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Journal

Thin Solid Films, 506-507

ISSN

0040-6090

Authors

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Publication Date

2006-05-26

DOI

10.1016/j.tsf.2005.08.114

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Peer reviewed

Ion flow and sheath physics studies in multiple ion species plasmas using diode laser based laser-induced fluorescence

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Available online 27 December 2005

Abstract

Diode lasers have proved to be a valuable light source for laser-induced fluorescence (LIF) measurements for plasma science since the early 1990s, and they have recently improved the state of the art of measuring ion flow from ion velocity distribution functions (ivdfs) at the sheath–presheath boundary in single and multiple ion species plasmas. In the case of a low temperature two ion species plasma (ArI+HeI), we were the first to show experimentally that ion species may reach the sheath edge flowing at a very different speed than that expected from the single species Bohm Criterion (ArII ions exceed the individual Bohm flow speed by almost a factor of 2 at the sheath edge). Simulation results are found to agree. Diode laser technology relevant to LIF measurements in multiple ion species plasmas is discussed with the aim of addressing outstanding problems in sheath formation in such plasmas.

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Keywords: Plasma sheath; Plasma diagnostics; Plasma spectroscopy

1. Introduction

The flow of multiple ion species onto plasma boundaries is an important feature of many plasma systems. Divertors and limiters in fusion plasma devices, substrates in etching and deposition plasmas, and Langmuir probes are among the many examples where the plasma–wall interaction is a prominent feature of the system and in which multiple ion species are often present. Understanding sheath formation in such plasmas, a longstanding problem [1–3], remains a practical problem of critical interest in plasma science [4–7]. Its recent revival among plasma theorists in both the DC and RF communities [8–13] has begun to be matched by relevant experiments, however it is still true that theory is ahead of experiment.

Riemann and others [9–11] have argued that for a multiple ion species plasma which is weakly collisional, individual ion species must satisfy a generalized Bohm criterion (GBC) expressed as

$$1 \geq \sum_j \frac{n_{jo}}{n_{eo}} \frac{C_j^2}{v_{jo}^2}, \quad (1)$$

where the ion sound speed for each species is $C_j = \sqrt{T_e/m_j}$, T_e is the electron temperature measured in eV, m_j is the ion mass, n_j the density, v_{jo} ion fluid speed, and j numbers the ion species. The zero subscript refers to quantities at the boundary between the sheath and presheath. Unlike the case of a single species plasma where the equality is normally satisfied by the ion species reaching its sound speed, in the case of multiple ion species, the equality may be satisfied by speeds faster or slower than the sound speed for a given ion species. Two simple solutions satisfying Eq. (1) are that all ions attain the same speed at the sheath edge, and that each species attains its own Bohm speed. The former solution

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¹ Work supported by DOE grant no.DE-FG02-03ER54728, and NSF grant no. CHE0321326.

works out to be the ion acoustic speed of a homogeneous plasma of multiple ion species with no ion drifts. Many authors quite naturally have assumed the latter solution [13,14]. If the neutral pressure is sufficiently high, however, the flow is mobility limited. In this case, Franklin [11,12] among others have shown that the ratio of ion flow velocities at the sheath edge is that of their mobilities. This ratio will not in general equal unity, nor will it equal the ratio of their Bohm speeds, thus eliminating the two simplest solutions cited above. However, preliminary experimental results found by Hala [15], based on ion acoustic wave measurements in an Ar+He plasma, suggest that ArII ions reach the same speed as HeII ions near the sheath edge.

Since then, we have demonstrated unambiguously that in such a two ion plasma, the ArII ions do indeed exceed their individual Bohm speed at the sheath edge, and approach the ion sound speed of the ArII+HeII plasma system, as calculated in the bulk plasma [16]. This was the first experimental work that measured ion flow at the sheath edge in multiple ion species plasmas using LIF which combined plasma potential measurements, and which served to locate the ivdfs relative to the spatial sheath structure. Our work, of course, was not the first to employ an LIF diagnostic to measure ion flow at the sheath edge: the work on a single ion species plasmas began with Gulick et al. [17], who found that in a high density magnetized ECR plasma ($n_e = 3 \times 10^{11} \text{ cm}^{-3}$, $kT_e \sim 10 \text{ eV}$) that the ions did not reach the Bohm speed at the boundary. In lower density plasmas ($n_e \leq 1 \times 10^9 \text{ cm}^{-3}$), other researchers did find that the flow velocity exceeded C , although Goree et al. [18] did not measure the plasma space potential and had to infer the location of the sheath edge, and although Batchet et al. and Carrere et al. [19,20] did measure the plasma potential, their potential measurements did not specifically locate the sheath edge. However, in all work other than Gulick's, there were unmistakable signatures in the ivdfs that indicated that some were measured between the sheath edge and the material boundary. All employed pulsed or CW dye laser systems, and in all cases, the ion flux, or the area under the curve of the ivdfs, appeared to rise as the ions fell into the sheath, a point to which we will return.

