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# *In situ* deuterium inventory measurements of a-C:D layers on tungsten in TEXTOR by laser induced ablation spectroscopy

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**Abstract**

Laser induced ablation spectroscopy (LIAS) is a diagnostic to provide temporally and spatially resolved *in situ* measurements of tritium retention and material migration in order to characterize the status of the first wall in future fusion devices. In LIAS, a ns-laser pulse ablates the first nanometres of the first wall plasma-facing components into the plasma edge. The resulting line radiation by plasma excitation is observed by spectroscopy. In the case of the full ionizing plasma and with knowledge of appropriate photon efficiencies for the corresponding line emission the amount of ablated material can be measured *in situ*. We present the photon efficiency for the deuterium Balmer $\alpha$-line resulting from ablation in TEXTOR by performing LIAS on amorphous hydrocarbon (a-C:D) layers deposited on tungsten substrate of thicknesses between 0.1 and 1.1 $\mu$m. An experimental inverse photon efficiency of $[\frac{D}{XB}]^{\text{a-C:D}\xrightarrow{\text{LIAS}}\text{D}}_{\text{D}_\alpha(\text{EXP})} = 75.9 \pm 23.4$ was determined. This value is a factor 5 larger than predicted values from the ADAS database for atomic injection of deuterium under TEXTOR plasma edge conditions and about twice as high, assuming normal wall recycling and release of molecular deuterium and break-up of $D_2$ via the molecular ion which is usually observed at the high temperature tokamak edge ($T_e > 30\,\text{eV}$).

Keywords: laser impact on surfaces, plasma diagnostic techniques and instrumentation, nuclear fusion power, plasma–material interactions, excitation and ionization by electron impact, safety, spectroscopy in chemical analysis

(Some figures may appear in colour only in the online journal)

## 1. Introduction

Plasma–wall interaction plays a key role for future fusion reactors, as the first wall lifetime and tritium retention are crucial factors for safety and economic performance. Therefore methods to characterize *in situ* material deposition and fuel retention on the wall in fusion devices are urgently needed for ITER, W7X and future fusion devices. Laser-based methods are promising candidates and are investigated for first wall characterization in fusion devices [1–4]. This work focuses on laser-induced ablation spectroscopy (LIAS) [5]. The steps performed for LIAS are shown in the flow chart in figure 1; example data can be found in [6].

In LIAS, a selected plasma-facing material deposit of unknown composition is ablated by intense laser radiation in

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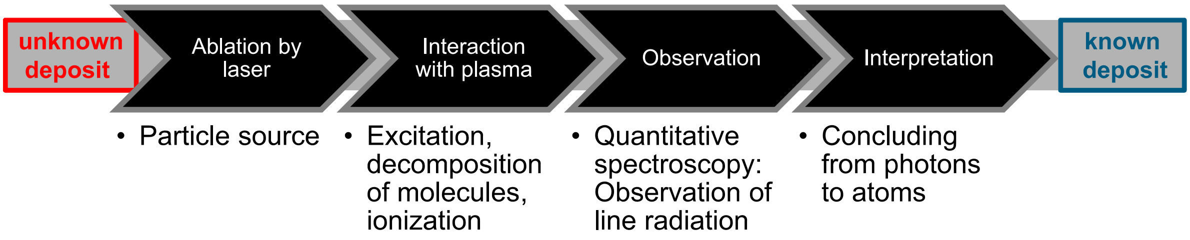

**Figure 1.** The principle of LIAS.

**Table 1.** Experimental conditions for LIAS.

| Experiment | a-C:D layer thickness (nm) | Side-view camera | Experiment time (ms) | $H_\alpha$ filter FWHM (nm) | Laser spot area ($mm^2$) | $F_{Laser}$ ($J\,cm^{-2}$) |
|---|---|---|---|---|---|---|
| A | 180, 520, 780 | Pike F-032, Allied Vision Technology | 5 | 3.24 | $24 \pm 3$ | 6.2 |
| B | 94, 344, 505, 1108 | PROXITRONIC camera with MCP | 20 | 0.81 | $16 \pm 3$ | 3.2 |

the presence of the fusion edge plasma. The ablated particles entering the plasma edge are excited and then observed by optical spectroscopy. From the detected photon flux of dedicated species, the composition, the amount of ablated material as well as the retained fuel can be determined. It is required that all emission occurs inside the observation volume of the full ionizing plasma. The thickness is concluded by identification of the transition from the (co-)deposit to the underlying bulk material. If the deposit is formed from the same chemical element as the bulk material (e.g. Be on Be) determination of the transition to the bulk material is still possible if the materials differ in their removal rates. The removal rates will differ due to hydrogen co-deposition in the layer (e.g. Be + D) or differences between thermal conductivity and density of deposit and bulk material. A common approach for interpretation of the observed light by utilizing inverse photon efficiencies [7], available for many species in the ADAS database, is routinely employed to measure impurity and fuel particle fluxes [8]. In this report, we show that the observed inverse photon efficiency of deuterium from LIAS of amorphous hydrocarbon (a-C:D) layers is different from an atomic or molecular deuterium source in the plasma edge, but in agreement with values observed for the release of deuterium from deuterated hydrocarbons.

