of $n_e$ and $T_e$ shown in Figure 4 are quite typical for this kind of microdischarge [9, 10] and are consistent with the predictions of a 1-D mathematical model, described in [10].

The $n_e$ data obtained by LTS were compared with those obtained by Optical Emission Spectroscopy (OES) [11] using Stark broadening of Balmer-$\beta$ emission resulting from H atoms due to the water impurities. Electron densities obtained by both techniques are compatible (see Figure 5).

**Figure 5.** Comparison of $n_e$ measurements obtained by LTS and OES.
Diode Laser Absorption measurements were carried out in a pure Ar microdischarge with an interelectrode gap of \( d = 300 \mu m \). Before every experiment, the dark current of the CCD array as well as the plasma emission (although it was highly suppressed by the aperture) were recorded. The measured intensity was automatically corrected by subtracting these background signals.

Figure 6. Profile of the ratio \( I/I_0 \) as a function of interelectrode distance and laser frequency (logarithmic scale).

Figure 6 shows the ratio of transmitted laser intensity with plasma OFF \( (I_0) \) and with plasma on \( (I) = I_0/I \) (on a logarithmic scale) – as a function of interelectrode distance and laser frequency. One can obtain spatially resolved profiles of the argon metastable density by integrating the profile of Figure 6 with respect to the laser frequency at different spatial locations.

Figure 7 shows the Ar(1s5) density profiles at different currents and pressures. The profiles are consistent with the properties of the cathode sheath and negative glow, where most of metastable production occurs. The peak metastable density increases with increasing pressure, but decreases with increasing discharge current. The latter trend was attributed to the gas heating [12]. As the pressure increases, the Ar(1s5) density gradient becomes stronger and the profile moves closer to the cathode [12].

The gas temperature \( T_g \) was extracted by analyzing the absorption lineshapes. Experimental profiles were fit to a Voigt function with \( T_g \) as the fitting parameter. The results of \( T_g \) measurements obtained by absorption spectroscopy are presented in Figure 8. This Figure also shows a comparison of the results obtained by DLAS and those obtained by OES [11]. OES measurements were based on the rovibrational bands of the N\(_2\) first positive system. Here, gas temperatures are compared at the spatial location of the peak argon metastable density (~50 \( \mu m \) from the cathode) and for \( P = 100 \) Torr. The same trend with discharge current is observed with both methods. However, the values obtained with OES are slightly higher than those obtained by absorption spectroscopy.

This discrepancy can be explained by the influence of the trace gas admixture on the microdischarge characteristics. Absorption spectroscopy was done in pure argon microdischarges, while OES was done in microdischarges of argon with a trace of N\(_2\). The insert of Figure 8 shows that, even small additions of
nitrogen (0.5%), result in higher discharge voltage for the same current (compared to pure argon), and therefore higher power deposition. This may explain the higher $T_g$ values obtained by OES.

Figure 8. Comparison of $T_g$ measurements obtained by absorption spectroscopy and OES. Insert: I-V characteristics of a pure argon microdischarge (black) and of an argon microdischarge with 0.5% of $N_2$ (red); $P = 100$ Torr.

4. Conclusions

Parallel plate slot-type DC microdischarges were characterized using laser spectroscopy. Laser Thomson Scattering (LTS) was employed for simultaneous direct measurements of electron density ($n_e$) and electron temperature ($T_e$) in argon microdischarges. $n_e$ values obtained by LTS were consistent with those found by Optical Emission Spectroscopy. Diode Laser Absorption Spectroscopy (DLAS) was used for spatially resolved measurements of argon metastable Ar(1s) density in argon microdischarges. The analysis of the absorption lineshapes also provided the gas temperature. By fitting the absorption lineshapes to Voight functions, the gas temperature was extracted as the fitting parameter. The values of $T_g$ obtained by DLAS were found to be in reasonable agreement with those obtained by OES, showing the same increasing trend of the gas temperature with current.

The authors thank the Department of Energy (DE-FG02-03ER54713), and the University of Houston GEAR program for financial support of this work.

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