Oksuz et al. and Meyer et al. [5,7] were the first to perform these experiments using a Littman type extended cavity diode laser system, based on the LIF schemes and seed laser, optical amplifier system worked out by McWilliams et al.'s group [21]. Compared with dye laser systems, diode lasers take up considerably less space (one quarter of a 4 by 8 ft optics table is sufficient), use considerably less power (one or two 120 V power outlets is sufficient), and cost considerably less (our present system based on a Littrow type cavity [22], costs on the order of \$10,000 [23]). In what follows, we will describe our experiment and LIF set up (Section 2), our results in one and two ion species plasmas (Section 3), and then make some remarks about the meaning of those results with a view to filling in gaps in what is known (Section 4). We wish to discuss experimental

aspects of these results not previously reported, particularly in connection with the diode laser based LIF diagnostic, and we introduce first results of particle-in-cell (PIC) simulations which agree with those results.

2. Experimental arrangements

The experiments with pure Ar plasmas and with Ar+He plasmas reported here were performed in a DC hot-filament multi-dipole plasma system [24]; details of emissive probe measurements of the plasma space potential and ion acoustic wave measurements to determine the relative ion concentrations are described in detail by Severn et al. and Hala and Hershkowitz [16,25]. It is sufficient to recall four facts: (a) the relative concentrations of the ions could not be independently varied because of Penning ionization between metastable He neutrals and Ar neutrals in the ground state, (b) the ratio of neutral pressures exceeded 25 ($P_{\text{He}}/P_{\text{Ar}} > 25$) in order to achieve an argon ion fraction nominally exceeding 50% ($n_{\text{Ar}}^+/n_e = 0.65$), (c) we estimated that the resonant charge exchange mean free path for Ar was larger than that of He, by a factor of approximately 3.5, and (d) the mean free path for Ar ion charge exchange on He I was larger than that of resonant charge exchange for He by a factor of 7 [26].

Argon ion flow through the bulk plasma into the presheath and sheath was measured using a diode laser based [21] laser-induced fluorescence diagnostic, as shown in Fig. 1. The optical cavity of the seed diode laser (Newport 2010) was a Littman–Metcalf type, capable of 5 mW output with 10 nm course tuning centered on 668 nm, and the optical amplifier (SDLTC30) was necessary in order to boost the intensity by a factor of 5. The typical incident laser power for these experiments ranged between 20 and 25 mW. The laser bandwidth ($\Delta\nu_l \sim 500 \text{ kHz}$) was smaller than the limited by the natural line width of the LIF excitation transition ($^4F_{7/2} - ^4D_{5/2}$) by a factor of 20, which in turn was smaller than the Doppler broadened FWHM by a factor

Fig. 1. The LIF apparatus comprises a tapered chip optical amplifier (SDL TC30) that is injection locked to a low power ($P_l < 10 \text{ mW}$) diode laser (Newport 2010ECU) in a Littman–Metcalf cavity, tunable over 20 nm and sets the bandwidth of the output beam at nominally 500 kHz. Typical operating power was 20–25 mW. Optical isolators protect both lasers from unwanted optical feedback and the $\lambda/2$ waveplate optimizes the input polarization.

Fig. 2. The spatial profile of plasma potential for the pure Ar plasma shows the presheath and the sheath. At the location of the sheath edge, within error, the rms velocity passes through the Bohm speed.

