

Argon ion laser-induced fluorescence with diode lasers

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Diode lasers have been used for ion temperature measurements in ArII plasmas by finding new laser-induced fluorescence (LIF) schemes suited to the present range of available wavelengths. The new LIF schemes require excitation at 664, 669, and 689 nm, all near industry-standard wavelengths. Conventional LIF measurements performed by dye lasers in ArII use 611.66 nm in vacuum, shorter than any commercially available red diode laser line, and depend on the population of the $3d' \ ^2G_{9/2}$ metastable state. The metastable state density of the conventional LIF scheme was found to be larger than the populations of the other metastable states by an order of magnitude or less. A master oscillator power amplifier diode laser was used both in a Littman–Metcalf cavity and as an optical amplifier for a low power diode laser which was in a Littman–Metcalf cavity. Both systems provided intensity of up to 500 mW, continuously tunable over 10 nm centered at 666 nm, and were used to obtain high resolution ion velocity distribution functions. © 1998 American Institute of Physics. [S0034-6748(98)04201-4]

I. INTRODUCTION

Diode laser technology has advanced to the point of being useful for basic plasma dynamics research in noble gas discharge plasmas. Already diode lasers are used extensively for atomic and molecular spectroscopy, and recent reviews document their virtues^{1–4} in that regard. The use of diode lasers specifically for plasma physics has been slower to develop, even though they have been used for spectroscopic studies in plasmas for years. While quantum physics issues have been the primary reason for those studies, they have nevertheless shown the usefulness of diode lasers for measuring the sort of physical quantities relevant to plasma dynamics, such as number density of a given atom or molecule, and its velocity space distribution functions, as shown by Uzelac⁵ in an Ar discharge. Typically the atoms so diagnosed have been neutral, but the laser induced fluorescence (LIF) measurement technique is independent of the of the charge state of the analyte atom.

LIF techniques have been used for plasma physics measurements for over twenty five years,^{6,7} and were made possible by the advent of high power tunable single mode dye lasers, and applied first to *Q*-machine plasmas with rare earth ions with well populated metastables states easily accessible with various dyes. Suitable LIF schemes found in noble gas discharges have been exploited for plasma dynamics measurements for over a decade.⁸ But before dye lasers, the most common instrument used to obtain ion velocity distribution functions (ivdfs) was the gridded energy analyzer.⁹ Goekner *et al.*¹⁰ used a dye laser to measure the ion temperature in a hot filament discharge plasma (ArII) and compared it with the results from gridded energy analyzers available in the literature. The analyzers always gave anomalously high values, and Goekner's work confirmed that indeed the ion temperature in those devices was essentially room temperature and that the energy resolution of the analyzers were of order

0.1 eV, much too large to measure room temperature ions (.025 eV) accurately. The speed resolution of the gridded energy analyzer, for argon ions, is approximately 10^3 m/s. The velocity resolution of cw dye laser systems and diode laser systems can achieve sub-Doppler resolution, approaching the ultimate limit imposed by the natural linewidth of the excitation transition.

The point of this work is to compare the usefulness of the diode laser with that of the dye laser for making LIF measurements of ivdfs in argon plasma discharges. Diode lasers have linewidths which are 10–100 times sharper than that of dye lasers, and are cheaper and easier to use. For ArII discharges, the Doppler width, $\Delta\nu_d = \nu_0 \sqrt{(8kT \ln 2)/mc^2}$, varies from 0.9 to 5.5 GHz as the ion temperature varies from 0.025 to 1 eV. For dye lasers and diode lasers, the laser linewidth is orders of magnitude smaller than the Doppler width. The difference in velocity resolution between diode and dye laser systems is insignificant if their linewidths are much smaller than the natural linewidth of the excitation transition. The true velocity resolution of a laser system ultimately depends on the convolution of the laser line and the spectral line associated with the excitation transition of the LIF scheme one is interested in. In order to make our comparison definite, we have in mind a diode laser system composed of a low power ($P < 10$ mW) extended cavity diode laser, optical isolators, polarization rotator, and a tapered chip optical amplifier; this is a system without active frequency stabilization. A comparable dye laser system,¹¹ for example, a tunable ring dye laser, also without active frequency stabilization, has a linewidth about the same as many natural linewidths, between 10 and 20 MHz. This is the range of natural linewidths involved in the LIF scheme in ArII that we examine in this article. Because of the narrower diode laser linewidth, the velocity resolution of the diode laser system is about a factor of two sharper than the dye system. A dye laser with active frequency stabilization will have velocity resolution comparable to the diode laser system.

