Laser Chem. Vol. 12, pp. 137–143
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A NEW TECHNIQUE FOR ULTRAFAST VELOCITY DISTRIBUTION MEASUREMENTS OF ATOMIC SPECIES BY POST-IONIZATION LASER INDUCED FLUORESCENCE (PILIF)

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(*Received 20 September 1991; in final form 15 October 1991*)

A new method for single shot velocity distribution measurement of metallic impurities of relevance for studies involving continuous sources, such as limiter experiments in fusion devises or sputtering experiments, based in the combination of Resonant Enhanced Multiphoton Ionization (REMPI) and Laser Induced Fluorescence (LIF) is proposed. High ionization yield and good time resolution are expected according to the numerical simulation of the experiment that has been run for several atomic species. Other possible applications of REMPI to plasma edge physics and to conventional techniques for velocity distribution measurements are briefly addressed.

1. INTRODUCTION

Velocity distributions, of great relevance in many fields of physics and chemical physics, can be readily measured by Time of Flight (TOF) techniques,[1] provided that free collision conditions exist in the region between the source and the detector. The accuracy of the method is restricted by the finite sharpness of the gating function and the finite dimensions of the detection volume, so that long flight distances are needed for good resolution, in detriment of the signal to noise ratio. On the other hand, Doppler shifted LIF spectroscopy has been widely used for this purpose,[1] although the full scan on the laser wavelength required for the velocity distribution measurement implies a continuous character of the experiment and narrowing the laser bandwidth is made on expenses of pulse energy.

In plasma fusion research, velocity distribution of neutrals are important not only for the evaluation of impurity fluxes but also to determine the mechanism responsible for their ejection.[2] The continuous character of the flow of sputtered particles and the impossibility to chop it make time of slight (TOF) techniques not applicable to *in situ* velocity measurements in fusion plasma experiments so that Doppler shifted excitation in LIF detection of neutrals is the only method extensively used until now for this purpose.[3] In general these measurements require many, reproductible, plasma discharges and new methods based on fast scanning of the dye laser frequency during a single discharge have been developed as alternative.[4] In any case velocity resolution has to be gained on expenses of the signal versus noise ratio and correction for the laser power at each wavelength is always needed due to the low saturation parameter required for these experiments.

In the present work a new method for single shot, *in situ* velocity distribution measurements based on REMPI in combination with Laser Induced Fluorescence (LIF), of relevance for impurity flux determinations in fusion plasma research is proposed, altogether with some other applications.

2.1 PILIF Experiment

The proposed experiment consist on crossing two laser beams (the ionizing and probing ones) in the scattering volume and to record the time evolution of the LIF signal after complete ionization has taken place. After the detection volume has been depleted of neutral atoms by the REMPI process an spatial hole in terms of neutral density is formed. As the sputtered atoms start to fill it, the density in the observation volume will continuously increase and therefore the LIF signal when used as a density diagnostic, i.e. bandwidth greater than the Doppler profile. This last requirement restricts the proposed experiment to those atomic systems where metastable levels do not act as a sink of the laser-populated level, as in 3 level systems.

Assuming a well collimated atomic beam with a given velocity distribution $f(v)$ and a scattering volume with dimensional parallel to the travelling direction ℓ the density at a given time after ionization is given by:

$$n(t) = n(0) \times \left[t/\ell \int_0^{\ell/t} f(v) v \, dv + \int_{\ell/t}^{\infty} f(v) dv \right] \tag{1}$$

where $n(0)$ stands for the density of neutrals before ionization takes place.

The accuracy of the velocity distribution obtained with method will be limited by that of the scattering volume dimensions and its resolution by the dimensions themselves and the minimum sampling interval, ultimately limited by the lifetime of the excited level, providing that ionization takes place in a large extend during the ionizing pulse.

The velocity distribution function, $f(v)$, can be reconstructed from the time evolution of the LIF signal by applying the expression:

$$f(v) = -\frac{1}{n(0)} \times \frac{t^3}{\ell} \times \frac{\delta^2 n(t)}{\delta^2 t} \tag{2}$$

where v in the case of a point source, far away from the scattering volume, is simply given by ℓ/t.