## 2. Experimental methods

To investigate the performance of LIAS, pre-deposited samples were used to ablate known amount of particles into TEXTOR. The light emitted by the material is then observed by radiometrically calibrated detectors. For the results presented in the following two different side-view intensified visible cameras equipped with narrowband interference filters for dedicated line transitions and a spectrometer, described in detail below, were used. Surface and transmission measurements were carried out in MirrorLab at Forschungszentrum Jülich[4].

### *2.1. Deposition and characterization of pre-deposited samples*

Ablated materials investigated resulted from a-C:D layers deposited by plasma-enhanced vapour deposition with D/C ratio between 0.5 and 0.6 and thicknesses of 0.1–1.1 $\mu$m on polished tungsten. The total amount of material ablated into the TEXTOR edge plasma was concluded from the crater area measured post mortem with a surface profiler (Stil Micromesure 2) multiplied with the atomic density of the samples determined *a posteriori* with both electron probe micro analysis (EPMA) and nuclear reaction analysis (NRA) for carbon, and NRA for the deuterium content. The two methods were in good quantitative agreement concerning carbon for a-C:D layers on tungsten, with NRA typically yielding higher carbon concentrations ($+16 \pm 3\%$). The uncertainty of the amount of removed atoms is determined by layer irregularities, surface roughness after laser exposition and uncertainty of the concentration measurements.

### *2.2. TEXTOR experiment*

TEXTOR experiments were carried out at the plasma–wall interaction facility [9], where samples can be introduced without breaking the vacuum of the main vessel through a limiter lock system. For detection spectroscopic observation systems described in [6] and references therein were used. The experimental conditions for the two experiments described in this work are listed in table 1.

The camera systems were used in conjunction with the respective interference filter for the side view observation of the experiment. The interference filters both have

[4] Mirrorlab website: https://tec.ipp.kfa-juelich.de/mirrorlab/ (access details: mirrorlab@fz-juelich.de).

a transmission maximum at 656.3 nm and cover the emission of hydrogen and deuterium. The transmission curve of the narrowband filter was measured with a spectrophotometer (PerkinElmer Lambda 950 UV/VIS/NIR). From the transmission curves for the wider filter used in experiment A it was found that CII lines within the full width half maximum (FWHM) of the filter at 657.83, 658.25 and 658.35 nm are transmitted with 62, 40 and 37%, respectively. Therefore, the recorded signal through the filter has been corrected with respect to the light pollution by comparing the transmission curve with sufficient high spectroscopic resolution by a Spectrelle spectrometer ($R = 20\,000$ at 656 nm with a FWHM of 3.3 pixel in stroboscopic mode with an exposure time of 5 ms and 5 Hz sampling rate) [10] for LIAS of an a-C:D layer under comparable plasma conditions. The CII emission is caused by the carbon from the a-C:D layer which is ablated at the same time. For the narrowband interference filter used in experiment B the CII line with the strongest contribution to the measured signal has a transmission well below 5% and its influence can be neglected.

For all systems the LIAS signal is obtained by subtracting the average of the previous and subsequent frame from the frame in which the laser is fired and the LIAS light is recorded. Additionally, the emission was monitored in experiment A by a photodiode equipped with a Balmer-$\alpha$ filter with a time resolution of $\sim 1\ \mu$s in order to be capable to resolve the LIAS pulse.

One megawatt neutral beam injector (NBI) heated discharges (hydrogen beam into deuterium plasma) with plasma parameters $I_{\mathrm{p}} = 350$ kA, $B_{\mathrm{t}} = 2.25$ T, $n_{\mathrm{e}} = 3.0 \times 10^{19}\ \mathrm{m}^{-3}$ were used to perform the experiments. Optical drift compensation was used to actively stabilize the plasma position. Current and magnetic fields were reversed during experiment A. The samples were placed about 4 cm behind the last closed flux surface at nominal TEXTOR radius $r = 50.0$ cm on a rotatable sample holder. To ablate the material a Nd:YAG (Innolas SpitLight 2000: $E_{\mathrm{L}} = 2$ J, $\lambda = 1064$ nm, $\tau = 7$ ns) laser system is expanded by a Galilean telescope and guided into TEXTOR via six dielectric mirrors with a beam path length of about 18 m. A convex lens focuses the beam from top onto the sample located the bottom of the torus. The laser energies were measured in front of the TEXTOR window with a laser power meter. A transmission of 85% was assumed to compute the energy fluence reported in table 1 based on the laser spot size. In all experiments LIAS was performed during the current and density flat-top phase of the discharge.