Dye laser systems exceed the cost of comparable diode

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laser systems by a factor of 5.¹¹ Diode laser systems simply plug into the wall in any random laboratory room, and are air or thermoelectrically cooled. Of course, the coarse tuning range of diode laser systems cannot compare with dye laser systems. The primary limitation of diode laser systems is that there are gaps in the spectrum of available wavelengths. But this is a limitation only if the spectral line(s) of interest happens to fall in one of those gaps. Happily, the gaps are shrinking. As late as the last decade, visible laser diodes were not available. Though much of the visible spectrum remains inaccessible, red diode lasers are available from roughly 630 nm to the infrared.¹² The interests of the spectroscopy community is of very little moment to diode laser manufacturers who respond primarily to the needs of larger industries (e.g., fiber optics communications, laser pumping, printers and data storage, remote sensing and interferometry, HeNe laser replacement). One is best served by looking for useful spectral lines in the range of available wavelengths. Available power is then an issue, since in discharge plasmas commonly used in basic plasma physics research, there is a fair amount of background light. Even then one must be sure that lasing modes can be tuned across the spectral line of choice and that mode hops do not prohibit the diode laser from reaching the target wavelength. This problem can be eliminated by placing the diode laser, if one or more facets have been AR coated, in a simple external optical cavity.^{2,13,14} The minimum power requirements, typically on the order of tens of mW, can be met by use of tapered optical amplifiers.¹⁵ One still needs to find a useful line, and the target wavelength depends upon the choice of working gas.

We describe the ArII LIF schemes relevant for commercially available diode laser systems in Sec. II A. The plasma confinement device used for the experiments is discussed in Sec. II B along with the diode laser systems LIF measurements. We present the ivdfs obtained with diode lasers in Sec. III. In an era of diminishing research support, it is handy to find a more cost effective diagnostic tool than the conventional one. Since the new tool is also simpler to use, we hope that a wider number of researchers will be attracted to its use.

II. EXPERIMENTAL SYSTEM DESCRIPTION

A. LIF schemes for ArII

A partial energy level diagram for ArII, shown in Fig. 1, depicts the commonly used LIF scheme. The laser induced excitation is from the $3d' \ ^2G_{9/2}$ metastable state to the $4p' \ ^2F_{7/2}^0$ and requires a photon of vacuum wavelength 611.66 nm. The observed fluorescent photon at 461.08 nm signals the atomic transition to the $4s' \ ^2D_{5/2}$ state and then to the ground state. The plasma is immersed in a magnetic field in the experiments reported here and the lines exhibit weak field Zeeman splitting. They are also Doppler broadened. For perspective, the maximum wavelength detuning, for this LIF scheme, between the farthest σ lines is ± 17 GHz/T, which in a field of 1 kG corresponds to a width of 1.7 GHz (or 1.2×10^{-3} nm), and is similar to the Doppler width of each ArII line at a temperature of 0.1 eV.

One of the ways that diode laser systems are becoming cheaper is by limiting the tunability and providing a user

