Laser induced fluorescence monitoring of atomic hydrogen densities and velocities in an expanding cascaded arc plasma

Citation for published version (APA):
Boogaarts, M. G. H., Brinkman, G. J., Heijden, van der, H. W. P., Vakan, P., Mazouffre, S., Mullen, van der, J. J. A. M., & Schram, D. C. (1997). Laser induced fluorescence monitoring of atomic hydrogen densities and velocities in an expanding cascaded arc plasma. In *proceedings of the 8th International symposium on Laser-aided plasma diagnostics, Doorwerth, The Netherlands, 22-26 September 1997* (pp. 109-114)

Document status and date:
Published: 01/01/1997

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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LASER INDUCED FLUORESCENCE MONITORING OF ATOMIC HYDROGEN DENSITIES AND VELOCITIES IN AN EXPANDING CASCADED ARC PLASMA

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In an expanding plasma created by a cascaded arc, spatially resolved densities, temperatures, and velocities of atomic species are obtained by applying two-photon excitation laser induced fluorescence. In order to obtain absolute number densities the set-up has been calibrated for atomic hydrogen using a titration in a flow tube reactor. In addition, the axial velocity of atomic hydrogen has been measured in the supersonic expansion of an Ar/H-plasma.

1 Introduction

When a cascaded arc created plasma is expanded into a vacuum vessel, a versatile high quality particle beam is obtained that has been shown to have very promising features.[1,2] An important application of such an expanding plasma is the fast deposition of e.g. amorphous silicon (a-Si:H) or carbon (a-C:H).[2,3] For these applications, but also from a fundamental point of view, it is interesting to study the composition of the beam, i.e. the degree of dissociation, excitation, and ionization, and to understand it in terms of the elementary plasma processes that take place.

Several laser-based diagnostic techniques are used to probe the local particle densities in the expanding plasma and their energy distribution. For the species-selective detection of atoms the very sensitive 'laser induced fluorescence' (LIF) technique can be used. However, due to the large energy spacings involved, monitoring of atoms in their ground-state requires the use of high energetic photons and the application of experimentally demanding techniques that are necessary for the generation of these photons. Problems connected with single-photon excitation of atoms can be avoided with the application of multi-photon excitation. Several schemes for 'two-photon excitation laser induced fluorescence' monitoring of atoms in plasmas (also refered to as 'two-photon absorption laser induced fluorescence' (TALIF)) have already been demonstrated for e.g. H,[4] O,[5] and N.[6] A major disadvantage of the LIF technique remains the difficulty to interpret the signals in terms of absolute number densities.

In this contribution measurements are presented on atomic hydrogen in the expanding plasma of a cascaded arc using two-photon excitation and detection of the resulting non-resonant laser induced fluorescence. The corresponding energy level scheme is shown in Fig. 1. H is excited with two 205.14 nm photons from the 1s ^2S ground state to the 3d ^2D and 3s ^2S states. The excitation is monitored by detection of the fluorescence on the Balmer-α line at 656.3 nm.

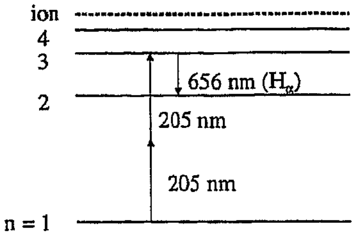

Fig. 1: Energy level scheme for the two-photon excitation laser induced fluorescence measurements on atomic hydrogen.

In order to obtain absolute number densities from these measurements, the fluorescence set-up is calibrated for atomic hydrogen by comparing the H-signal from the plasma with that from a flow tube reactor where in the latter the amount of H can be determined by titration.[7] The absolute number densities of the atomic species are not only important to gain information on the fundamental plasma processes and conditions, but also to obtain a quantitative idea of the sensitivity of this 2-photon LIF technique for various atomic species.

In addition the axial velocity of H in the supersonic expansion is measured by accurately determining the Doppler-shift with a frequency-calibrated laser. These measurements are important for e.g. the interpretation of the density data in terms of atomic flux and degree of dissociation.

2 Experimental

A scheme of the experimental set-up is depicted in Fig. 2. The cascaded arc plasma source has already been described in detail elsewhere.[8] In the experiments described in this paper it is operated on a 40 A dc current and a gasflow of 3.0 slm Ar and 0.5 slm H_2. The plasma expands from a 4 mm diameter channel into a roots-blower pumped vessel with a background pressure of 7 Pa. The plasma conditions for this case are close to those used for deposition of a-C:H and a-Si:H. The cascaded arc plasma source is mounted on a translation arm. Spatial scans through the expanding plasma can be made by moving the cascaded arc relative to the intersection of laser beam and detection volume.

