Laser induced fluorescence of argon ions in a plasma presheath

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The characteristics of presheaths near an electrically floating plate in weakly collisional argon multipole plasmas are investigated with a combination of data from laser induced fluorescence using a diode laser, Mach probes, emissive probes, and Langmuir probes. It is shown that ion–neutral collisions result in an increase in ion temperature from approximately room temperature in the bulk plasma to 0.13 eV, 0.5 cm from the plate, the location of the closest measurement. In addition, at that point, the presheath plasma potential drop is greater than \( T_e/2 \), and the drift velocity is equal to 0.5 \( c_s \), where \( c_s \) is the ion sound velocity. © 2001 American Institute of Physics.

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I. INTRODUCTION

Unmagnetized confined plasmas are usually separated into three regions; the bulk plasma, the presheath, and the sheath. The sheath thickness is the order of Debye lengths and for high densities is very small and hardly observable. The presheath thickness is the order of the ion collision lengths.\(^1\) The presheath is the nonuniform quasineutral transition region where ions gain energy to reach the Bohm velocity before they enter the sheath. Bohm showed that ions must have a kinetic energy at the sheath/presheath boundary that is at least half of the electron temperature (i.e., \( T_e/2 \)). Riemann\(^2\) has shown for weakly collisional plasma that the presheath potential near the sheath can be written

\[
\varphi = \frac{T_e}{e} \sqrt{\frac{x}{\lambda}},
\]

where \( \lambda \) is the ion–neutral collision length. While sheaths have been studied in considerable detail, the literature on presheaths is not extensive and experimental verification of most predictions does not exist. This is in part because predicted presheath electric fields are small and difficult to measure accurately and it has not been clear how to measure ion drift velocities and ion distribution functions near plasma boundaries. Nevertheless, the understanding of presheaths is important for understanding plasma/wall, limiter, and divertor interactions in fusion plasmas, ion scattering in presheaths in industrial plasma applications, and ion behavior in ion thrusters as well as basic plasma diagnostic techniques. In all cases, the presheath determines the ion flux to the boundaries and provides ions at the sheath boundary with energies of at least the Bohm velocity.

In weakly collisional, weakly ionized (<1%) plasmas, such as those used for high ion density (\( n_i > 10^{11} \text{cm}^{-3} \)) plasma etching, presheath lengths are determined by charge exchange and elastic ion–neutral collisions.\(^4\) The ion–neutral collision lengths \( \lambda \) satisfy \( \lambda_D < \lambda < L \) where \( \lambda_D \) is the Debye length and \( L \) is the order of the plasma dimension. In collisionless plasmas, ions can be accelerated across the presheath by potential changes equal to \( T_e/2e \). However, weakly collisional plasmas require potential steps greater than \( T_e/2e \).\(^5,6\) In addition to reducing the ion drift velocity (the first velocity moment of the ion distribution function), the presence of charge exchange, elastic collisions, and ionization broadens the ion distribution function, increasing the ion temperature as the ions approach the plate.

Laser induced fluorescence (LIF) is a well-established diagnostic technique for determining ion and neutral velocity distribution functions. It can be used to determine ion and neutral density profiles and temporal variations. LIF is a nonintrusive diagnostic technique, whose usefulness depends on the specific species being investigated.

Mach probes provide an alternative way to determine ion drift velocity. They consist of back to back Langmuir probes with an insulator between them. For uniformly drifting, unmagnetized, collisionless plasma, Hudis and Lidsky\(^8\) used Bohm presheath theory to derive the ion drift velocity on the ratio of ion saturation currents to the two sides of the probe. Hutchinson\(^9\) derived the current ratios as a function of the Mach number for a magnetized plasma with ion gyroradius smaller than the probe size. Hutchinson included the particle diffusion into the presheath as well as particle diffusion out from the presheath and neglected ionization. Gulick et al.\(^10\) used Mach probes to measure the ion drift velocity in an argon electron cyclotron resonant (ECR) plasma. They compared the experimental Mach number results from laser induced fluorescence (LIF) data and Mach probe results. Based on LIF measurements, they concluded that the Hutchinson model is the best for ECR plasma in the unmagnetized region. They pointed out that neither of the models give accurate plasma potential variation in the presheath. In an investigation of Mach probes in density gradients, we have found that the saturation currents to each side of the Mach probe depend on the current at the respective probe presheath–plasma boundaries and are insensitive to probe radius \( a \) when probes are smaller than the presheath dimensions, i.e., \( a/\lambda \ll 1 \).