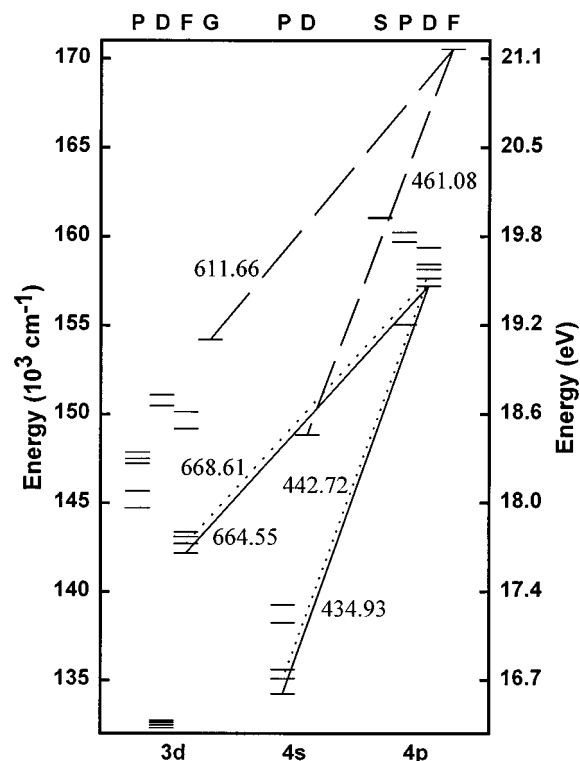


FIG. 1. A partial Grotrian diagram for singly ionized Ar, depicting the LIF schemes 1–3 in Table I.

specified center wavelength as NEW FOCUS has done. The tuning range, 100 GHz, specified by NEW FOCUS is smaller than the 120 GHz difference between the air and vacuum wavelengths (668.61_{vac} nm, 668.43_{air} nm) for the transition $3d \ ^4F_{7/2} - 4p \ ^4D_{5/2}^0$, part of an important LIF scheme in ArII. Clearly this difference must be taken into consideration in purchasing such a system. Further, the air wavelength of the same transition varies between that measured in San Diego, CA and Boulder, CO by 20 GHz, a significant fraction of the proposed tuning interval. Argon plasma LIF is done in vacuum. Specifying the vacuum wavelengths removes these potential difficulties, and we adopt this convention in this article. Of course, specifying the wavelengths at STP works as well. Our point really is that users and vendors of these sorts of diode laser systems must be careful about specifying the desired wavelength.

In any case, the 611.66 nm LIF scheme has three strong merits. First, for a variety of different plasma discharges such as microwave induced plasmas,¹⁶ ohmic discharges,¹⁷ and thermionic filament discharges,^{10,18} the scheme is found to provide an adequate LIF signal. These different discharges represent ion temperatures in the range of 0.025–5 eV, electron temperatures from 1 to 20 eV, ion densities of 10^9 – 10^{12} cm⁻³, for which the LIF signal is sufficient to determine ion temperatures and measure ion diffusion coefficients. Second, the wavelength for the excitation, 611.66 nm, is accessible at high power in single mode in dye lasers with a very long lived dye: Rhodamine 6G. This minimizes maintenance. Third, many photomultiplier tubes have their maximum sensitivity and minimum equivalent noise intensity between 400 and 500 nm. Apart from the high cost in terms of

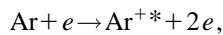
TABLE I. Summary of LIF schemes in ArII. For comparison, the standard scheme is included. This list of course is not exhaustive; only the schemes with the highest transition probabilities from the excited states have been included. The metastable state is state 1 in the three level LIF scheme. The fluorescence transition connects the excited state (state 2) with the state to which it decays (state 3). The absolute spontaneous transmission probabilities (A_{12}, A_{23}) and the branching ratio (β_{23}) for the fluorescence transition are also given. The last column gives the minimum velocity resolution (Δv) of that scheme. All wavelengths are given for vacuum conditions.

Scheme No.	Metastable state	Fluorescence transition	λ_{12} (nm)	λ_{23} (nm)	A_{12} (10^8 s^{-1})	A_{23} (10^8 s^{-1})	β_{23}	Δv (m/s)
1	$3d' \ ^2G_{9/2}$	$4p' \ ^2F_{7/2}^0 - 4s' \ ^2D_{5/2}$	611.66	461.08	0.200	0.789	0.665	12.2
2	$3d \ ^4F_{7/2}$	$4p \ ^4D_{5/2}^0 - 4s \ ^4P_{3/2}$	668.61	442.72	0.107	0.817	0.616	7.2
3	$3d \ ^4F_{9/2}$	$4p \ ^4D_{7/2}^0 - 4s \ ^4P_{5/2}$	664.55	434.93	0.147	1.171	0.810	9.8
4	$3d \ ^4F_{7/2}$	$4p \ ^4D_{7/2}^0 - 4s \ ^4P_{5/2}$	688.85	434.93	0.009	1.171	0.810	0.6
5	$3d' \ ^2G_{7/2}$	$4p' \ ^2F_{7/2}^0 - 4s' \ ^2D_{5/2}$	612.51	461.08	0.009	0.789	0.665	0.5
6	$3d \ ^4D_{7/2}$	$4p \ ^4P_{5/2}^0 - 4s \ ^4P_{5/2}$	440.22	480.74	0.304	0.780	0.574	13.4