of 100, and the corresponding velocity space resolution was about 10 m/s. The air wavelengths of the excitation and fluorescent photons were 668.4293 nm and 442.6001 nm, respectively. The details of the collection optics, and how the volume in space diagnosed by LIF was located with respect to the boundary plate and probes, is discussed elsewhere [16]. We mention here that the signal acquired from the Lock-in Amplifier as the frequency of the laser was scanned was averaged over 5 to 10 scans. Individual scans were examined for indications of mode-hops and eliminated if found. Mode-hops were infrequent for scans narrower than about 50 GHz, a bandwidth roughly 5 times wider than our data scans. The beam passed through an iodine gas cell before entering the plasma, so that I_2 and ArII fluorescence signals could be obtained simultaneously. The frequency of the I_2 fluorescence corresponding to an ArII ion was used as the fiducial mark from which detuning frequencies were calculated. The fiducial mark on I_2 spectrum corresponding to absorption of a photon by an Argon ion at rest was determined by interpolating between known lines in a standard I_2 atlas [27], as described by Severn et al. [21]. Argon ion velocities were then calculated by the first order Doppler shift, $\omega = \omega_l - kv_z$, or $v_z = \lambda (f - f_l)$, where the laser beam was defined to be in the $-\hat{z}$ direction; f_l is the frequency of the laser photon in the lab frame, and Δf is the Doppler shift measured from the iodine spectrum.

The LIF signal is proportional to the reduced one-dimensional ion velocity distribution function (ivdf) along the beam direction, $f(v_z, z)$. Standard references [28] to the meaning of the LIF signal describe the integration along the velocity space coordinates normal to the beam direction. We

used the LIF signal to calculate both the second and inverse second moment of the ivdfs,

$$\langle v_z^2 \rangle = \frac{\int_{-\infty}^{\infty} v_z^2 f(v_z, z) dv_z}{\int_{-\infty}^{\infty} f(v_z, z) dv_z}, \quad (2)$$

so as to arrive at the $v_{\text{rms}} = \sqrt{\langle v_z^2 \rangle}$ and $v_{\text{rms}} = \sqrt{\langle 1/v_z^2 \rangle}^{-1}$, for comparison with the ArII ion sound speed, C_1 . The empirical constant of proportionality relating the LIF signal to the ivdf that depends on efficiency of light collection as described above, cancels out.

3. Results

Our results for the single species Ar plasmas were described previously [16]. Our confidence in our technique and our estimate of the fiducial point in the I_2 fluorescence spectrum was confirmed by our calculation of the v_{rms} as a function of position, showing that it passes through the Bohm velocity, within error, just at the sheath edge (Fig. 2). The Argon neutral pressure was 0.1 mTorr and there were two Maxwellian electron components at 1 eV and 4.5 eV. The harmonic mean of these was used to calculate ion sound speed, which was $C_1 = 2.07$ km/s in this case. The measured thickness of the sheath (6.0 ± 0.5 mm) agreed to within 5% with the calculation of the sheath thickness using the Child–Langmuir model.

Neutral He was added to create a two ion plasma, keeping the Ar neutral pressure fixed. For the He–Ar plasmas of this experiment, Langmuir probe measurements again revealed a bi-Maxwellian electron temperature, with cold and hot components of 0.97 and 4.68 eV, respectively. The effective average kT_e previously described was used to calculate the C_j for each species, which was 1.62 and 5.12 km/s for ArII and HeII ions, respectively. The plasma density in the bulk plasma remote from the walls of the

Fig. 3. A family of ivdfs for the two ion plasma (Ar+He). The ion acoustic speed of the two ion plasma is given by v_{ph} .

chamber and remote from the plate was approximately 10^9 cm^{-3} giving $\lambda_D = 0.25 \text{ mm}$. A typical ion acoustic velocity in the bulk plasma far from the plate was $v_{ph} = 3.28 \text{ km/s}$, giving the relative ion concentrations, $n_{Ar}/n_e = 0.66$ and $n_{He}/n_e = 0.34$. A family of ivdfs is given in Fig. 3. The ivdfs in Ar–He plasma were considerably noisier than the case of the pure Ar plasma. However, it is clear that for $z < 1 \text{ cm}$, the peak of the distribution exceeds the ArII Bohm speed. The asymmetry on the $v_z = 0$ side of the ivdfs was present as was the case for the ivdfs in pure Ar plasma, consistent with the effects of charge exchange collisions. Beyond the sheath edge, we observed ivdfs that became distended on the high velocity side of the peak of the distribution, indicating the presence of ions in the sheath.

The ArII Bohm velocity, C_1 , was compared with v_{rms} and v_{rms} as a function of position relative to the plate, and is shown in Fig. 4. Emissive probe measurements show that the sheath edge, z_0 is $3.0 \text{ mm} < z_0 < 4.0 \text{ mm}$, and the calculation of the Child–Langmuir sheath thickness was $d_{CL} = 2.5 \pm 0.3 \text{ mm}$. LIF measurements clearly show that the rms speed of the ArII ions exceeds its own Bohm speed. We should point out that ion sound speed of the system ($v_{ph} = 3.28 \text{ km/s}$) itself of course is much faster than the ArII Bohm speed (103% greater than C_1). At the sheath edge, the rms velocity is 77 % greater than C_1 , and rapidly approaches ion sound speed of the system. But it is clear that ArII ions fall into the sheath moving significantly faster in the Ar–He plasma than they do in the pure Ar plasma.