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Mach probes were moved so that, at a given position, data on each side were taken at that position. For locations where the Mach probe diameter was comparable to the separation from the plate, the experimental data were somewhat questionable because of shadowing effects. However, data taken with a Mach probe with a diameter $=3.2$ mm agreed with the $9.5$-mm-diam probe data to within 1 cm from the plate, but had larger uncertainty. Data were not taken simultaneously from the two sides of the probe. The noncollecting side was grounded. This work will be reported elsewhere.

Goeckner et al.\textsuperscript{11} compared LIF measurements of ion temperature with those made with a gridded ion energy analyzer for a dc hot filament discharge and with other ion energy analyzer results in the literature. Their LIF results gave a bulk ion temperature approximately equal to room temperature compared to the ion energy analyzer results which were the order of 0.1 eV or more, i.e., higher than the LIF results. They concluded that the resolution of the ion energy analyzer was poor. LIF experiments have usually been carried out using dye laser systems. Recently, Severn et al.\textsuperscript{12} made the ion temperature measurements in a dc multipole system using a diode laser. They excited the metastable argon ion at 668.61 nm.

In this paper measurements of ion distribution functions in the nonuniform presheath in an argon plasma are presented. Measurements were made with a diode laser system similar to that described by Severn et al.\textsuperscript{12} Emissive probe and Mach probe results are compared with the LIF results to clarify the role of collisions. The experimental setup is described in Sec. II. The experimental results are given in Sec. III. General comments and a discussion are given in Sec. IV.

II. EXPERIMENTAL CONSIDERATIONS

The experiments were performed in two different dc hot filament multipole plasma\textsuperscript{13} systems. The working pressure was approximately 1 mTorr and argon gas was used. The plasma was obtained by heating thoriated tungsten filaments was approximately 1 mTorr and argon gas was used. The experiments were performed in two different dc hot filament discharge systems. Recently, Severn et al.\textsuperscript{12} compared LIF measurements of ion energy analyzer results with those made with a gridded ion energy analyzer for a dc hot filament discharge and with other ion energy analyzer results in the literature. Their LIF results gave a bulk ion temperature approximately equal to room temperature compared to the ion energy analyzer results which were the order of 0.1 eV or more, i.e., higher than the LIF results. They concluded that the resolution of the ion energy analyzer was poor. LIF experiments have usually been carried out using dye laser systems. Recently, Severn et al.\textsuperscript{12} made the ion temperature measurements in a dc multipole system using a diode laser. They excited the metastable argon ion at 668.61 nm.

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II. EXPERIMENTAL CONSIDERATIONS

The experiments were performed in two different dc hot filament multipole plasma\textsuperscript{13} systems. The working pressure was approximately 1 mTorr and argon gas was used. The plasma was obtained by heating thoriated tungsten filaments (diameter $=0.02$ cm) biased at approximately $-100$ V with respect to the grounded chamber wall. Presheaths were measured near conducting plates mounted in each of the plasmas. The diameters of the plates were 7.5 cm for the small chamber and 15 cm for the large chamber. The cylindrical chamber dimensions were length $=40$ cm and diameter $=35$ cm and length $=75$ cm and diameter $=65$ cm, respectively. LIF measurements were collected by a 1.3-cm-diam lens (which focused onto a pinhole) at a perpendicular distance 15 cm from the laser beam axis for the small chamber and 25 cm for the large chamber. Two different approaches were taken to eliminate laser beam reflections from the plate. In the small chamber, the plate axis was oriented at a small angle ($\approx 4^\circ$) with respect to laser direction. In the large chamber, the beam was along the plate axis and a small hole (diameter $=0.2$ cm) was opened at the center of the plate. The detection system was not moved during the experiments. The target metal plate was moved instead.