both money and time in maintaining a dye laser system, it is an excellent LIF scheme.

A number of alternative LIF schemes^{19,20} are presented in Table I, along with the usual scheme for comparison. The schemes with excitation wavelengths near the diode laser industry standard wavelengths of 660, 670, and 685 nm jump off the page (schemes 2–4). Not every conceivable scheme has been included in the list, only those for which the fluorescent transition has the maximum branching ratio of all the allowed transitions. Fortunately, those listed all have fluorescent wavelengths between 400 and 500 nm and are characterized by branching ratios equal to or greater than that of the most commonly used scheme. The remaining concerns for making use of these alternative schemes for use with diode lasers are: (1) the population density of the alternative metastable states and, (2) the background light generated by the plasma close to the wavelength of the LIF emission lines. The amount of diode laser power required depends on these critical magnitudes.

The metastable state, $3d' \ ^2G_{9/2}$, some 34 eV above the ground state of the argon neutral atom is thought¹⁰ to be populated primarily by a single ionization event with an energetic electron,



because the metastable state density has been found to be proportional to the discharge current. A two step process, such as described by Cherrington as dominant in the relevant excitation processes in Ar^+ cw lasers²¹ would scale quadratically with the discharge current, since this quantity enters into the independent probabilities of ionization and excitation. The observed linear scaling with discharge current is consistent with single step creation of even higher excited states followed by radiative decays into the metastable state. This process is especially relevant for the background signal, where, for example, the cross section for excitation of the $4p' \ ^2F_{7/2}^0$, scheme 1, shows evidence²² of strong radiative cascade contributions. It is difficult to perform an *ab initio* calculation to assess the relative densities of the alternative metastable states compared with the $3d' \ ^2G_{9/2}$ state since the internal states of the ions are not in thermal equilibrium with plasma. That equilibrium is radically disturbed by the non-thermal energetic electron beam which creates the plasma.

These primary electrons are accelerated through a potential difference of 300 V in our experiment, roughly ten times the threshold potential for producing argon ions in the $3d' \ ^2G_{9/2}$ state. Given this amount of energy in excess of the required amount, it seems unlikely that the alternative metastables would have very different densities since they are energetically so close together. In any case, the means of plasma production is one particular aspect of the experimental apparatus, and to its detail we now turn.

B. Experimental apparatus

The experiments reported here were performed in the Irvine Torus,²³ a plasma discharge device with a thermionic emission plasma source. Argon was the neutral chosen for the experiments described here and the typical operating pressure was 1×10^{-4} Torr. Typical plasma parameters were as follows: plasma density, $n_e \leq 1 \times 10^{11} \text{ cm}^{-3}$, electron and ion temperatures, $T_e \leq 5 \text{ eV}$, $0.1 < T_i < 2 \text{ eV}$. The discharge was maintained usually with a 16 ml filament biased 300 V below (vacuum chamber) ground, with 100 mA of emission current.

Our wavelength and power requirements led us to try two related diode laser systems using a tapered amplifier, currently available from SDL (SDL 8630). The diode laser system functions in the fashion typical of diode laser chips, as the gain element bounding an extended cavity arrayed in a Littman–Metcalf geometry. It serves as a laser source capable of the highest cw power commercially available (0–500 mW), an order of magnitude higher than the extended cavity visible diode laser systems currently on the market, and is continuously tunable over a 10 nm bandwidth with a center wavelength specifiable between 665 and 675 nm. Such high power chips at visible wavelengths currently represent the state of the art of solid state fabrication technology. This system has the capability of lasing in a single longitudinal mode but this was difficult to realize in practice. The spectra is broadband, which is to say multiline, consisting of lasing cavity modes $\sim 1.7 \text{ GHz}$ apart modulated by the broad gain envelope of the diffraction grating, and the etalon formed by the chip facets giving a 17 GHz modulation. We shall call this laser system B.

