Xenon ion laser-induced fluorescence using a visible tunable diode laser near 680 nm

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Laser-induced fluorescence (LIF) measurements have been performed for the first time in a low temperature ($T_e \approx 0.6$ eV) Xe plasma using a tunable diode laser in the visible range of wavelengths. The transition in Xe II involved the $(^3P_1)5d[3]_{7/2}$ metastable state and the excitation wavelength was found to be 680.570±0.001 nm (air). LIF measurements of I_2 in a room temperature iodine gas cell were used to monitor the wavelength of the laser during the measurements. © *2007 American Institute of Physics.* [DOI: 10.1063/1.2813880]

This paper reports the first measurements of laser-induced fluorescence (LIF) signals in a low temperature Xe plasma obtained with a diode laser based diagnostic operating in the visible range of wavelengths, near 680 nm. Measuring ion velocity distribution functions (ivdfs) in xenon plasma discharges is of importance in a wide range of research endeavors including laser physics,[1] electric propulsion including Hall-effect thrusters (HETs),[2–4] and plasma-boundary physics, which includes a wide variety of plasma processing applications.[5–9] The LIF diagnostic technique for measuring ivdfs (Refs. [10–12]) is well established for use in plasma discharges generally and in xenon discharges, in particular.[2–4] The technique has been based entirely on dye laser technology in the visible range of wavelengths. Diode laser based diagnostics[13,14] hold many advantages over dye laser based diagnostics, but accessible LIF schemes must be shown to exist in order for these advantages to be realized. Unfortunately, most of the LIF schemes in the xenon ion that require excitation with visible light are inaccessible to readily available diode lasers. Further, as will be shown, there was reasonable doubt whether the one scheme that is easily accessible would actually work. Nevertheless, we describe a working LIF diagnostic for xenon ions in laboratory plasma discharges using a diode laser tuned to a wavelength near 680 nm.

The LIF diagnostic used to accomplish the measurements is depicted in Fig. 1. The light source for the diagnostic is a single extended cavity diode laser[15] in the Littrow configuration (Sacher TEC-100-680-20), capable of an output power of 20 mW and mode hop free tuning of about 5 GHz. Detuning from the line center was accomplished with an external voltage ramp. The laser beam serially passed through a room temperature iodine cell and the laboratory plasma to measure the iodine spectra and the Xe II fluorescence at the same time. The iodine spectra obtained with the gas cell were used as a wavelength calibration.[16] The Xe II fluorescence collected with a photomultiplier tube was filtered by a 50% transparent interference filter[17] cen-

tered at 492±0.2 nm with a full width at half maximum (FWHM) of 2 nm; the wavelength was chosen to exploit the transition from the excited state of the LIF scheme with the greatest probability of spontaneous decay. The LIF scheme itself is an essential part of the technology of the LIF technique and will be discussed presently. The intensity of the laser beam and of the fluorescence was modulated using a mechanical chopper rotating at 3.1 kHz placed in between the iodine cell and the vacuum chamber. The fluorescence signal was enhanced relative to the background light noise using a lock-in amplifier (Stanford Research Systems SR830). The beam was steered to a Burleigh WA-1000 wavemeter in order to coarse-tune the laser and to make wavelength and frequency measurements.

The experiments were carried out in a dc multidipole plasma chamber which is described in detail elsewhere.[8] The cylindrical surface of the chamber was surrounded by 12 rows of permanent magnets to enhance the plasma density and uniformity. The xenon gas pressure was maintained at 0.45 mTorr and the neutrals were ionized by energetic electrons thermionically emitted from hot thoriated-tungsten filaments biased at −60 V with respect to the grounded chamber wall. The emission current was 0.75 A. A Langmuir probe was used to obtain the electron density and temperature in the bulk plasma. The electron density was 5.4×10^9 cm^{-3} and the effective electron temperature T_e was 0.61 eV.