The laser setup employed the design of Severn et al.\textsuperscript{12} and consisted of a small tunable diode seed laser (center wavelength $=668.61$ nm) and a power amplifier (PA) which had a 10 nm range. The laser system excited a metastable Ar II ion level in the presheath near the floating plate. The metastable level\textsuperscript{12} was chosen since it is difficult to excite ions from their ground states because their resonance wavelengths are too short for commercially available lasers.\textsuperscript{14}

A computer controlled the tunable diode laser. The output of the tunable laser was approximately 4 mW and approximately 1 mm in diameter. The PA laser amplified the seed laser light to approximately 200 mW; this beam was directed into the plasma. The LIF signal was collected perpendicular to the incoming laser beam. The tunable diode seed laser could be scanned over a range of approximately 100 GHz in steps of approximately 100 MHz. The LIF system setup is shown schematically in Fig. 1. The LIF signal was chopped, the modulated LIF signal was detected by a photomultiplier tube, and the electrical signal was measured by a lock-in amplifier. The output was digitized and collected with a computer.

A Mach probe was used to measure the drift velocity profile over the presheath region. Drift velocity was determined from Mach probe ion saturation current ratios\textsuperscript{15} using

\[
\frac{J_U}{J_D} = \exp\left(4 \sqrt{\frac{T_i}{T_e}} u_M \right).
\]

Here, $J_U$, $J_D$ are the upstream and downstream current ratios and $u_M$ is the Mach number, i.e., the ratio of the ion drift velocity to the ion acoustic velocity. The ion temperature at the probe was taken as 0.18 eV, which was 10% of the electron temperature.

Plasma potential measurements were made using an emissive probe in the limit of zero emission technique.\textsuperscript{16} Plasma density measurements, far from the plate, were made by double-sided Langmuir probes (diameter $=0.95$ cm).

In this experiment, metastable argon atoms were excited. The main sources\textsuperscript{10} of the metastable argon ions are electron–ion collisions or electron–neutral collisions leading to ionization and excitation by photons. The excitation laser wavelength (668.61 nm) and the laser induced fluorescence wavelength (442.7 nm) are both in the visible region. The collected fluorescence signal was filtered because the background emissions from the filaments were very high. A bandpass filter, centered at 442.6 nm, with a full width at half maximum of 0.75 nm, was used to reduce the noise and a photomultiplier tube was used as the detector.
Laser induced fluorescence has been used for many years to determine plasma characteristics. The LIF technique for velocity measurements takes advantage of the Doppler effect. If the metastable ion is moving parallel to the laser beam with velocity $v$, the laser light at frequency $\nu$ can be absorbed when

$$\Delta \nu = \frac{v}{c} \nu = \frac{v}{\nu},$$

(2)

where $c$ is the speed of light. The laser had a scanning range from 668.55 to 668.675 nm, so the maximum measurable metastable argon ion velocity is $5.3 \times 10^4 \text{ m/s}$ in this system. Determining the zero velocity for the Ar II from the LIF data in the bulk plasma and measuring the Doppler frequency shift yielded the ion distribution function in the drifting plasma.

III. EXPERIMENTAL RESULTS

The plasma density was measured to be approximately $5 \times 10^{10} \text{ cm}^{-3}$ with a Langmuir probe. Emissive probe measurements were made to determine the plasma potential profile. Representative results corresponding to an electron temperature $T_e = 1.1 \text{ eV}$ are given Fig. 2. The expected maximum ion energy, at the closest position to the plate that was measured, is limited to the energy the ions pick up in falling through the presheath to approximately $T_i/2$, assuming no reduction in velocity from collisions. The ion temperature is expected to be in between room temperature and the electron temperature (i.e., 0.025–2.0 eV). For Maxwellian distribution functions, the ion temperature is given by

$$T_i = \frac{m c^2 \Delta \nu^2}{8 \ln 2 \nu_0^2}.$$

(3)

