High time resolution laser induced fluorescence in pulsed argon plasma

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(Received 1 May 2006; presented on 10 May 2006; accepted 23 May 2006; published online 21 September 2006)

A submillisecond time resolution laser induced fluorescence (LIF) method for obtaining the temporal evolution of the ion velocity distribution function in pulsed argon plasma is presented. A basic LIF system that employs a continuous laser wave pumping and lock-in aided detection of the subsequent fluorescence radiation is modified by addition of a high frequency acousto-optic modulator to provide measurements of the ion flow velocity and ion temperature in a helicon generated pulsed argon plasma with temporal resolutions as high as 30 μs. © *2006 American Institute of Physics.* [DOI: 10.1063/1.2217919]

I. INTRODUCTION

Laser induced fluorescence (LIF) is powerful tool for plasma diagnosis since it provides nonperturbative determination of the one-dimensional[1–3] (1D) and two-dimensional[4–6] (2D) particle (ions, atoms, and molecules) velocity distribution in a plasma. Typically, to obtain the particle velocity distribution function (vdf) in steady state plasma, weak LIF emission is discriminated against background light either by external modulation of the laser beam or by using pulsed lasers and, in both cases, subsequent phase synchronous detection. What are usually obtained are precise particle velocity ($\sim$50 m/s) and temperature ($\sim$0.1 eV) measurements with high spatial resolution (few mm^3). To investigate transient phenomena with short characteristic time scales, high time resolution LIF methods must be employed. Since the lower temporal bound of a LIF measurement is set by the lifetime of the upper optically pumped level (usually few nanoseconds), time resolutions as high as few nanoseconds could theoretically be achieved. In practice, however, the time resolution is limited by the statistics of LIF collected photons for a reasonable signal to noise, the *RC* time constants of electrical components of the LIF system, signal acquisition speed of the available electronics, and the particular plasma conditions. Time resolved LIF measurements with 0.2–20 μs resolution have been performed by employing as discriminator either a two-channel box car integrator/averager[7] or multichannel scaler.[8,9] In previous works[10,11] we demonstrated LIF time resolutions of 1 ms using a standard lock-in amplifier and a fast digital oscilloscope with a ring dye laser and a low power tunable diode laser. The 1 ms resolution limit arose from the requirement that the mechanical chopping frequency be a few times faster than the lock-in integration time (for reasonable signal-to-noise levels). In this work, we report an improvement in the LIF time resolution of a factor of $\sim$30. The improved time resolution, $\sim$30 μs, was made possible by replacing the mechanical chopper with a high-speed acousto-optic modulator and by digital signal processing of the raw data.

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II. EXPERIMENTAL APPARATUS

The experiments were performed in a pulsed plasma source discussed in detail elsewhere.[12] Briefly, 750 W of rf power was matched through a $m=+1$ helical antenna to a 1.5 m long, 10 cm diameter helicon source filled with flowing argon at 20 sccm (SCCM denotes cubic centimeter per minute at STP) and an operating pressure of 2.5 mTorr. The magnetic field strength on axis was 700 G. For these operating conditions, but in steady state mode, typical plasma parameters are electron temperature of $\approx$7 eV and electron density of $\approx 1.2\times10^{12}$ cm^{-3} as measured with a rf compensated Langmuir probe.[4]

For parallel argon ion LIF, we used a classical LIF scheme in which the Ar II $3d'\ ^2G_{9/2}$ metastable state is optically pumped by 611.66 nm (vacuum wavelength) laser light to the $4p'\ ^2F^0_{7/2}$ state. The $4p'\ ^2F^0_{7/2}$ state decays to $4s'\ ^2D_{5/2}$ state by emission at 461.09 nm. The laser used is a single-mode tunable ring dye laser pumped by a 6 W argon-ion laser yielding 200 mW of output power. A schematic of the LIF system used for parallel ion velocity distribution function (ivdf) measurements in pulsed helicon plasma is shown in Fig. 1. After passing through a 10% beam splitter, the laser beam is modulated with an acousto-optic modulator (AOM) at 100 kHz and then coupled into a multimode, nonpolarization preserving, fiber optic cable. The optics for parallel injection of laser light includes a 2.54 cm collimating lens, followed a Galilean telescope for beam waist reduction, and followed by a linear polarizer-quarter wave plate combination for conversion of the unpolarized laser light exiting the fiber into circularly polarized light to pump only one of the two σ transition clusters, specifically the $\Delta M=+1$ transition. The much smaller internal Zeeman splitting of the σ lines is ignorable during analysis of the parallel LIF data for magnetic field strengths less than 1000 G. The 10% portion of the laser beam is passed through an iodine cell for a consistent zero velocity reference. Spontaneous emission from the iodine cell absorption lines is recorded with a photodiode for each scan of the dye laser wavelength. The fluorescence radiation from the plasma is collected at 90° with respect to the laser beam and focused into a 200 μm diameter fused silica optical fiber. The intersection of 5 mm diameter in-