## 3. Results and discussion

LIAS relies on the ability to conclude from observed photons from atomic and ion line radiation to the number of ablated atoms by utilizing the photon efficiency for the observed spectroscopic line. To determine the inverse photon efficiency for $D_\alpha$, a-C:D layers of different thicknesses $d_{\mathrm{i}}$ ($94 \leqslant d_{\mathrm{i}} \leqslant 1108$ nm) were ablated in TEXTOR. The number of laser pulses required to remove the layer depended on the layer thickness, spanning from one laser pulse for the 94 nm layer up to seven pulses for the complete removal of the thickest layer. Five laser pulses were fired during the flat-top phase of the TEXTOR-discharge. The photons observed for each laser pulse with the camera were background subtracted in order to remove the recycling light and then added up for all LIAS frames with significant signal until the complete removal of the layer has been achieved. The integrated amount of photons is shown as a function of ablated atoms which have been determined by NRA analysis and crater measurements depicted in figure 2. The error bars for the deuterium inventory originate from the uncertainties of the NRA atomic area density, the crater volume measurement and the layer homogeneity. The main contribution to the uncertainty in the integral number of photons results from the background subtraction and the fluctuation of the $H_\alpha$-emission from fuel recycling during the plasma flat-top phase. Here, the standard deviation of the emission over all recorded frames before, in between and after the LIAS signal was used to estimate the uncertainty of the background frame as well as the frame with the LIAS signal. For experiment A (blue circles) the correction factor for the CII lines also contributes to the reported error.

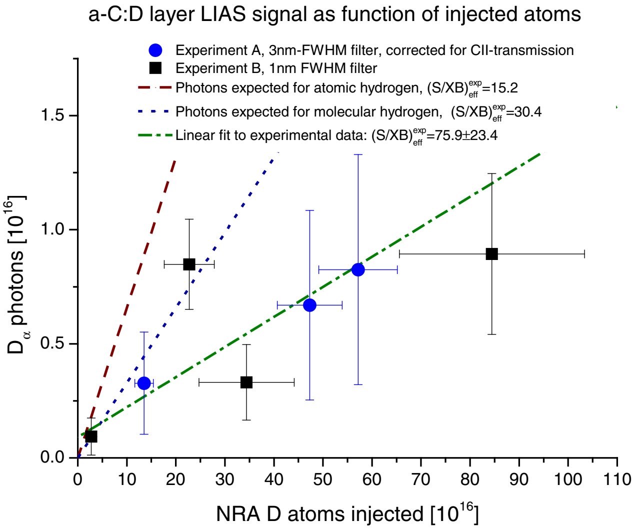

**Figure 2.** Calibration curve for a-C:D layers on tungsten. A linear fit to the measured values of two TEXTOR experiments is indicated.

A green dashed–dotted line indicates a single linear fit to both datasets. The inverse of the slope ($D_\alpha$ photons/D atom) found by the fitting algorithm is the experimentally determined effective inverse photon efficiency for $D_\alpha$ from D atoms resulting from LIAS on a-C:D layers and amounts to

$$\left[\frac{\mathrm{D}}{\mathrm{XB}}\right]_{\mathrm{D}_\alpha(\mathrm{EXP})}^{\text{a-C:D} \underset{\mathrm{LIAS}}{\rightarrow} \mathrm{D}} = 75.9 \pm 23.4. \qquad (1)$$

This can be compared with the $\left[\frac{\mathrm{S}}{\mathrm{XB}}\right]_{\mathrm{D}_\alpha(\mathrm{ADAS})}^{\mathrm{D}}$-value from the ADAS database[5] for atomic deuterium, which corresponds in the limiter plasma edge of TEXTOR of typically $T_{\mathrm{e}} = 50$ eV to an inverse photon efficiency of $\sim 15$ for $D_\alpha$-emission and a purely atomic source. The expected number of $D_\alpha$ photons for an atomic deuterium source of which the strength is determined via NRA is indicated by dashed red line in figure 1, the dotted line in blue indicating the effective $S/XB \sim 30$ value for the thermal

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[5] Open ADAS 1.0, open.adas.ac.uk.