Here, $m$ is the ion mass, $T_i$ is the ion temperature, $\Delta \nu$, $\nu_0$ are the full width at half maximum and the centerline frequency of the Gaussian-shaped ion distributions. It was assumed that the metastable ions are a good indicator for the plasma ions. For the small chamber, the location of zero drift velocity was found to be 4.9 cm from the plate from the emissive and Mach probes data. The ion temperature at zero drift velocity was found to be 0.039 eV. With the exception of points $\leq 1$ cm from the plate the one-dimensional fluid velocities were determined from the first moment of the distribution function, normalized by the area under the distribution curve,

$$v_d = \frac{\int_{-\infty}^{\infty} v f(v) dv}{\int_{-\infty}^{\infty} f(v) dv}.$$

(4)

The ion temperature was calculated using

$$T_i = \frac{\int_{-\infty}^{\infty} m (v - v_d)^2 f(v) dv}{\int_{-\infty}^{\infty} f(v) dv}.$$

(5)

In the small chamber, data were taken by giving a small scattering angle to the plate to eliminate reflected laser effects. Unfortunately, for distances $\leq 1$ cm, reflected laser light was not fully eliminated, so data for negative velocity are not meaningful. LIF measurements on the axis of a circular plate for a neutral pressure of 1 mTorr argon are given in Fig. 3. One prominent feature is the broadening of the distribution function as the ions approach the plate. It appears that there is a large contribution from charge exchange neutrals, close to the plate. The ion temperature was found to be $0.039 \pm 0.008$ eV. 5 cm from the plate. The electron temperature in the smaller chamber was 1.85 eV, which corresponds to a Bohm velocity of $2.15 \times 10^3 \text{ m/s}$. At the 4 cm location, the ion temperature increased to $0.056 \pm 0.017$ eV. The peak velocity shifted from $-100$ to $311 \text{ m/s}$ and the fluid velocity increased to $155 \text{ m/s}$. At the 1 cm location, the ion temperature changes to $0.081 \pm 0.01 \text{ eV}$ and the peak velocity reaches $1.24 \times 10^3 \text{ m/s}$, which is almost 64% of Bohm velocity. The drift velocity is $1.08 \times 10^3 \text{ m/s}$, which is less than the peak velocity and almost 50% of the Bohm velocity.

LIF measurements on the axis of a circular plate in the large chamber for a neutral pressure of 1 mTorr argon are given in Fig. 4. Data were taken with a hole in the plate to eliminate laser reflections. Unfortunately, the hole also allows ions to leak in from the direction opposite to the laser, so again data for negative velocity are not meaningful.

In the large chamber, the 8 cm point was identified as the location of zero velocity. The ion temperature at this location $0.035 \pm 0.010$ eV was colder than in the small chamber but agreed within the experimental uncertainty. Both appeared to be slightly warmer than room temperature. As the ions get closer to the metal target plate, the ion temperature increased.
as it did in the small chamber. In the large chamber, the electron temperature was 1.1 eV, which corresponds to an ion sound velocity of $1.66 \times 10^3$ m/s, so the ions enter the presheath of the plasma with a velocity approximately equal to 15% of the Bohm velocity, which is similar to results from the small chamber. At 0.5 cm, the closest location at which measurements could be made, the ion temperature increased to $0.13 \pm 0.02$ eV, the ion peak velocity reached to $1.12 \times 10^3$ m/s, and the ion fluid velocity was 846 m/s. In both chambers, the plasma densities were high and the sheath thicknesses were very small.

The Mach numbers calculated from the average flow velocities and the peak velocities versus position are shown in Figs. 5 and 6, respectively. The data are in good agreement. The Mach numbers calculated from the average LIF flow velocities are also compared with the Mach numbers found from the Mach probe in the small chamber in Fig. 5. These data are also in good agreement.

The ion temperature increases as the ions get closer to the plate. The change in the ion temperature is shown in Fig. 7. The ion velocity distribution takes on a different shape near 1 cm. Two peaks are apparent. The “peak” which has the higher ion velocity reaches 64% of the ion sound velocity at approximately 1 cm from the plate. The slow peak is centered at zero velocity. The negative part of the peak in this case is the result of the reflected laser light from the plate. The emissive probe results show that at the same location the potential drop is more than half of the electron temperature but the ion velocity is much less than the Bohm velocity.