The laser system that is used to produce the tunable UV-radiation is based on a pulsed (50 Hz repetition rate) Nd:YAG/dye-laser combination (Spectra-Physics, GCR-230/PDL-3). The tunable dye-laser has a bandwidth of 0.07 cm^{-1} and is used around 615 nm. The output of the dye-laser (typically 60 mJ) is frequency-tripled in two stages using first a KD*P-crystal and then a BBO-crystal resulting in typically 0.5 mJ of tunable UV light around 205 nm with an estimated bandwidth of 0.2 cm^{-1}.

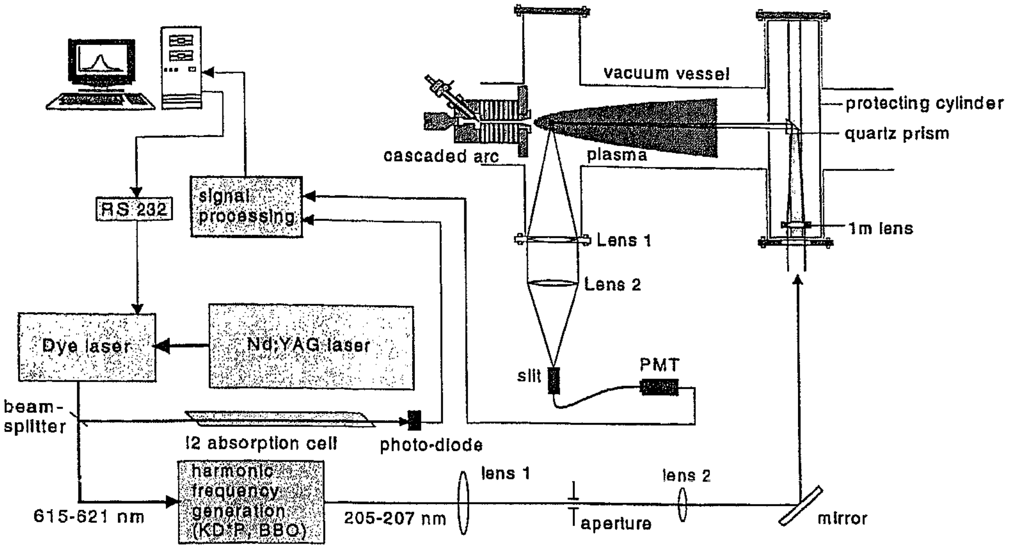

Fig. 2: Scheme of the experimental set-up. The cascaded arc plasma source is mounted on a translation arm that allows for spatially resolved measurements.

The frequency-tripled laser light is focussed into the plasma with a 60 cm lens. The laser induced fluorescence originating from the focus is imaged in a direction perpendicularly to the laser beam, via a slit of 2 × 0.2 mm, onto a photo-multiplier (Hamamatsu, R928). The slit is placed parallel to the laserbeam and defines the spatial resolution. The continuous background light emitted by the plasma is strongly reduced by a H_α bandpass filter in front of the photo-multiplier tube (PMT). The background signal is further reduced by using a gated integrator that accumulates the PMT-signal only during a time interval of some 200 ns around the arrival of the laser pulse.

All fluorescence measurements are performed by tuning the excitation laser and recording the resulting spectral profile of the two-photon absorption for various axial and radial positions. The spectral scans are all fitted to a Gaussian profile. From these fits three local parameters can be obtained. The local density is obtained from the integrated intensity (i.e. the area under the curve), while the local temperature is derived from the Doppler-width of the spectral profile. In addition, the component of the velocity in the direction of the laser can be determined from the absolute shift of the center frequency of the spectral profile.

The density profiles measured via laser induced fluorescence are relative. To obtain absolute atomic number densities, the LIF set-up needs to be calibrated using an independent source of a well-known amount of atomic hydrogen. This is accomplished via a titration in a flow tube reactor.[7] Atomic H is produced in the flow tube reactor in a diluted He/H_2-flow by dissociation of H_2 in a microwave cavity. H is then transported by the He-flow via an inert tube to a position close to the scattering volume of the laser where it is detected in the same way as in the expanding cascaded arc plasma. To determine the amount of H present in the scattering volume, a fast titration reaction for hydrogen: $H + NO_2 \rightarrow NO + OH$, with a reaction coefficient of $1.3.10^{-10}$ $cm^3/s\cdot$molecule, is used.[7]

NO_2 can be added via a small insert that enters the main transport tube near to the end of it. The original amount of H is then deduced from a careful measurement of the NO_2-flow needed to let disappear the H-signal. By scaling the resulting H-density with the ratio between the integrated LIF intensity of the cascaded arc plasma and that of the flow tube reactor, we are able to determine the spatially-resolved absolute H number density in the expanding Ar/H plasma. The accuracy of the density values is at this moment limited to about 50%, which is mainly due to the incertainty in the determination of the pressure in the vessel.