Another useful feature of the tapered amplifier is that it

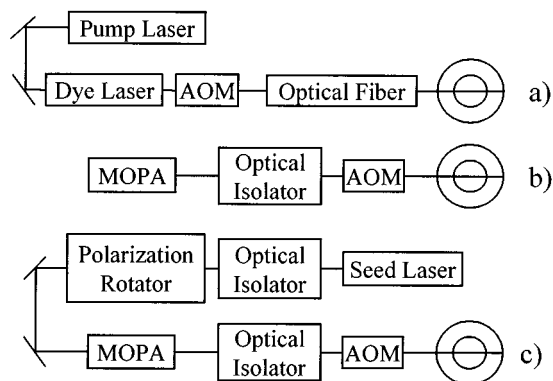


FIG. 2. The three lasers systems used for this experiment are: (a) the dye laser system A, (b) the tapered amplifier in an external cavity, system B, and (c) the seed laser combined with the tapered amplifier chip which formed system C.

can be injection locked to the output of a lower power diode laser which can operate with a single longitudinal mode, in a master oscillator power amplifier (MOPA) configuration. Marquardt *et al.*¹⁵ have in this way obtained gains as high as 100 while leaving the linewidth of the seed, or master oscillator, laser unchanged. The SDL 8630 can be easily modified to operate in this configuration by simply removing the grating and drilling a hole in the rf enclosure protecting the tapered amplifier chip (also called the MOPA chip), optical cavity, and output optics train.

Diode lasers are famous for extreme sensitivity to stray optical feedback and the MOPA chip is no different in this regard. Optical isolators capable of 30 dB of isolation are necessary for the system to operate safely and stably. We used a low power extended cavity diode laser system from NEW FOCUS as the seed laser. We will call this configuration system C, and the conventional dye laser arrangement is called system A. All three systems schematics are shown in Fig. 2. The beam passed through an I_2 gas cell before entering the plasma, and a LIF spectrum of I_2 was obtained as the laser was tuned. The I_2 fluorescence was collected with a NTE3032 photodiode. The collection optics for the fluorescence from the plasma was the same for each system, consisting of imaging lenses, IF filter, and photomultiplier tube (PMT). This is described elsewhere in this journal.¹⁸

III. RESULTS AND DISCUSSION

We found the schemes accessible to system C, with excitation at 664.55 and 668.61 nm, and show them in Fig. 3 against the backdrop of the LIF spectrum of I_2 , which was obtained in order to provide an absolute wavelength calibration. We were happy to find that LIF I_2 spectrum had a much higher signal to noise ratio than that available in a commonly used atlas of I_2 obtained by absorption spectroscopy,²⁴ although the regions surrounding 668.61 and 664.55 nm have relatively few identified absorption lines. The diode laser system C performed just as well as the dye laser system for the plasmas in our experiment. Ion velocity space distribution functions obtained for each system are compared in Fig. 4. Both the dye laser and the diode seed laser were continuously tuned while the stand alone tapered amplifier chip was