There are a number of Xe II LIF schemes currently in use,[2–4] but the main scheme in the visible range of wavelengths requires dye laser excitation at 605.1 nm, a wavelength inaccessible to readily available diode lasers. This scheme $((^3P_2)5d[3]_{7/2} \rightarrow (^3P_2)6p[2]_{7/2}^0)$ makes use of a metastable state at 95 437.67 cm^{-1} above the ion ground state. The new Xe II LIF scheme $((^3P_1)5d[3]_{7/2} \rightarrow (^3P_1)6p[2]_{5/2}^0)$ takes advantage of the wide availability of tunable diode lasers operating in the visible range of wavelengths near 680 nm, and is shown in Fig. 2. The new scheme has one dominant spontaneous emission transition,[18] and depends on a significant population in the ion metastable state at 108 423.07 cm^{-1} in order to produce a fluorescence signal. Excitation is accomplished with a wavelength of approxi-
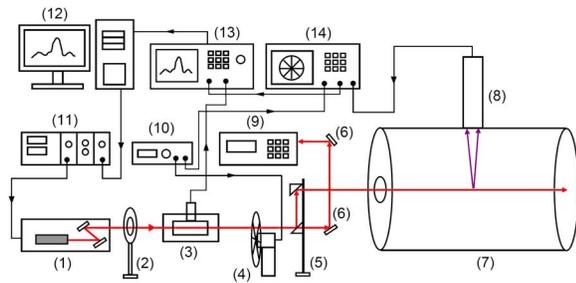
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FIG. 1. (Color online) Schematic of the LIF diagnostic measurement system: (1) laser head, (2) iris, (3) iodine cell, (4) mechanical chopper, (5) periscope, (6) mirrors, (7) chamber, (8) photomultiplier tube, (9) wavemeter, (10) chopper controller, (11) laser driver, (12) PC, (13) oscilloscope, and (14) lock-in amplifier.

mately 680.57 nm (in air). The detected photon comes from the transition $(^3P_1)6p[2]^0_{5/2} \rightarrow (^3P_1)6s[1]_{3/2}$ at 492.1 nm.

The new LIF scheme has an interesting history. Both the metastable and excited states can be found expressed in the *LS* coupling scheme as $5d^4F_{7/2}$, and $6p^4D^0_{5/2}$ in the tables compiled by Moore[19] in the 1950s. However, these proved to be associated with the wrong energy levels, nearly that of the now accepted levels[20] assigned to the doublet states $5d^2F_{7/2}$, and $6p^2D^0_{5/2}$. Prompted by persistent discrepancies in measured lifetimes and transition probabilities in Xe II, a new assessment of spectroscopic terms and levels was published by Hansen and Persson[21] in 1987. They found that *jK* coupling described many levels better than *LS* coupling, and they made designations of new levels as well. One of new
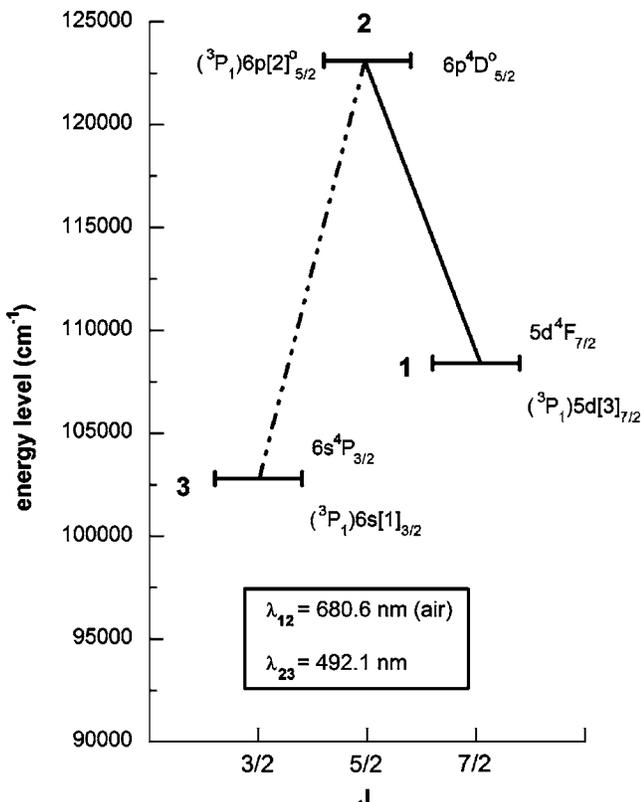