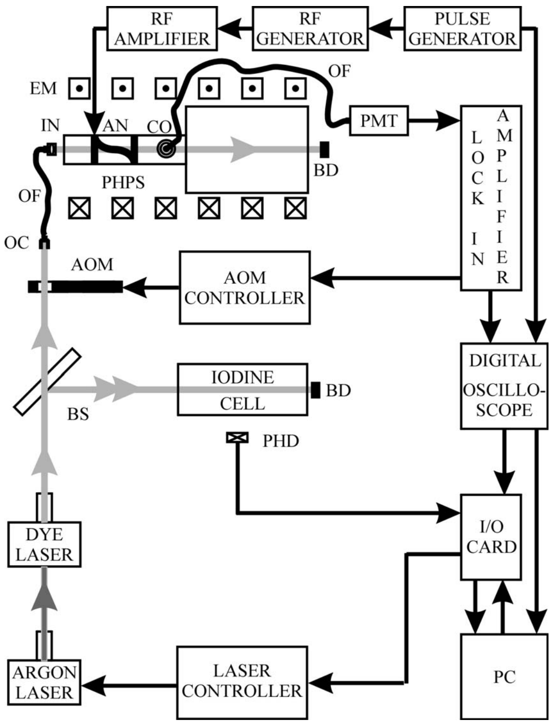

FIG. 1. Experimental setup for time resolved LIF diagnostic: PHPS—pulsed helicon plasma source, IN—injection optics, AN—antenna, CO—collection optics, BD—beam dump, EM—electromagnets, PMT—photomultiplier tube, OF—optical fiber, OC—optical coupler, AOM—acousto-optic modulator, PHD—photodiode, and BS—beam splitter.

jected laser beam and 0.8 mm diameter collection focus spot yields a measurement volume of ≈4 mm^3. Light exiting the collection fiber passes through a 1 nm bandpass interference filter centered at 461 nm. Following the filter is a photomultiplier tube (PMT) detector with an integrated 10 MHz bandwidth preamplifier. The PMT signal is composed of fluorescence radiation, electron impact induced radiation, and electronic noise. A lock-in amplifier, that provides the reference modulation signal to the AOM driver, is used to isolate the LIF signal from background emission at the fluorescence wavelength. The high-speed real and imaginary portions of the lock-in amplifier output are sent to a digital oscilloscope that is triggered off the rf modulation signal where they are averaged over few hundred plasma pulses and sampled at the digitization rate of the oscilloscope. The time resolution of the averaged signal is limited by the integration time setting of the lock-in amplifier and the digitation rate of the oscilloscope. The parallel drift velocity of the ions along the laser path is determined from the shift of the LIF peak relative to the iodine signal after correcting for the Zeeman shift of the σ absorption line. Since Doppler broadening dominates the width of the measured vdf, the parallel ion temperature is obtained from the full width at half maximum (FWHM) of the distribution. From our previous studies, for a reasonable signal to noise ratio, the minimum necessary "on/off" cycles within the lock-in integration time for a mechanical chopper (4 kHz chopping frequency) was 3–4. The AOM was driven directly by the lock-in amplifier at up to 100 KHz. Requiring three "on/off" cycles limited the integration time no less than 30 μs. As shown in Fig. 2(a), for a fixed integration time the LIF signal increased almost linearly with the log(modulation

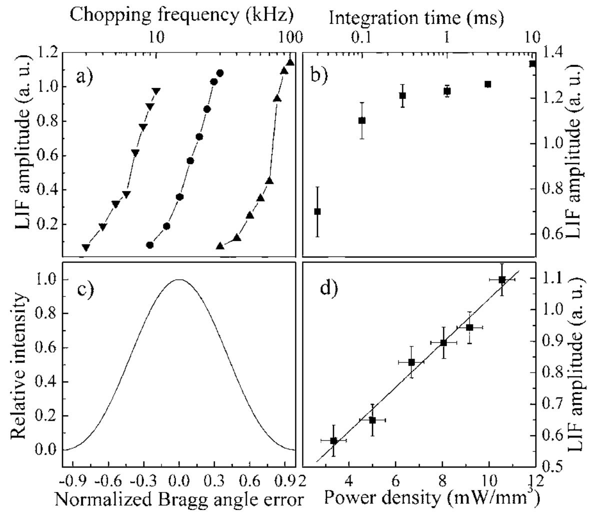

FIG. 2. (a) The LIF signal amplitude dependence vs chopping frequency (2—100 μs, (Š—300 μs, 1—1 ms integration times). (b) The LIF signal amplitude dependence vs integration time for 100 kHz chopping frequency. (c) The relative intensity of the AOM transmitted light in the first diffraction order vs Bragg angle misalignment. (d) The dependence of LIF signal amplitude vs injected laser power density.