Fig. 4. The plasma potential profile for the Ar+He plasma reveals that the rms velocity of the ArII ions is well beyond the individual Bohm speed C_1 and approaching the ion sound speed of the system (as measured in the bulk) at the sheath edge. The arrow marks the sheath edge as determined by PIC code simulation, and simulation (○) shows good agreement with measurement.

4. Discussion and conclusions

The generalized Bohm criterion predicts that the j th individual ion species may reach the sheath edge traveling at speed which differ from C_j ; our results are certainly consistent with this. One of us (M.M.T.) has written a 1-D PIC code to model these results, based on a conventional particle-in-cell model with Monte-Carlo collisions. While code results show significant disagreement with experiment in the presheath, there is good agreement with the fundamental result, that is, the argon ions reach the sheath edge traveling significantly faster than their individual Bohm speed. The simulation gives results in terms of dimensionless variables (e.g., potential normalized to kT_e , flow speed normalized to C , and so on), and we analyzed their predictions for our particular parameters. The input parameters of the simulation are similar to experiment: $n_e = 1 \times 9 \text{ cm}^{-3}$, $P_{He}/P_{Ar} \gg 1$, and $\langle n_0 v_0 \rangle = \Gamma_0 = 2 \times 10^{18} \text{ m}^{-2} \text{ s}^{-1}$, the total ion flux incident upon the sheath edge. For the case that the ratio of the Ar flux to the whole was about half, the simulation suggests that the Ar ions reach the sheath edge traveling somewhere between 10 and 30% faster than C_1 , accelerating past the ion sound speed of the system well into the sheath, consistent with the experimental data, as shown in Fig. 4. The simulation also predicts that the He ions should reach the sheath edge traveling somewhat less than C_2 , but we could not diagnose the He ions with LIF. But this was not the only experimental limitation.

Further, because of Penning ionization, the relative ion concentrations could not be independently varied so as to observe how our results varied with ratio of the ion fluxes and with collisionality. It is desirable to replace He with another noble gas atom for which the metastable state neutrals have less internal energy than the ionization potential of neutral Ar in the ground state and which possess LIF schemes accessible by currently available diode lasers. Xe is a candidate in this regard, and it is possible that Kr is as well. Since only 25 mW was required to produce ivdfs in the Ar+He plasma, it is hoped that a single seed laser could diagnose both ions. One of us has worked out two suitable LIF schemes in XeII ($^4F_{9/2} - ^4D_{7/2}^0$, excitation at 699.3 nm, detectable photon at 485 nm) and ($^4F_{7/2} - ^4D_{5/2}^0$, excitation at 680.8 nm, detectable photon at 502 nm). The Littrow type optical cavity is more efficient than the Littman–Metcalf type cavity, although it is not quite as dispersive, hence the Littrow cavity produces a linewidth perhaps 20 times broader (10 MHz compared with 500 kHz), but is capable of producing 20–25 mW. The Sacher-Laser Technik TEC 100 [23] is such a laser, it has already been used in Ar plasma to create ivdfs, and modular diode laser heads can convert the cavity so as to produce 680.5 nm as well, so in principle, the same laser can diagnose both ions. The TEC100 is quite small (120 mm long, roughly the volume of a box of 4 table-tennis balls) and is consistent with the trend in extended cavity diode laser systems to become smaller. Soon such laser systems will become MEMs devices.

Finally, we point out that this result indicates that the flux of ions incident upon the sheath, which determines the flux of ions incident upon the material boundary, may vary considerably from that expected from the single species Bohm Criterion, especially if kT_e is a significant fraction of the voltage drop across the sheath. Further, if the distinction between presheath and sheath is lost in the case that the GBC is not satisfied, then sheath potentials extend some considerable distance in the plasma, degrading the velocity space anisotropy of the ions. Exploring experimentally the density parameter regime within which the GBC is satisfied is of critical importance for many plasma processing applications.

Acknowledgments

One of us expresses thanks for the support of the Department of Energy (DOE grant no. DE-FG02-03ER54728), the National Science Foundation (CHEM0321326). He is also grateful to Fred Skiff for the useful discussions about LIF and to Jason Wohlgemuth for the help with analysis.

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