To be able to determine the axial velocity of atomic hydrogen in the expansion of the cascaded arc plasma, the LIF set-up is adapted so as to excite H on the symmetry axis of the expansion with a counterpropagating laserbeam (Fig. 2). The slitmask for the PMT is also rotated over 90° to keep it parallel with the laserbeam, and the laser is now focussed with a 1m lens to be able to keep the lens well away from the plasmabeam. In order to obtain quantitative velocity data, the wavelength of the laser needs to be calibrated. This is done by using a part of the dye-laser beam for the simultaneous recording of the absorption spectrum of molecular iodine at the fundamental dye-laser freqyency. The I_2 absorption lines in this region are accurately tabulated.[9]

3 Results

A simultaneous recording of the H LIF signal and the I_2 absorption spectrum is shown in Fig. 3a (top left). By comparing each I_2 absorption line with the literature value, the horizontal axis of the spectral scan can be determined with a minimum error of 0.01 cm^{-1}. The line center (velocity), width (temperature) and integrated intensity (density) of the H two-photon transition are derived from a Gaussian fit through the LIF data.

The axial density profile of H is shown in Fig. 3b (bottom left). This density plot is measured with the laser perpendicular to the plasma beam axis. The density scale is calibrated using the flow tube reactor. The H-density on the axis is about 5.10^{20} m^{-3} close to the nozzle, and decays rapidly with distance from the nozzle. In the first part of the expansion the axial density profile can be fitted well to an exponential decay with a 1/e length of 4 mm, while for larger distances the decay becomes less pronounced. To determine if and where the H-density will reflect a shock, the measurements need to be extended to still larger distances.

Fig. 3c (top right) shows a plot of the axial H-velocity as a function of distance from the nozzle. The velocity reaches a maximum of 3800 m/s, which agrees well with the most probable velocity in the supersonic expansion[10] of a gasmixture at a temperature of 1 eV and with an average mass that matches that of the Ar/H_2 mixture used in these experiments. This implies a complete coupling of H and Ar in the expansion region where the plasma is accelerated. Part of this acceleration may be visible in the figure. The minimum standard deviation in the velocity measurements is 300 m/s.

In Fig. 3d (bottom right) the temperature, as derived from the same spectral scans as the data in the Fig. 3c, is plotted against distance. The temperature on the axis starts at about 2500 K near the nozzle and decreases until a minimum of about 700 K is reached at distances between 20 and 60 mm. After that the temperature sharply rises again. From this figure we get first evidence for the shock to lie around 80 mm. Simple gasdynamic formulas can be used to derive from these temperature data the local acoustic velocities assuming a continued coupling between H and Ar. These values are indicated in the velocity plot as well. A comparison of these estimates with the experimental velocities yields another indication that the plasma shocks (i.e. becomes subsonic) at or shortly after 80 mm.