FIG. 2. LIF schemes for Xe II fluorescence used in the work reported here in low temperature plasmas. We label the states with the *LS* and *jK* notations for ease of identification, although the *jK* notation is now preferred. The excitation transition is distinguished by the solid line.
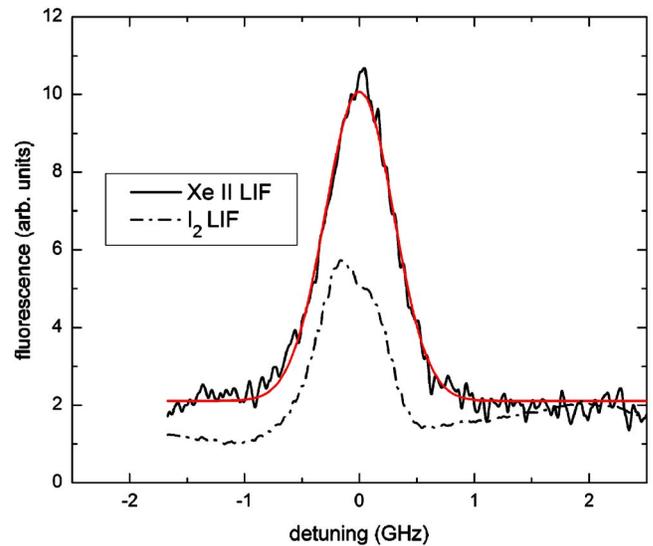

FIG. 3. (Color online) Xenon II and molecular iodine fluorescence as a function of detuning frequency from the line center.

levels, at 108 423.07 cm^{-1}, a metastable state, was designated as $(^3P_1)5d[3]_{7/2}$, with the best *LS* coupling designation, albeit considerably more imperfect, being $5d^4F_{7/2}$. However, in footnote 57 of their paper, the authors claimed that the quartet state $(6p^4D^0_{5/2})$ could not be the excited state for the 680.574 nm transition, yet it is, as we and others have discovered experimentally. In that footnote, they assigned $(^3P_2)6p[3]_{5/2}$ as the excited state for that transition, but in Table III of the same paper they assigned $(^3P_1)6p[2]^0_{5/2}$ as the excited state, a state designated in the *LS* scheme as $6p^4D^0_{5/2}$. This is the first recognition of this particular inconsistency. Mitchell *et al.*,[22] who along with Broström *et al.*,[18] did indeed discover that the quartets described above had been mistaken for the doublets, referenced this very footnote in their sleuthing about lifetime measurements of excited state in question, but seemed not to notice the error.