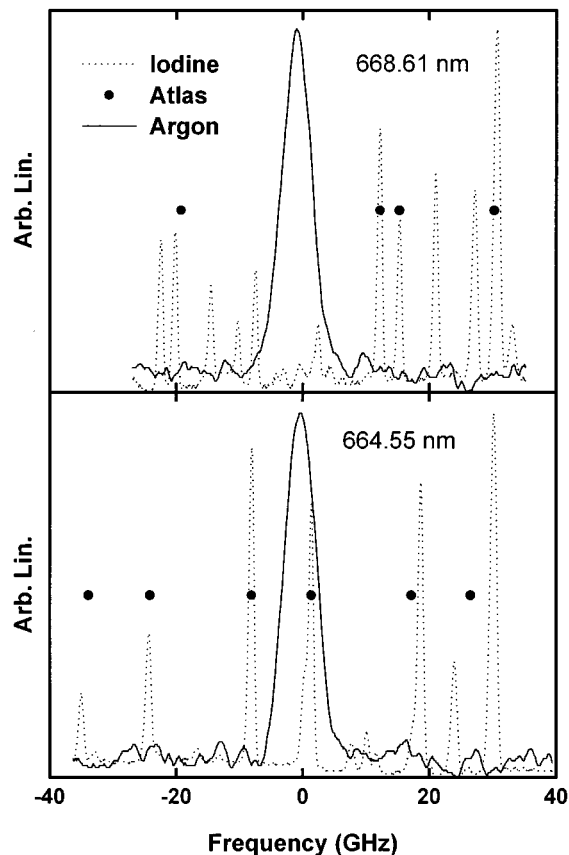


FIG. 3. LIF spectrum of ArII and I_2 about (top) scheme 1, requiring excitation at 668.61 nm, and (bottom) scheme 2, requiring excitation at 664.55 nm. The I_2 was used to obtain an LIF map of the surrounding lines of I_2 to aid in finding the ArII LIF schemes and to provide an absolute wavelength calibration.

temperature tuned through a discrete set of temperatures, limited by the resolution of the temperature controller. The signal to noise ratio obtained using the lock-in amplifier for systems A and C are identical. The diode laser systems can examine the ivdfs with marginally greater resolution than the dye laser owing to their smaller linewidth. The smoothness of the curves in this Fig. 4 is determined by the ability of the lock-in amplifier to eliminate the background light from the plasma. For these plasmas, the fluorescence signal is small compared to the background light, of order 10^{-2} for the dye laser operating in single mode with the conventional LIF scheme with an intensity between 15 and 30 mW. The lock-in technique need only eliminate 30–40 dB of noise to produce high quality ivdfs using the diode laser system C. Diode lasers certainly can begin to replace dye lasers where suitable absorption lines exist in the analyte ion.

The distribution function as produced by system B is inferior to the other two systems. This does not have anything to do with the linewidth of lasing modes, which are also very narrow for the MOPA acting as a stand alone diode laser, but stems from the multimode output of the laser. It was difficult to obtain a stable mode structure as the laser tuned. There was a fair amount of mode hopping. In Fig. 5, the mode spectra of the diode laser systems are compared. The modulation of the cavity modes due to the finite width of

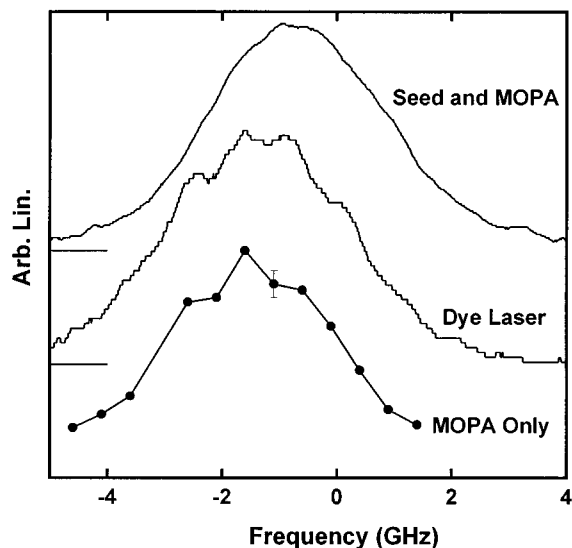


FIG. 4. A comparison of ivdfs obtained by (top) our dye laser system, system A, (center) system B, and (bottom) system C, the diode laser systems. For system B, tuning was accomplished by changing the temperature of the chip, which had a resolution of 0.1 °C.

the chip serve to reduce the number modes to actually produce fluorescence to just a few for system B, but there is a significant amount of power in laser lines detuned from the target line. Using the tapered amplifier chip injection locked to a low power, single mode seed laser gave the highest quality results of the two diode laser systems.