The most appealing way to carry out the experiment would be by using the same experiment set-up as for LIF detection. This in many instances consists of a dye laser pumped by an excimer one, typically XeCL at 308 nm, so that a 4.03 eV high power photon source is readily available. This photon energy combined with that of the pumping photon is able to bring all the neutral atoms to the ionization continuum in most of the metals typically monitored in limiter experiments. *Table 1* shows some of them:

Table 1

Atom	*Excitation, nm (eV)*	*Ionization (eV)*
Fe I	302.0 (4.11)	3.79
CrI	428.9 (2.92)	3.85
TiI	293.3 (4.23)	2.61
BeI	234.9 (5.28)	4.04

In order to completely ionize all the atoms in the sampling volume, the right splitting of the excimer power into direct (ionizing) beam and pumping one has to be made. That in principle will depend on the particular atomic system under consideration, but a simple calculation based on the rate equations for a three level system plus ionization[5] shows that if, for example, a 0.5 Jul/pulse XeCl laser and a ratio 1:1 between the power used to pump the dye laser and that of the ionizing beam are used, so that a modest 50 μJul/pulse UV radiation is obtained after frequency doubling, still high enough to saturate the resonant transition (S = 150 for Be, S = 100 for Fe, focussing in $2 \times 2\,mm^2$), ionization will take place to a 100% in a time shorter than the laser pulse (15 to 20 nsec), even if the excimer radiation is focussed in an area several times larger, thus minimizing alignment problems. As an example, Figure 1 shows the results for the Fe atom assuming an excimer laser square pulse of 20 nanoseconds and an ionization cross section, for the excited atom, of $10^{-18}\,cm^2$.

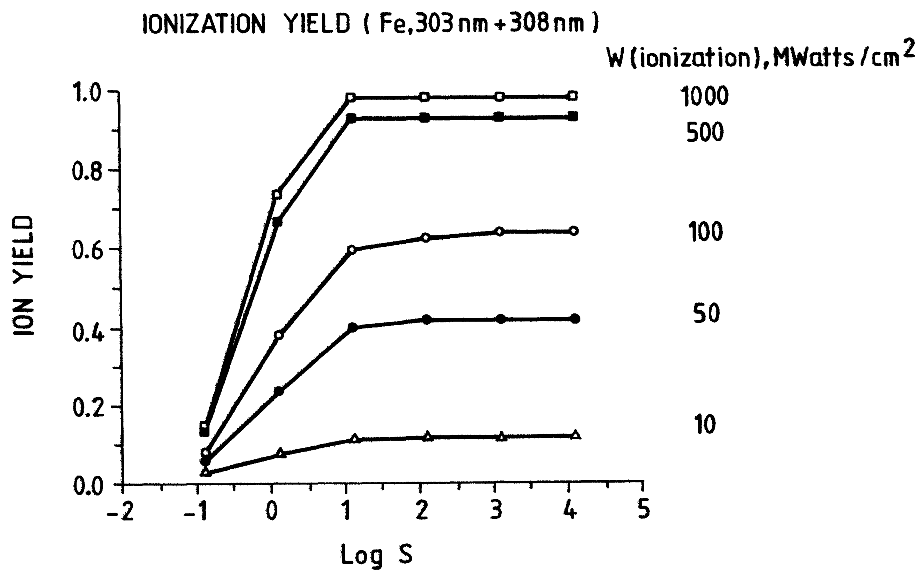

Figure 1 Calculated ionization yield of Fe atoms as a function of probe and ionizing laser powers at the end of a 20 nanoseconds pulse. $U_{Saturation} = 70\,KWats/cm^2 \times nm$.

The conditions used for the simulation are similar to that used for REMPI detection of Fe atoms in sputtering experiments[6]. As it can be seen a high degree of ionization is expected even at relatively low saturation parameter and modest excimer laser power.

Sampling of the LIF signal in a relevant time scale could be achieved by optically delaying successive reflections of the probing laser or by using a long pulse dye laser (several hundred nanoseconds) and crossing the excimer at the beginning of the pulse, thus obtaining a continuous LIF signal. The first scheme implies long distances (optical delay > pulse duration) and correction for the laser divergence, so that the second scheme will be more feasible. Due to the short time required by the TOF exeriment (see below) synchronization of the two lasers should not be critical.

2.2 Model Calculation for Extended Sources

The time evolution predicted by Eq. 1 is not directly applicable to extended sources as one has a limiter experiments. Convolution over all the emitter area and the different paths across the sampling volume depending on geometry, as well as attenuation through the plasma edge has to be taken into account.

The resuls of the calculation for a Be bar limiter, where a 0.6 cm diameter hole is used to look at the scattering volume, a prism of $2 \times 2 \times 4$ mm placed at 1 cm from the limiter in this simulation (see Figure 2) are displayed in Figure 3. Plasma edge temperature and density profiles as well as ionization and excitation rate constants are the same as in Ref. 7. No contribution to the refilling of the hole due to CX or electron recombination is considered as they are expected to be not fast enough to effectively complete with the direct flux of sputtered atoms in the relevant time scale. A cosine distribution for sputtered particles is assumed.