The LIF signal obtained as function of frequency is shown in Fig. 3. These measurements were taken in the center of the vacuum chamber, far from any sheaths, and where the electric field was essentially zero. If we assume no hyperfine splitting, the ion thermal temperature that corresponds to the observed Doppler broadening (587 MHz) is roughly 450 K, very close to the wall temperature of the vacuum chamber. We will return to this assumption presently. The iodine fluorescence was compared with the atlas of Salami and Ross.[16] Our laser was able to tune stably without mode hops and without other forms of hysteresis over about 4−5 GHz. Patching the iodine spectra together as well as possible over a range of 20 GHz, and paying attention to the asymmetries of the lines, we found a sequence of peaks with symmetries and gaps that matched the atlas of Salami and Ross in the range of wavelengths between 680.570 and 680.590 nm. It proved easier to shift the atlas spectrum to match our measured iodine spectrum, and as a result, we found that the best match was obtained by shifting the atlas by +0.0111 nm (about 7 GHz), as shown in Fig. 4. The calibration offset will of course differ from wavemeter to wavemeter. The important point is that the line center of our Gaussian Xe II LIF signal was coincident with a feature of
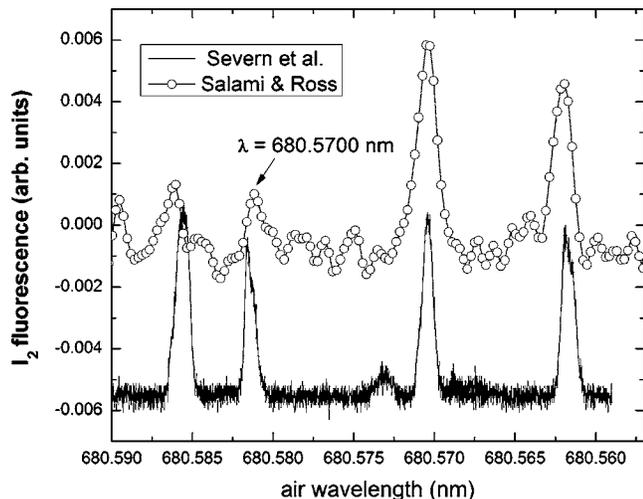
FIG. 4. Comparison of experimental molecular iodine fluorescence spectrum with the absorption spectrum of Ref. 16, corrected for air wavelengths. The feature in the atlas indicated by the arrow is also the location of the line center of the Xe II fluorescence signal. Reading the wavelength on the abscissa one sees that the calibration offset between wavemeter wavelength measurements and the atlas is about 0.01 nm, for this particular wavemeter.

the iodine spectrum corresponding to a wavelength of 680.570 ± 0.001 nm (air).

The accepted lifetime τ_l of the excited state is 7.2 ns.[22] We estimate[23] that saturation intensity, $I_{sat}=3hc/(\pi\lambda^3\tau_l)$ ≈ 9 mW/cm^2, where h is Planck's constant, c is the speed of light, and λ is the excitation wavelength. Given a roughly ellipsoidal beam area of 0.5 ± 0.1 cm^2 at the location of the focus of the photomultiplier tube collection optics, and estimating a roughly 10% loss through the vacuum view port, we estimate that the laser power measured at the laser head at which saturation should be observed is roughly 5 mW. This was corroborated experimentally; we found that the LIF signal at fixed frequency was linearly increasing up to an output power between 5 and 6 mW, and that above this output power the LIF signal flattened out considerably.

The hyperfine structure (hfs) constants A and B are not known for the metastable state $(^3P_1)5d[3]_{7/2}$; therefore, the hfs for the transition is unknown. The standard LIF scheme, however, is well understood[24,25] in that regard. The isotope shifts, which are small compared to the hfs splittings and small even compared to the expected Doppler broadening at room temperature (480 MHz). The hfs splittings, however, for the two odd Z isotopes (129, 131) cover a span of 6 GHz roughly centered on the mass 132 peak. Holt *et al.*[24] found that some of the hfs peaks are half as intense as that of the mass 132 peak. It is speculation of course to suppose the hfs splittings for the new LIF scheme studied here would be of similar magnitudes. We observed no other peaks in the Xe II fluorescence other than that shown in Fig. 3 within ± 10 GHz of the observed line center. That we see no other peaks suggests that the amplitudes of the other hfs components are in the noise. Without knowing the A and B hyperfine structure constants for this transition, one cannot perform a deconvolution analysis to obtain the ivdf from LIF signal. On the other hand, since we can only see the signal from the dominant isotopes, our LIF signal is a good approximation to the ivdf. This assumption is validated in that the ivdfs which we infer from LIF signals such as that shown in Fig. 3 do indeed satisfy the Bohm criterion near a negatively biased plate in a pure Xe plasma.[9]

Our summary result is that we have observed a useful LIF signal with excitation at 680.570 nm (air) arising from the Xe II metastable state $(^3P_1)5d[3]_{7/2}$ for the first time with a tunable diode laser in a low temperature xenon plasma. We would like to point out that finding a LIF signal with the fast ion beam-laser technique of Refs. 18, 22, 24, and 25 was no guarantee that a LIF signal could be measured in a low temperature xenon plasma with the diagnostic described in this paper. On the other hand, finding a useful fluorescence signal with the new LIF scheme and new diagnostic in a laboratory plasma makes the likelihood of finding this new LIF scheme useful for higher temperature, higher density HET plasmas quite high.