Tuning the seed laser was accomplished by ramping the voltage of a PZT wafer, controlled with (GPIB). A complete scan for the LIF schemes 2 and 3 can be accomplished by tuning through 80 GHz, which corresponds to roughly 15 ion temperatures at 1 eV, and could be done in about 5 s. A family of ivdfs obtained in this fashion is shown in Fig. 6. Each curve corresponds to a different incident laser intensity spanning the range from 5 to 100 mW for this particular data set. We varied the forward current through the tapered amplifier chip rather than the power of the seed laser. The seed

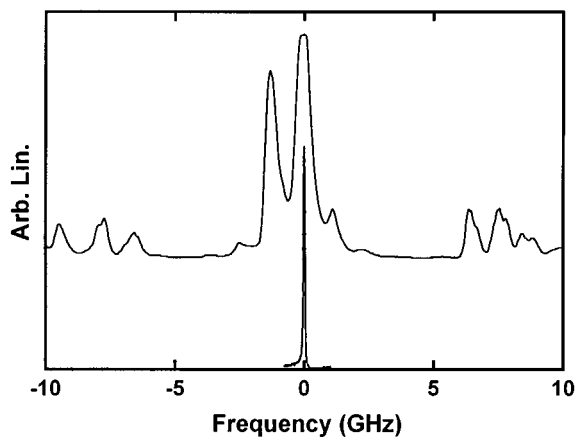


FIG. 5. A comparison of the mode structure of the diode laser systems: the top curve is the broadband MOPA chip output, system B, and the bottom curve is the seed laser plus MOPA chip, system C.

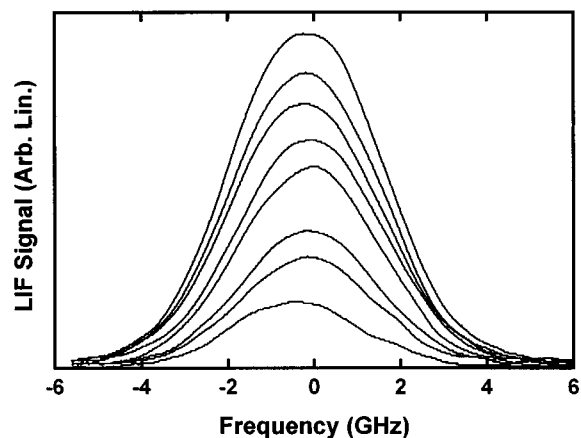


FIG. 6. A family of ivdfs obtain from system C by varying the output intensity; the values of which were in ascending order: 5.55, 11.13, 16.74, 33.18, 44.46, 67.92, 81.24, and 108.6 mW, respectively.

laser wavelength tunes slightly when its power is changed, a common feature of diode lasers. Marquardt¹⁵ has shown that this particular tapered amplifier chip saturates at about 500 mW for an input seed laser intensity between 2 and 3 mW when the forward current through the chip is near its maximum value of 2.2 A. Once one tunes to the line, obtaining ivdfs is a turn-key operation. One can shut the laser off and come back the next day and find the laser still tuned to the line! There is no need to rehearse what is true of dye laser systems. Tuning the diode laser, however, can lead to unwanted shifts in the output intensity if the seed laser tunes through a maximum of a chip mode of the MOPA laser. But this is a problem only if the MOPA is operated near its power limit, otherwise the constant power feature of the MOPA current controller works well.

We found that the LIF signal strength varied quadratically with input laser power at 668.61 nm, with system C, as shown in Fig. 7(a). This is as expected when the incident intensity is such that the stimulated emission is significant compared with optical pumping, or the total spontaneous emission rate to the other states. The saturation intensity as given by

$$I_{\text{sat}} = \frac{8\pi hc^2}{\lambda_{21}^5} \frac{\sum_j A_{2j}}{A_{2j}} \delta\lambda,$$

is about 0.1 mW/mm² at 668 nm and a linewidth of 100 kHz well below the spectral intensities used in our experiments with laser system C, and thus consistent with our results. We observe that the power broadening of the ivdf, shown in Fig. 7(b), is still fairly weak. There are therefore competing effects evidenced here. The relatively narrow linewidth available in diode laser systems serve to modestly increase velocity space resolution, while the need for reasonable signal to noise ratio implies a modest power broadening, and therefore a modestly reduced velocity space resolution.