frequency). Below a threshold value of the integration time, ~300 μs [see Fig. 2(b)], the LIF signal drops abruptly. The minimum integration time at which LIF signal could still be detected was 30 μs. At an integration time of 30 μs, the LIF signal is about 57% of the threshold value. At shorter integration times, the lock-in was unable to discriminate between the induced emission and spontaneous emission. However, the LIF signals were recorded at a digitization rate of 10 kHz thus limiting the time resolution to 100 μs. The transmitted laser power is significantly lower with the AOM than with the mechanical chopper. The transmitted intensity in the first diffraction order is a sensitive function [see Fig. 2(c)] of Bragg angle alignment,[13]

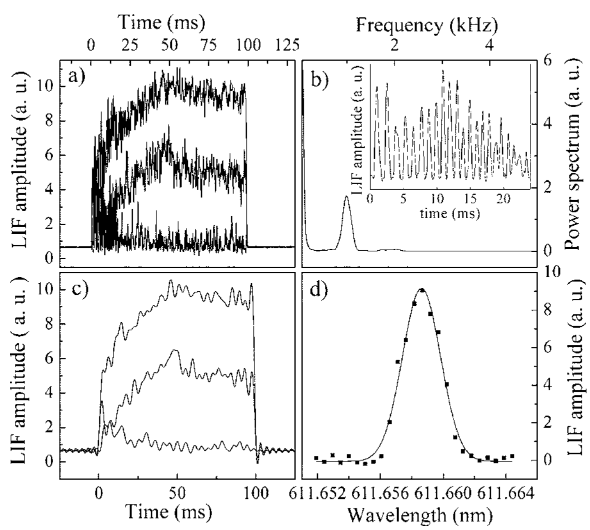

FIG. 3. (a) Raw LIF signal during the 100 ms discharge pulse (only 3 of 25 wavelengths are plotted). (b) Oscillations of the LIF signal amplitude observed on the first 26 ms of the pulse (as insert) and corresponding power spectrum showing 1 kHz oscillation frequency. (c) LIF signal after low pass filtering. (d) Argon ion vdf at $t=50$ ms into the pulse.

$$I_1/I_0 \propto [\sin^2(\pi\delta/\theta_B)]/(\pi\delta/\theta_B)^2, \quad (1)$$

where I_1 and I_0 are the intensity of the first order beam and the intensity of the zeroth order beam when the acoustic energy in the AOM medium is zero, respectively, $\theta_B = \lambda f/2v$ (λ—the laser wavelength in vacuum, f—acoustic frequency, v—acoustic velocity in the AOM medium) the Bragg angle, and δ the angular misalignment with respect to θ_B. For our AOM (Isomet 1205C-2 crystal with a Isomet 222A1 driver), I_1 is ~35% of I_0.[14] Since during the AOM "on" interval only ~10% of the light remains in the $m=0$ order, we chose to inject light from the $m=0$ beam that was modulated between 100% (during the "off" interval) and 10% (during the "on" interval). Losses in the injected light path, particularly in coupling into the fiber, reduce the injected light to 20%–30% of the laser output. For these levels of injected power (~40 mW), the LIF is in a linear regime [see Fig. 2(d)].

III. TIME RESOLVED LIF MEASUREMENTS

LIF measurements were obtained at a time resolution of 30 μs but with poor signal to noise. Therefore, only the 100 μs resolved data are shown here. Typically, 25 equally spaced wavelengths centered on the wavelength of peak LIF signal were used and the signal averaged over 300 plasma pulses. A wavelength span of 0.012 nm is sufficient to measure the entire vdf for argon ions with a 0.4 eV temperature. Increased fluorescence as the laser wavelength is tuned through the absorption line is evident in Fig. 3(a). Although noisy, LIF signal appears through the pulse except at the very beginning. During the first approximately 26 ms of the pulse, the LIF signal oscillates with a characteristic frequency of about 1 kHz [see Fig. 3(b)]. The oscillations are observed at all laser wavelengths, are unaffected by the modulation frequency, and vanish at long integration times. The oscillations are not electronic noise pickup as they also vanish if plasma light entering the collection optics is blocked. It is expected that any naturally occurring fluctuations in the plasma with frequencies on the order of 1 kHz would be rejected by the lock-in detection scheme, i.e., on the 100 kHz modulation time scale of the AOM the background light signal fluctuations at 1 kHz would be essentially constant and therefore result in a net null signal. However, if the oscillations result from a large and decaying initial oscillation (as in a damped oscillator), the change in oscillation amplitude as a function of time could result in a finite signal even with the lock-in detection scheme. Such an interpretation is consistent with the vanishing of the 1 kHz signal later in the discharge pulse, i.e., when the oscillation amplitude becomes more constant in time. To eliminate the 1 kHz oscillation, the raw data were digitally low pass filtered. The processed signals are shown in Fig. 3(c). A typical vdf (at $t=50$ ms into the discharge pulse) is shown in Fig. 3(d). The measurements are well fit by a single Maxwellian distribution. The time evolution of the vdf is shown in Fig. 4(a) with 100 μs time resolution. These higher time resolution measurements reveal features of the argon ion vdf that were not observed with 1 ms time resolution measurements: the signal amplitude and ion temperature increase slowly during the pulse and require approximately 45 ms to reach their steady state values [see Fig. 4(b)], the ion flow speed reaches its stationary value of ~1.9 km/s much more quickly (after approximately 25 ms), and an average ion flow of over 1.5 km/s appears within the first few hundred microseconds of the discharge. The LIF signal is roughly proportional to the ion density[12] and therefore the 45 ms time scale to achieve steady state LIF amplitude and ion temperature likely reflects the time necessary for the discharge to completely break down and reach a steady state ion density as well as heat the ions from room temperature to 0.4 eV. The more rapid ion acceleration suggests that the time scale needed to create the electric fields responsible for ion acceleration (discussed in Ref. 12) is shorter and distinct from the overall discharge evolution.