[1] M. Jánossy, P. Mezei, and P. Horváth, Opt. Commun. **65**, 287 (1988).
[2] W. A. Hargus, Jr. and M. A. Cappelli, Appl. Phys. B: Lasers Opt. **72**, 961 (2001).
[3] N. Dorval, J. Bonnet, J. P. Marque, E. Rosencher, S. Chable, F. Rogier, and P. Lasgorceix, J. Appl. Phys. **91**, 4811 (2002).
[4] R. J. Cedolin, W. A. Hargus, Jr., P. V. Storm, R. K. Hanson, and M. A. Cappelli, Appl. Phys. B: Lasers Opt. **65**, 459 (1997).
[5] H. Tanaka, K. Akinaga, A. Takahashi, and T. Okada, Jpn. J. Appl. Phys., Part 2 **43**, L585 (2004).
[6] T. Ide, J. Hashimoto, K. Nozaki, E. Mizuta, and T. Baba, Jpn. J. Appl. Phys., Part 2 **45**, L102 (2006).
[7] P. Y. Oh, J. H. Lee, S. H. Jeong, H. S. Moon, S. B. Lee, K. B. Song, Y. Jung, Y. Kim, G. Cho, H. S. Uhm, and E. H. Choi, IEEE Trans. Plasma Sci. **34**, 317 (2006).
[8] D. Lee, G. Severn, L. Oksuz, and N. Hershkowitz, J. Phys. D **39**, 5230 (2006).
[9] D. Lee, N. Hershkowitz, and G. D. Severn, Appl. Phys. Lett. **91**, 041505 (2007).
[10] R. A. Stern and J. A. Johnson III, Phys. Rev. Lett. **34**, 1548 (1975).
[11] D. N. Hill, S. Fornaca, and M. G. Wickham, Rev. Sci. Instrum. **54**, 309 (1983).
[12] A. Fasoli, T. N. Good, F. Anderegg, F. Skiff, P. J. Paris, M. Q. Tran, and M. Yamada, Phys. Rev. Lett. **63**, 2052 (1989).
[13] G. D. Severn, D. A. Edrich, and R. McWilliams, Rev. Sci. Instrum. **69**, 10 (1998).
[14] A. Keesee and E. Scime, Rev. Sci. Instrum. **75**, 4091 (2004).
[15] Sacher Lasertechnik LLC, Lynx Tunable Littrow External Cavity Diode Laser Instruction Manual (TEC100) Hannah Arendt Str. 3–7, 35037 Marburg, Germany, 2006.
[16] H. Salami and A. J. Ross, J. Mol. Spectrosc. **233**, 157 (2005).
[17] Dell Optics, 71 Orange Ave., Walden, NY 12566.
[18] L. Broström, S. Mannervik, A. Passian, and G. Sundström, Phys. Rev. A **49**, 3333 (1994), see Fig. 4, in particular.
[19] C. J. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, DC, 1958), Vol. 3.
[20] E. B. Saloman, J. Phys. Chem. Ref. Data **33**, 765 (2004).
[21] J. E. Hansen and W. Persson, Phys. Scr. **36**, 602 (1987).
[22] R. E. Mitchell, S. D. Rosner, T. J. Scholl, and R. A. Holt, Phys. Rev. A **45**, 4452 (1992).
[23] C. J. Foot, *Atomic Physics* (Oxford University Press, Oxford, 2005), p. 142.
[24] R. A. Holt, S. D. Rosner, and T. D. Gaily, Phys. Rev. A **15**, 2293 (1977).
[25] L. Broström, A. Kastberg, J. Lidberg, and S. Mannervik, Phys. Rev. A **53**, 109 (1996).