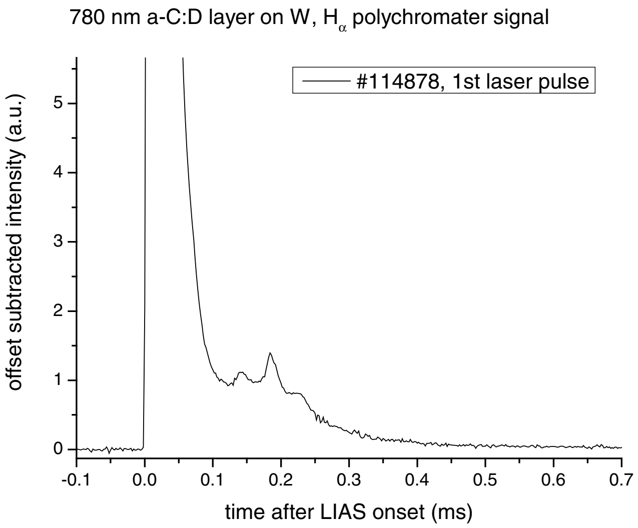

**Figure 3.** Time resolved $H_\alpha$ signal.

$D_2$ source from recycling in TEXTOR [11]. Following the analysis of hydrocarbon injection described in [12] we define the normalized photon-per-particle production efficiency for Balmer-$\alpha$ light as

$$\eta = \left[\frac{\mathrm{S}}{\mathrm{XB}}\right]_{(\mathrm{ADAS})} \Bigg/ \left[\frac{\mathrm{D}}{\mathrm{XB}}\right]_{(\mathrm{EXP})}. \tag{2}$$

For our experimental value $\eta_{\mathrm{D}\alpha}^{\mathrm{LIAS(a\text{-}C:D)}} = 0.20 \pm 0.07$ is found.

While atomic injection of atoms is expected to lead to $\eta = 1$ this factor can be affected by different effects: an increase in the value would be observed for $n_e$ increase as well as $T_e$ decrease due to local plasma perturbation by local cooling. On the other hand, $\eta = 0.2$ was observed for $CH_4$ injection in ohmic discharges in TEXTOR [12]. Here this was thought to be due to the fact that more protons than atoms are produced during the complex catabolism of the methane molecule[6]. To investigate the ablation process the $H_\alpha$- and $D_\alpha$ light (and possibly polluting CII light as discussed above) was recorded temporally resolved with a polychromator-system described in [6]. A time trace for the first laser pulse on a 780 nm a-C:D layer from experiment A is shown in figure 3.

While the temporal profile resembles the shape found for LIAS on EK98 bulk graphite [6] (figure 2), the decay of the peak signal is interrupted by burst-like maxima 0.10 and 0.25 ms after the onset of LIAS emission, which might be attributed to a C cluster release. We note that cluster formation is generally observed in laser ablation [13], but only accounting for $<10\%$ of the ablated material in laser blow-off of metallic targets [14]. No quantitative cluster release has been investigated for a-C:D layers. In ablation of a-C:H layers spallation [15] and the formation of polycyclic aromatic hydrocarbons [16] have been observed. However, from the present data we cannot quantify to what extent the observed photon per particle production efficiency results from the catabolism of hydrocarbons and to what extent it results from the ablation of clusters and fragments.

## 4. Summary and outlook

In this work LIAS was performed on a-C:D layers on tungsten. As LIAS is investigated as a quantitative *in situ* diagnostic for unknown layers, from absolute calibrated spectroscopy and pre-characterized samples exposed in TEXTOR an inverse experimental photon efficiency for $D_\alpha$ from deuterium atoms has been determined from a-C:D layers of different thickness.

The determined experimental inverse photon efficiency for LIAS $[\frac{\mathrm{D}}{\mathrm{XB}}]_{\mathrm{D}_\alpha(\mathrm{EXP})}^{\mathrm{a\text{-}C:D}\xrightarrow{\mathrm{LIAS}}\mathrm{D}} = 75.9 \pm 23.4$ allows for quantitative measurement of the deuterium content in a-C:D layers.

This value is not in agreement with the ADAS S/XB value for an atomic deuterium source and a factor $\sim 2.5$ larger than the value observed for thermal $D_2$ in TEXTOR. The deviation is thought to be due to molecular injection of hydrocarbons and/or fragments created by ablation observed in time resolved measurements.

This will be further addressed in future experiments by measuring radial profiles of hydrocarbon emissions and comparison with reaction kinetic calculations utilizing the HYDKIN (see footnote 6) database.

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