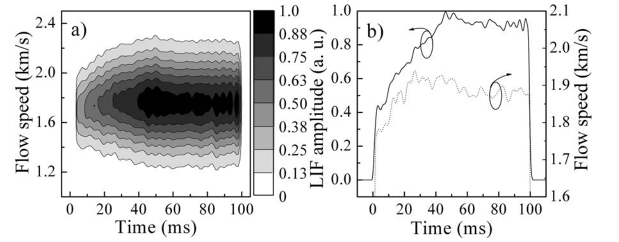

FIG. 4. (a) Contour plot of the evolution of the argon ion vdf during the 100 ms pulse with 100 μs time resolution. (b) Evolution of the LIF signal amplitude (continuous line) and ion flow velocity (dashed line) during the pulse.

The improved time resolution LIF technique described here has achieved high quality measurements at 100 μs with only minor modifications to a standard LIF diagnostic. Similar quality data at a time resolution of 30 μs are possible with the same electronics and slightly improved light collection. At lower time resolutions, the large oscillations in optical emission from the plasma had gone undetected. We note that the oscillation amplitude vanishes at approximately the same time that the ion acceleration ceases.

ACKNOWLEDGMENTS

The authors thank Professor Greg Severn from the University of San Diego for the loan of the AOM. This work was performed with support from NSF Grant No. PHY-0315356.

[1] R. A. Stern and J. A. Johnson III, Phys. Rev. Lett. **34**, 1548 (1975).
[2] D. H. Hill, S. Fornaca, and M. G. Wickham, Rev. Sci. Instrum. **54**, 309 (1983).
[3] X. Sun, A. M. Keesee, C. Biloiu, E. Scime, A. Meige, C. Charles, and R. Boswell, Phys. Rev. Lett. **95**, 025004 (2005).
[4] C. Biloiu, E. Scime, X. Sun, and B. McGeehan, Rev. Sci. Instrum. **75**, 4296 (2004).
[5] R. McWilliams, H. Boehmer, D. Edrich, L. Zhao, and D. Zimmerman, Thin Solid Films **8**, 113 (2005).
[6] D. Zimmerman, R. McWilliams, and D. A. Edrich, Plasma Sources Sci. Technol. **14**, 581 (2005).
[7] G. Bachet, L. Cherigier, C. Arnas-Capeau, F. Doveil, and R. A. Stern, J. Phys. III (France) **6**, 1157 (1996).
[8] B. Pelissier and N. Sadeghi, Rev. Sci. Instrum. **67**, 3405 (1996).
[9] G. Bachet, F. Skiff, M. Dindelegan, F. Doveil, and R. A. Stern, Phys. Rev. Lett. **80**, 3260 (1998).
[10] E. Scime, C. Biloiu, C. Compton, F. Doss, D. Venture, J. Heard, E. Choueiri, and R. Spektor, Rev. Sci. Instrum. **76**, 026107 (2005).
[11] C. Biloiu *et al.*, Plasma Sources Sci. Technol. **14**, 766 (2005).
[12] X. Sun, C. Biloiu, R. Hardin, and E. Scime, Plasma Sources Sci. Technol. **13**, 359 (2004).
[13] C. Davis, *Lasers and Electro-Optics: Fundamentals and Engineering* (Cambridge University Press, Cambridge, UK, 2000), p. 495.
[14] All about Bragg angle errors in AO modulators and deflectors: Application Note IM 1022, www.ISOMET.com