As it can be seen in Fig 3a, discrimination between sputtering (Thompson model) and thermal distributions should be obvious even in a short time after the ionizing pulse ($t = 0$). It must be recalled at this point that accurately measuring the Doppler profile for a thermal distribution requires an extremely narrow laser bandwidth, not always available. A higher resolution in this case, by PILIF will be easily achieved by simply enlarging ℓ. A factor of two in the binding energy will be also distinguishable after several tens of nanosecond. Figure 3b shows the velocity distributions for these two cases (Eb = 3.32 eV and 1.66 eV respectively) as they would be measured by Doppler shift in the same geometry and plasma edge conditions.

3. EXPERIMENTAL SET-UP

The available experimental set-up, where the proposed experiment will be undertaken, has been previously described.[8] Basically, a DC Glow Discharge in an inert gas (He, Ar) is produced, the stainless steel chamber acting as the cathode (= 17% Cr). The sputtered Cr atoms are detected by LIF at 429 nm (two level system) by using an excimer pumped (XeCl) dye laser (Lambda Physik LPX 205i- *ibid*. FL 3002E). The available bandwidth can be reduced from 0.2 to 0.04 cm^{-1} by an intracavity etalon,

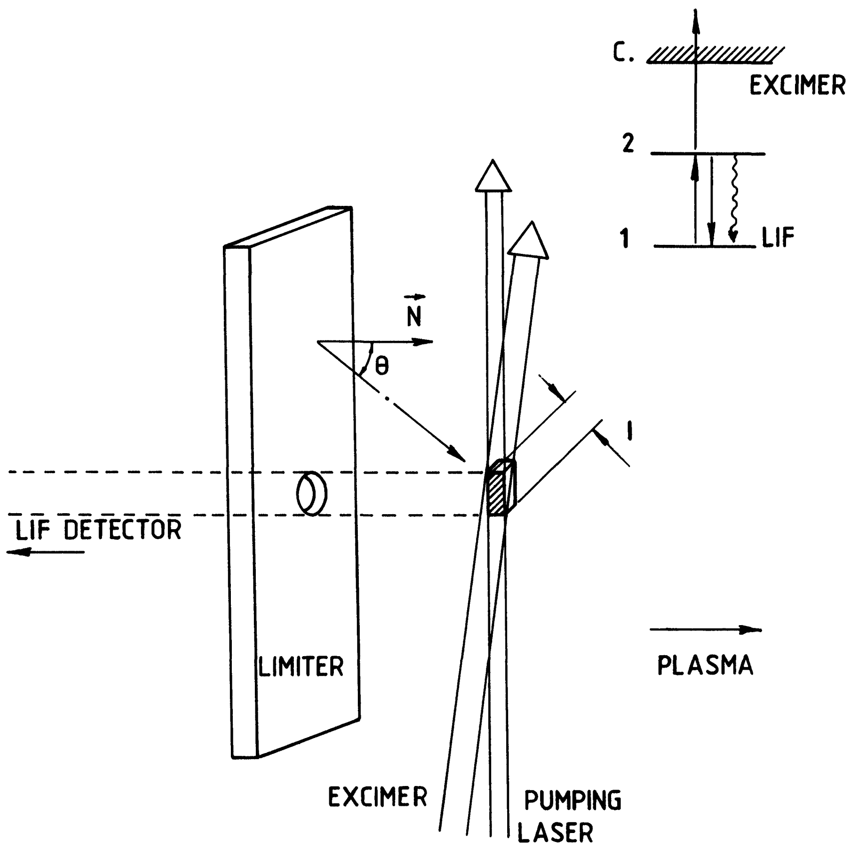

Figure 2 Geometry assumed in the model calculation for a Be limiter. The interaction with the plasma is assumed to take place within a rectangle of 2 cm wide and 4 cm high. See text for the rest of the parameters used.

so that a velocity resolution of ≈ 0.5 Km/s can be obtained for the standard Doppler shifted LIF experiment (see Figure 3b), providing that saturation of the transition is avoided ($S = 0.6\,kW/cm^{-2}\,s^{-1}$). Splitting of the excimer radiation at 308 nm will still allow 200 mJ/pulse for the ionizing beam and > 20 mJ/pulse for pumping of the transition, so that the required conditions for the REMPI-LIF experiment can be easily fulfilled, even without focussing of either laser beam. A flashlamp-pumped dye laser (1 J/pulse, pulse duration $\approx 1\,\mu s$, bandwidth ≈ 1 nm), presently under construction, will be used for "long time" excitation under saturation conditions. The LIF time evolution will be recorded in a fast digital oscilloscope (Tektronix DSA 601,1 GS/s) and the data transferred to a PC. Synchronization of the excimer and the dye laser beams will be achieved by optically delaying part of the 429 nm radiation used to trigger the excimer laser via a fast photodiode.