**IV. DISCUSSION AND CONCLUSIONS**

The experiments performed in two different size chambers showed similar results in the presheath region of the metallic collector. At the closest points measured, 1 cm from the plate in the small chamber and 0.5 cm from the plate in the large chamber, the ions did not reach the Bohm velocity in the weakly collisional plasma. In both plasmas, the potential drop from bulk plasma to the location where the ion velocities are measured is greater than $T_e/2e$. If the ion motion were collisionless, the measured potential changes would correspond to ion velocities greater than the Bohm velocity. Presheath plasma potential drops greater than $T_e/2$ are in agreement with previous predictions\(^6\) and somewhat similar to the experimental result found in the previous LIF experiment.\(^11\) Unlike that experiment, the Mach probe data agree with the LIF data. It is expected that the sheath thickness is the order of the Debye length for the floating wall and the Debye length is very small ($\lambda_D \approx 10^{-2}$ cm). Therefore, the presheath/sheath boundary is still approximate 0.5 cm from the location of the closest measurement. Fitting to the presheath potential given by Riemann,\(^3\)

$$\varphi = \frac{T_e}{e} \sqrt{\frac{x}{\lambda}}.$$
provided a way to extrapolate the potential to the location of the sheath/presheath boundary near \( x = 0 \). This gives a total potential drop of \( 1.1 \pm 0.1 T_v/e \) from the bulk plasma to the sheath edge. Assuming ion flux conservation and electrons that satisfy the Boltzmann relation, the velocity at the sheath edge is predicted to be \( 1.0 \, c_s \).

The relative importance of ionization and charge exchange as sources of cold ions can be estimated by the ratio \( \langle \sigma_i v_i \rangle / \sigma_{cx} v_i \), where \( \langle \sigma_i v_i \rangle \) is the product of the electron velocity and the ionization cross section averaged over a Maxwellian electron distribution function with electron temperature \( T_e \) and \( \sigma_{cx} \) is the charge exchange cross section for an ion with velocity \( v_i \). For argon, \( \sigma_{cx} v_i \approx \text{constant} = 2 \times 10^{-9} \, \text{cm}^3/\text{s} \). Since \( T_e \) is \( 1-2 \, \text{eV} \), \( \langle \sigma_i v_i \rangle \) is approximately \( 10^{-11} \, \text{cm}^3/\text{s} \), so ionization can be neglected. The standard Langmuir probe theories assume a potential drop of \( T_v/2e \) in the presheath and the Boltzmann relation to obtain a \( 0.6 = e^{-1/2} \) multiplication constant in the expression for the ion saturation current. However, it is now clear that this multiplication constant is a variable depending on the plasma conditions. For example, for the weakly collisional plasma described in this paper, the potential drop is on the order of the electron temperature, which corresponds to \( 0.36 \) for the multiplication constant.

Note that the maximum peak in velocity is approximately at \( 0.7 c_s \) while the average flow velocity at the closest distance was somewhat less. Assuming collisionless ion motion, treating the ions as single fluid, and assuming constant ion temperature are clearly questionable assumptions. The ion temperature changes by more than a factor of 3 in the measurable distances. Therefore, the charge exchange and elastic scattering must be considered as ion heating mechanisms. Goeckner et al.\(^{11} \) pointed out that ion energy analyzers indicated higher ion temperature. It can now be concluded that this is not the result of poor ion energy analyzer resolution but is actually the result of the increase in ion temperature that occurs in front of the floating first grid of the ion energy analyzer. In this experiment, with plasma density \( \approx 5 \times 10^{10} \, \text{cm}^{-3} \) and the neutral gas density \( \approx 3 \times 10^{11} \, \text{cm}^{-3} \) at 1 mTorr, recycling is not expected to be an issue. A final conclusion from the agreement of Mach probe and LIF data is that Mach probes can be used to measure ion drift velocities in density gradients in unmagnetized plasmas.

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