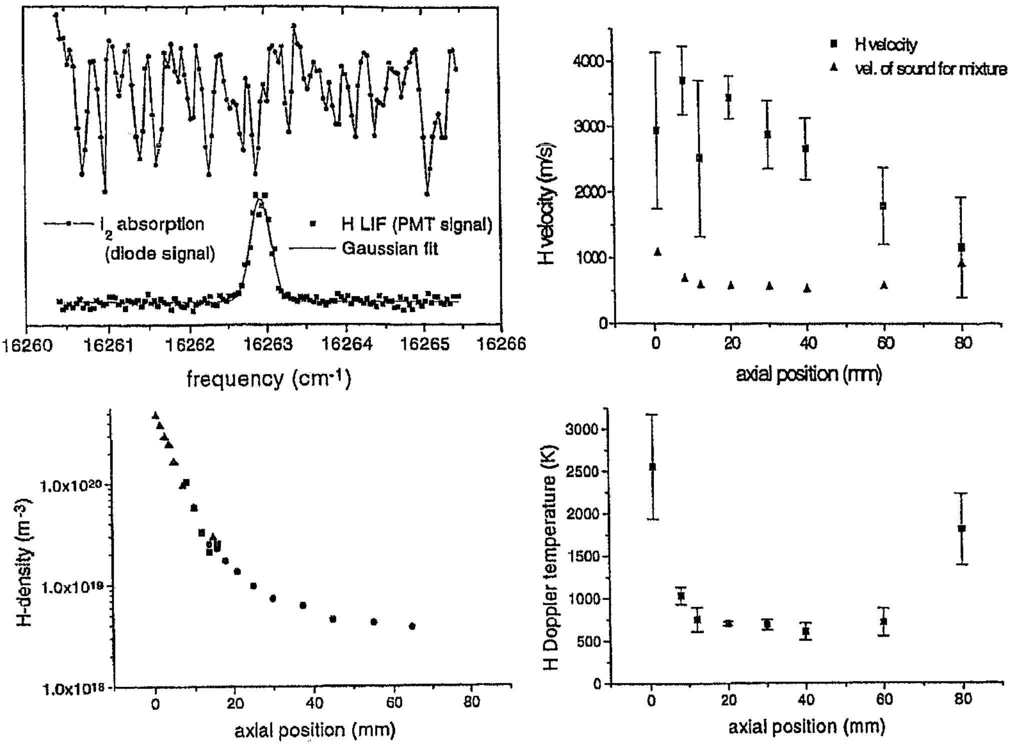

Fig. 3: a (top left): Spectral scan with simultaneous recording of the H LIF signal (lower trace) and the I_2 absorption signal (upper trace). b (bottom left): Calibrated axial profile of the H-density. c (top right): Axial H-velocity as a function of distance from the nozzle. d (bottom right): H-temperature as a function of distance from the nozzle. The data in these graphs have been acquired with the laser anti-parallel with the plasma beam axis, except for the density data in graph b that have been measured in a perpendicular configuration.

4 Conclusions

Two-photon excitation laser induced fluorescence measurements enable the spatially-resolved determination of the density, the temperature, and the velocity of atoms. This LIF technique has been applied to monitor atomic hydrogen in the expanding plasma of a cascaded arc operating on an Ar/H_2-mixture.

The setup has been calibrated for H using a titration in a flow tube reactor. The H number density in the Ar/H-plasma has a maximum of 5.10^{20} m^{-3} near the nozzle for operating conditions that are typical for deposition research. From these measurements it is concluded that the detection limit for atomic hydrogen of our fluorescence setup lies well below a density of 5.10^{17} m^{-3}.

The axial velocity of H in the supersonic expansion has been measured in a configuration with a counterpropagating laser. In these measurements the laser-frequency is calibrated by simultaneously recording the I_2 absorption spectrum. The observed maximum velocity of 3800 m/s implies a complete coupling of Ar and H in the acceleration region.

The interpretation of the data in terms of net atomic flux and dissociation efficiency of the cascaded arc is currently being undertaken. For this it is important to determine also the radial density profiles of the atoms, which is one of the next objectives of our research.

Acknowledgement

This work is financially supported by the "Stichting Technische Wetenschappen (STW)". The authors would like to thank Dr. U. Czarnetzki and coworkers at the University of Essen for using their flow tube reactor, and H.M.M. de Jong, M.J.F. van de Sande and A.B.M. Hüsken for their skillful technical assistance.

References

1. R.F.G. Meulenbroeks, R.A.H. Engeln, M.N.A. Beurskens, R.M.J. Paffen, M.C.M. van de Sanden, J.A.M. van der Mullen, and D.C. Schram, *Plasma Sources Sci. Technol.* 4 (1995) 74.
2. M.C.M. van de Sanden, R.J. Severens, R.F.G. Meulenbroeks, M.J. de Graaf, Z. Qing, D.K. Otorbaev, R. Engeln, J.W.A.M. Gielen, J.A.M. van der Mullen, and D.C. Schram, *Suface and Coatings Technology* **74-75** (1995) 1.
3. M.C.M. van de Sanden, R.J. Severens, J.W.A.M. Gielen, R.M.J. Paffen, and D.C. Schram, *Plasma Sources Sci. Technol.* **5** (1996) 268.
4. U. Czarnetzki, K. Miyazaki, T. Kajiwara, K. Muraoka, M. Maeda, and H.F. Döbele, *J. Opt. Soc. Am. B* **11** (1994) 2155.
5. A. Goehlich, T. Kawetzki, M. Katsch, E. Quandt, and H.F. Döbele, *Proc. Frontiers in Low Temperature Plasma Diagnostics II, Bad Honnef* (1997) p. 139.
6. G.J. Brinkman, S. Mazouffre, H.F. Döbele, M.G.H. Boogaarts, J.A.M. van der Mullen, and D.C. Schram, *Proc. Frontiers in Low Temperature Plasma Diagnostics II, Bad Honnef* (1997) p. 97.
7. D.W. Setser, *Reactive Intermediates in the Gas Phase* (Academic Press, New York, 1979).
8. M.C.M. van de Sanden, J.M. de Regt, G.M. Janssen, J.A.M. van der Mullen, D.C. Schram, and B. van der Sijde, *Rev. Sci. Instum* **63** (1992) 3369.
9. S. Gerstenkorn and P.Luc, *Atlas du spectroscopie d'absorption de la molecule d'iode* (CNRS, Paris, 1978); S. Gerstenkorn and P.Luc, *Rev. Phys. Appl.* **14** (1979) 791.
10. G. Scoles, *Atomic and molecular beam methods* (Oxford University Press, 1988).