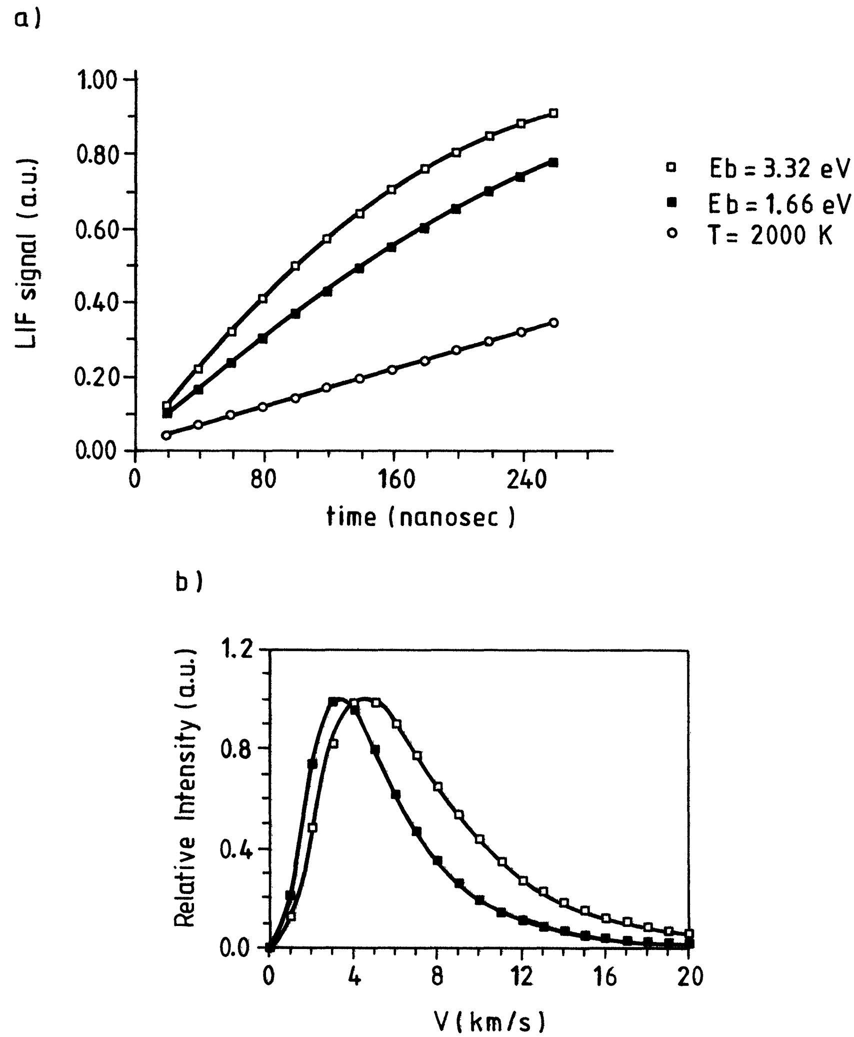

Figure 3 Results from the model calculation: (a) Time evolution of the LIF signal. Thompson model for Eb (Be) = 3.32 eV (top) and Eb = 1.66 (middle) and thermal distribution (bottom) for $T = 2000$ K. (b) Velocity distribution along the detection line of sight for the Eb = 3.32 and Eb = 1.66 eV cases for the same geometry as above.

4. OTHER EXPERIMENTS

The good spatial and temporal resolution of laser diagnostics could be used in the REMPI experiments to probe the plasma edge. Although no detailed calculations have been performed yet, the screening properties of the plasma edge could, in principle, be tested without perturbing other plasma parameters in combination with neutral atomic beam diagnostics. Besides, REMPI could be used to create a highly located, time resolved high intensity pulse of single charged ions which propagation through the plasma, followed by optical methods, could yield information concerning particle transport, among others.

In atomic beams experiments, the proposed ionization scheme could be used to optically chop the beam with an extremely narrow equivalent gate function, thus improving the velocity resolution in the conventional TOF experiment.

Acknowledgements

The author would like to thank Dr. J. Hackmann, Dr. C. Niesband and Dr. M. Bessenrodt.Weberpals for the very useful discussion and suggestions about the proposed experiments during the author's stay at the Düsseldorf University and to the TJ-I team.

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