Given that the LIF signal is proportional to the metastable state density

$$I_{\text{LIF}} \propto n P_l \lambda^3 A_{23} \beta \int g(v) dv \Rightarrow n \propto \frac{V_{\text{ac}}}{P_l \lambda^3 A_{23} \beta},$$

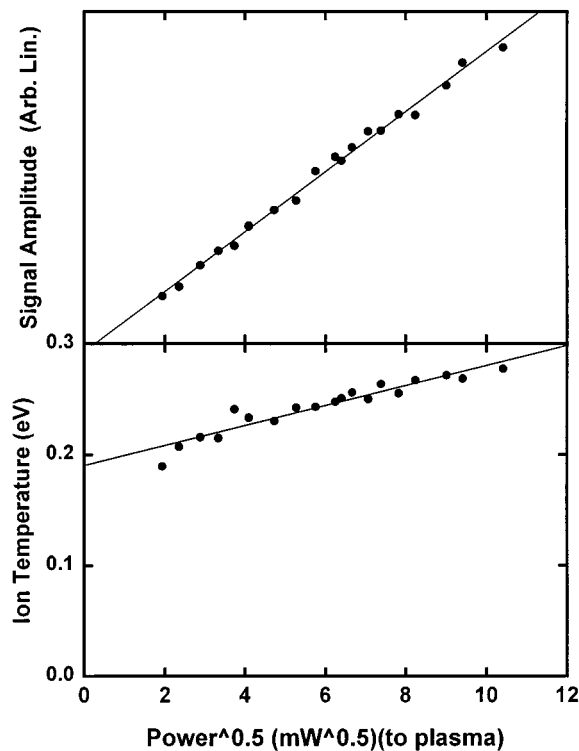


FIG. 7. The LIF signal depended quadratically on the incident laser intensity for system C (top), consistent with our estimate of the saturation intensity of 668.61 nm. The ion temperature (bottom) shows the slight effects of power broadening. The ion temperature is from a Gaussian fit to the ivdf and is uncorrected for Zeeman splitting.

where the V_{ac} is the voltage measured using a lock-in amplifier, the light signal in phase with a 50% duty cycle modulation of the beam, A_{23} , and β are the atomic transition probability of the fluorescent transition and the branching ratio, respectively, and P_l is the input laser intensity, we found that $n_{F_{7/2}}/n_{G_{9/2}} \approx 0.1$, and $n_{F_{7/2}}/n_{F_{9/2}} \approx 1$. The first ratio gives the ratio of the metastable state population of an alternative LIF scheme (664.55 nm excitation) to the metastable state population of the commonly used one (611.66 nm excitation). This ratio involves LIF measurements with different interference filters and thus the inference is indirect. We believe that the alternative metastable state is less well populated than the standard one in the UCI plasma, but the gap is not very wide, perhaps one order of magnitude difference or less. The second ratio given above shows that the metastable state densities for alternative schemes 2 and 3 are essentially unity, and the inference is direct, since the optics train for the fluorescence measurement is identical for both measurements.

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- ¹¹The comparison we are thinking of is between two systems that can be tuned with an external voltage ramp, that do not make use of external reference cavities. The diode laser system (tunable low power diode laser system, e.g., NEW FOCUS Vortex 6000 at \$8000, MOPA laser, SDL 8630 at \$18 000, Newport MOPA controller, Model 6000, at \$5000, and two Optical Isolators, NEW FOCUS Model 5568 at \$6000) is nominally \$37 000. The dye laser system (tunable ring dye laser, e.g., Coherent 899-05 at \$76 000, ion argon pump laser, Coherent Innova TSM 15 at \$80 000,) costs nominally \$156 000, a factor of 4.2 greater. In addition, for the dye laser system, one must arrange for a heat exchanger and substantial electrical power which may increase that factor by quite a bit.
- ¹²See diode laser reference guide on the web page of Thor Labs, Inc., <http://www.thorlab.com/>. Industry standard wavelengths in the red are 635, 650, 670, 675, 680, 685, and 690 nm. These are solitary diode lasers. Tuneable systems made from these fill in the gaps in this range.
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